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GaAsSbN/GaAs strained-layer single quantum wells grown on a GaAs substrate by molecular-beam epitaxy with different N concentrations were studied using the photoluminescence (PL) technique in the temperature range from 9 to 296 K. A strong redshift in optical transition energies induced by a small increase in N concentration has been observed in the PL spectra. This effect can be explained by the interaction between a narrow resonant band formed by the N-localized states and the conduction band of the host semiconductor. Excitonic transitions in the quantum wells show a successive red/blue/redshift with increasing temperature in the 2–100 K range. The activation energies of nonradiative channels responsible for a strong thermal quenching are deduced from an Arrhenius plot of the integrated PL intensity. © *2003 American Institute of Physics.*
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I. INTRODUCTION

Several research groups have recently shown that a small nitrogen incorporation into III–V semiconductor alloys induces a strong reduction of the band-gap energy.[1–4] This effect has attracted a great deal of attention to the possibility of using these classes of III–N–V materials to produce lasers emitting within the 1.3–1.55 μm wavelength range, which are strategic for telecommunications.[5,6] Among these materials, $In_xGa_{1-x}N_yAs_{1-y}$ is an attractive alloy which has been used in active layers of lasers with pulsed and continuous-wave emission at 1.31 μm.[6,7] However, it has been difficult to obtain lasers of good quality using InGaNAs alloys at 1.55 μm wavelength emission.[3,8,9]

It has been shown that N-containing samples present a strong photoluminescence (PL) property degradation.[3,9–15] Many works have demonstrated that the disorder in the InGaNAs alloy has a strong effect on the carrier motion, and that the radiative recombinations are generally dominated by localized excitons.[11–15] These works also suggest that the microscopic origin of the localized states is related either to the formation of In–N clusters,[11,13,15] or to the well width fluctuations and/or the local strain field induced by the presence of N.[11,14] Ungaro *et al.*[3] have recently demonstrated that semiconductor alloys from the material—GaAsSbN—grown on a GaAs substrate can be used to prepare optical devices that emit light at room temperature in the 1.3–1.55 μm wavelength range.[3,9] In particular, 100-Å-thick $GaAs_{0.825}Sb_{0.15}N_{0.025}$/GaAs quantum wells (QWs) have demonstrated emission at 1.57 μm.[9] In the present study, we investigated the temperature dependence of excitonic recombination in the GaAsSbN/GaAs QWs in the 9–296 K range. Two QWs with the same width and different N concentrations were analyzed. The thermal activation energies of the nonradiative channels responsible for the thermal quenching were obtained by integrated PL intensity.

II. EXPERIMENTAL DETAILS

Two GaAsSbN/GaAs QW structures with 150 Å width and different N concentrations ($GaAs_{0.843}Sb_{0.15}N_{0.007}$/GaAs-No. 72 637, $GaAs_{0.85}Sb_{0.13}N_{0.02}$/GaAs-No. 72 624) were grown by molecular-beam epitaxy on undoped GaAs (100) substrates. N-containing layers were grown in the 450–480 °C temperature range. Atomic nitrogen was generated from high-purity N_2 gas flowing into a radio-frequency plasma cell manufactured by ADDON. Arsenic and antimony solid sources were used. The QWs were surrounded by $Al_{0.3}Ga_{0.7}As$ cladding layers to produce greater confinement of photogenerated carriers inside the GaAsSbN active layer. The N and Sb compositions in the QW were determined from x-ray diffraction and PL studies. Further details on the growing process and the N incorporation can be found in Refs. 3, 4, and 9. PL measurements were performed in the 9–296 K temperature interval using the 5145 Å line of an Ar^+ laser with a ~20 W/cm^2 power density as the exciting source. The temperature variation was obtained by a closed-cycle helium cryostat. The spectral analysis of the luminescence measurements was carried out by the automated Jobin Yvon T-64000 system and detected by the Ge detector with a standard lock-in technique.

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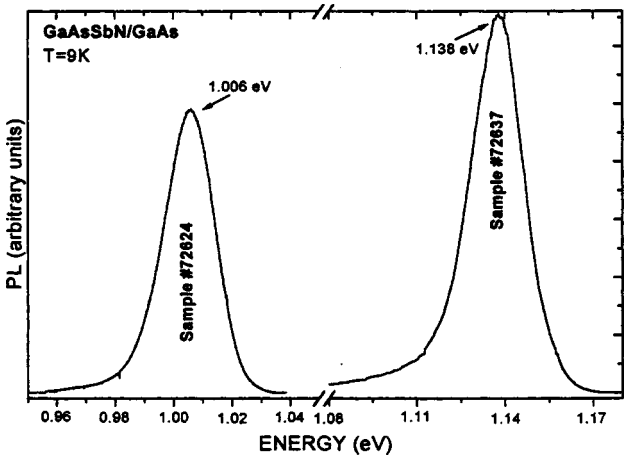

FIG. 1. The PL spectrum for sample No. 72 637 (0.7% of N) and for sample No. 72 624 (2.0% of N) at 9 K. The energy positions of the PL peaks are 1.138 eV and 1.006 eV for sample Nos. 72 637 and 72 624, respectively. The FWHM value is ~20 meV for both samples.

III. RESULTS AND DISCUSSION

Figure 1 shows the PL spectra of the samples 72 637 and 72 624 at 9 K for the same levels of excitation. The peak energies of the excitonic recombinations are centered at 1.138 eV (No. 72 637) and 1.006 eV (No. 72 624), respectively. The full width at half maximum (FWHM) in both samples is practically the same—about 20 meV at $T=9$ K. As Fig. 1 shows, the luminescence efficiency reduces with increasing nitrogen concentration. Similar results have been found in InGaNAs and GaAsN samples.[1,10,12] At low temperature, the PL spectra of both samples have an asymmetric behavior with an exponential tail at the lower-energy side, a characteristic of localized excitonic recombinations due to potential fluctuations.[16–20] Several studies have shown that in QW heterostructures, random fluctuations of the alloy composition and roughness of the barrier–well interfaces are determinant factors of sample quality.[21–24] These defects create fluctuations in the confinement potential and lead to the formation of a band tail in the excitonic density of states.[18,23–25] Depending on the energy excess (obtained from the laser pumping energy) and the magnitude of the potential fluctuation, the excitons will relax either to the local minima or to the absolute minimum of the confinement potential via phonon emission before the radiative recombination takes place.[17,19] Therefore, the PL spectrum reflects the exciton distribution in the energy states generated by potential fluctuations. Figure 2 shows the PL spectra for sample No. 72 637 in the 9–296 K temperature range. The PL peak energy shows a successive red/blue/redshift at low temperatures. This anomalous behavior, also known as the "S-shaped" emission shift, has been observed in both InGaN (Ref. 17) and AlGaN (Ref. 26) bulk materials and in QWs heterostructures such as GaAs/InGaP,[19] CdS/ZnSe,[20] and InGaNAs/GaAs.[11,16] This phenomenon is represented by the dotted line in Fig. 2(a). The thermal energy and the local potential magnitude determine the exciton dynamics. When the temperature increases, the excitons can escape from a relative potential minimum and reach a deeper potential fluctuation. Recombination from deeper levels causes a shift of

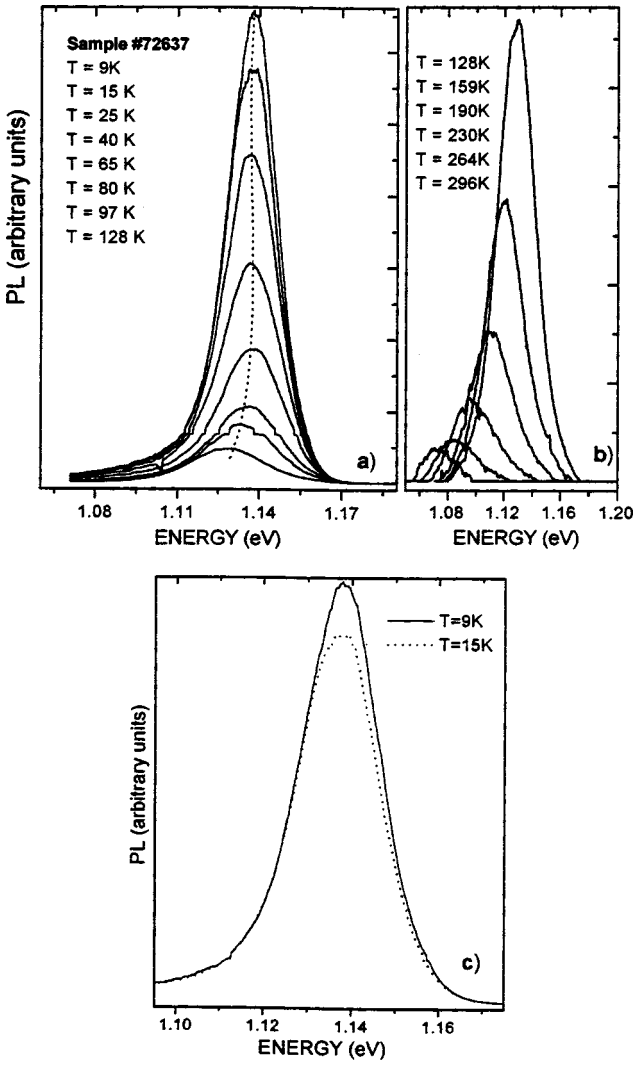

FIG. 2. The PL spectra of sample No. 72 637 at different temperatures. (a) The behavior of the PL spectrum in the 9–128 K temperature range. The dotted line is a guide for the eye of the PL peak variation with temperature. (b) The behavior of the PL spectrum in the 128–296 K temperature range. In this temperature interval, the redshift of the PL peak is originated from the temperature dependence of the recombination of the free excitons. (c) The comparison of PL spectra for sample No. 72 637 at temperatures of 9 K and 15 K. With a slight increase of temperature, the carriers move to states of minimum energy in the band tail. Consequently, there is a decrease in the PL intensity and a redshift of the PL peak energy.

the PL peak energy toward a lower energy.[17–20] This small redshift of the PL spectrum, due to the transfer of exciton population from higher- to lower-energy states of the band tail, can be observed in Fig. 2(c). With an additional increase in the temperature ($T>40$ K), the excitons are thermally transferred to higher-energy states of the band tail, which produces the blueshift of the PL peak energy observed in Fig. 2(a). For $T\geqslant 80$ K, the thermal energy prevents the exciton localization, and the line shape of the PL spectrum becomes a characteristic line shape of the recombination of the free excitons; the further redshift of the PL spectrum in this temperature range arises from the temperature dependence of the band gap. The behavior of the PL spectra originated from the recombination of the free excitons for different temperatures is shown in Fig. 2(b). In the first analysis, the redshift ob-

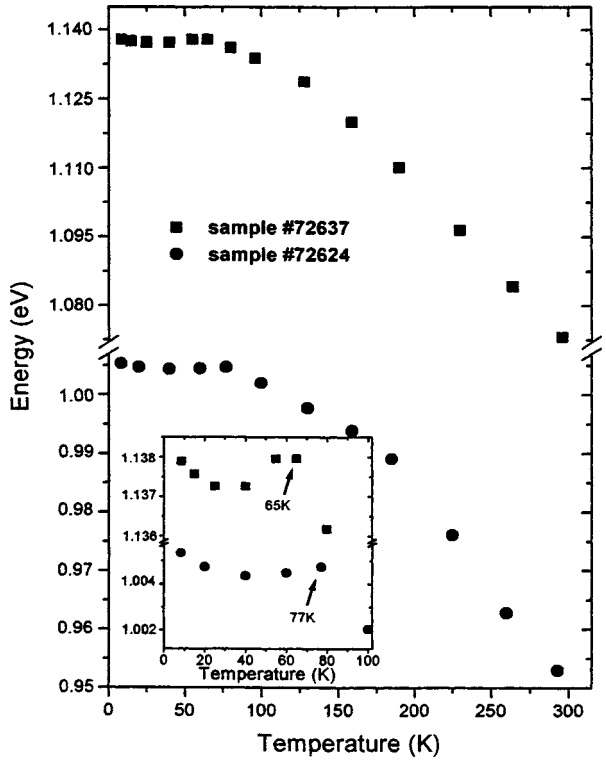

FIG. 3. Temperature dependence of the PL peak energy for sample Nos. 72 637 and 72 624, in the 9–296 K interval. In the inset, we can see the temperature dependence of the PL peak at $T \leqslant 100$ K with a higher resolution. The redshift reaches its maximum value of ~1 meV at 40 K (sample No. 72 637) and 50 K (sample No. 72 624). Over the temperatures of 120 K (No. 72 637) and 150 K (No. 72 624), these samples present a linear temperature dependence of the emission peak.

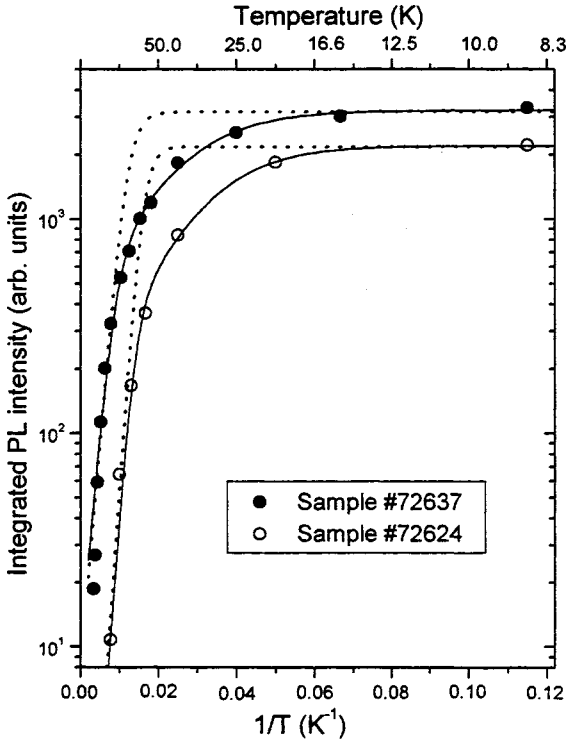

FIG. 4. Arrhenius plot of the integrated PL intensity. Dotted lines (continuous) are fitted using Eq. (1) [Eq. (2)].

served to $T \geqslant 80$ K, is similar to that observed in regular N-free semiconductor materials.[27,28] Figure 3 shows the PL peak energy as a function of temperature in the 9–296 K range. Figure 3 more clearly shows the red/blue/redshift of the PL peak energy in both samples. In a smaller temperature scale, the inset of Fig. 3 shows that the redshift reaches its maximum value of ~1 meV at 40 K for sample No. 72 637 and at 50 K for sample No. 72 624. At temperatures between 40–65 K (No. 72 637) and 50–80 K (No. 72 624), a blueshift is observed to arise from the transfer of thermally activated excitons from lower- to higher-energy band-tail states. In the 40–100 K interval, a competition between the blueshift associated with the recombination of localized excitons and the regular redshift of the band-gap energy induced by the lattice thermal expansion and the temperature dependence of the electron–phonon interaction takes place.[27,28] These latter phenomena dominate the band-gap dependence at temperatures over 120 K (No. 72 637) and 150 K (No. 72 624), as evidenced by a rapid linear redshift of the PL peak energies.[27,28] Polimeni *et al.*[15] observed similar S-shaped PL peak energy behavior in the InGaNAs/GaAs QWs, which was interpreted as arising from the detrapping of strongly localized excitons, and subsequent motion of the excitons in weakly localized states, that lie relatively close to the band edge. The temperature at which this detrapping occurred was found to increase with the N concentration of the QW, indicating that the number and/or potential depth of the carrier trapping centers increase with increasing N concentration.[15] Similar behavior is observed in the detrapping temperature in GaAsSbN/GaAs QWs (see inset of Fig. 3).

The integrated PL intensity as a function of reciprocal temperature is shown in Fig. 4. The integrated PL intensities for both samples remain constant, between 9 and 20 K, and fall by approximately 2.5 orders of magnitude at temperatures over 20 K. The temperature dependence of the integrated intensity of the PL spectra in III–V QW heterostructures with low defects density is usually described by a phenomenological expression:[29–32]

$$I(T) = \frac{I_0}{1 + A \exp(-E_a/kT)}, \tag{1}$$

where I_0 is the intensity at $T = 0$ K, A is the ratio between the exciton radiative lifetime in the QW and the exciton escape time from the QW to a nonradiative center,[29,30,33] and E_a is the thermal activation energy of the nonradiative center. The dotted lines in Fig. 4 represent the best fits obtained by Eq. (1). The thermal activation energies obtained from the fits were: $E_a = 42 \pm 5$ meV for sample No. 72 637 and $E_a = 46 \pm 7$ meV for sample No. 72 624. However, these fits, which consider only one nonradiative channel, differ significantly from the integrated PL intensity in the 18–90 K temperature range. This discrepancy led us to assume that there is a second nonradiative channel. Thus, the integrated PL intensity can be described by the following expression:[17,32–34]

$$I(T) = \frac{I_0}{1 + A \exp(-E_a/kT) + B \exp(-E_b/kT)}, \tag{2}$$

where E_b is the thermal activation energy of the second nonradiative channel. The coefficients A and B measure the efficiency of each quenching mechanisms.[17,32] These preexponential factors are inversely related to the nonradiative lifetime, i.e., to the density of nonradiative centers (for more details see Refs. 32 and 34). The solid lines in Fig. 4 represent the best fits using Eq. (2). The thermal activation energies obtained from the fits were: $E_a = 42 \pm 5$ meV and $E_b = 7.2 \pm 0.5$ meV for sample No. 72 637, and $E_a = 46 \pm 7$ meV and $E_b = 7.6 \pm 0.5$ meV for sample No. 72 624. Although the activation energies are uniquely determinated, using the expression (2), factors A and B are not uniquely determinated and must be interpreted with some caution.[32,34] However, they provide some indication of the relative efficiency of the two mechanisms within a given sample. We observed that the ratio of the number of nonradiative type A centers (high activation energy) to the number of type B centers (low activation energy) increases with increasing N concentration. The low activation energy corresponds to the nonradiative channel that determines the PL intensity quenching at temperatures below 80 K. The observation that both the initial red/blueshift of the PL peak energy and the PL quenching, related to the nonradiative recombination centers of lower activation energy (type B), occurs in the same temperature range (10 K to 80 K) suggests that these two phenomena are correlated. In addition, the 7.2 ± 0.5 meV (No. 72637) and 7.6 ± 0.5 meV (No. 72624) activation energies are consistent with the delocalization energies [$k_B T \cong 5.6$ meV (No. 72637) and $k_B T \cong 6.6$ meV (No. 72624)] measured in the inset of Fig. 3. We believe that the thermal excitation of the carriers contributes to radiative recombinations at different energies of the band-tail states and to the capture of these carriers by nonradiative recombination centers. Thus, we conclude that this first nonradiative channel is easily accessible from the localized states of the band tail, and it is likely to have originated from N-related defects.

When the thermal activation energy is close to the difference between the PL emission energy of the QW and the band-gap energy of the barrier, the thermal quenching of PL intensity at temperatures over 80 K is attributed to the loss of electron–hole pairs from the QW to the barrier.[29,31] However, our results (Fig. 4) cannot be explained by this model. The difference between the PL peak energy of the QW and the band-gap energy of the barrier ($E_g^{\mathrm{GaAs}} = 1.517$ eV at $T = 9$ K) is 379 meV for sample No. 72 637, and 511 meV for sample No. 72 624, which is significantly greater than the measured "type A" activation energies (42 ± 5 meV and 46 ± 7 meV). This result agrees qualitatively with recently published studies; where no correlation was established between the thermal activation energy of nonradiative traps and the band-gap difference of QW and barrier materials.[33,35–37] In this case, two hypotheses are considered to explain the quenching mechanism of the PL intensity for $T > 80$ K: (i) the quenching is related to the escape of only one type of carrier (electrons or holes) from the QW to the barrier[31,33] and (ii) the quenching is related to the presence of defect states in the semiconductor structure.[33,36,37]

In the first hypothesis, the activation energy, E_a, would be given by the band offsets in the conduction or valence band.[31] In order to verify this hypothesis, the energy band alignment at the GaAsSbN/GaAs interface is analyzed. Experimental data and theoretical adjustments have suggested a type-II band alignment for $GaAs_{1-x}Sb_x$/GaAs QW structures and that, in this material, the valence band offset (ΔE_V) is higher than the conduction band offset (ΔE_C).[38–40] Teissier *et al.*[40] verified that the conduction band position of the $GaAs_{1-x}Sb_x$ in the $GaAs_{1-x}Sb_x$/GaAs QW system is situated only ~11 meV over the conduction band position of the GaAs, considering $\Delta E_V = 245$ meV at $x = 0.15$.[40] On the other hand, a simple model has been proposed to describe the effect of the N incorporation in the band structures of III–N–V semiconductor alloys.[2,41] This model, known as the band anticrossing (BAC) model, has been used to explain the strong band-gap reduction in the $In_xGa_{1-x}N_yAs_{1-y}$ alloy.[41] It has also been verified that, in small N concentrations, only the conduction band is altered whereas the valence band position remains practically constant.[2] Considering the results of Teissier *et al.*[40] and assuming that the BAC model could also be applied to the GaAsSbN alloy, the GaAsSbN/GaAs QWs can be classified as type-I QW structures, where the conduction band offset is mainly determined by the N composition. In our samples, a strong confinement for both types of carriers is expected for the Sb and N compositions used, and it is not likely that electrons or holes can be thermally emitted into the adjacent barriers.

Another possible explanation for the high-temperature thermal quenching involves the states associated to defects in the semiconductor material.[33,36,37] The degradation of PL efficiency at low N concentration in III–V semiconductor materials has been reported by several research groups.[12,42,43] Buyanova *et al.*, for example,[43] observed similar nonradiative recombination channels, with activation energy of 50 meV, in the GaAsN/GaAs system. They suggested that these efficient competing nonradiative channels are located in the GaAsN layer, and that these channels are activated as soon as carriers are thermally detrapping from the localized states. Although the origin of these centers is not clearly understood at present, they might have several origins including the presence of extrinsic defects like impurity inclusions related to N incorporation.[44,45] Therefore, the model which holds that the quenching arises from defect or impurity centers induced by the presence of nitrogen, is consistent with the data and it is probably correct.

In conclusion, the temperature dependence of the optical recombination energy in the GaAsSbN/GaAs QWs showed that a small N concentration is responsible for a strong reduction in the optical transition energies in quantum wells, as predicted for III–N–V materials.[2,41] The position of the PL peak energy shows a successive red/blue/redshift at low temperatures, which is a characteristic behavior of the recombination of trapped excitons to localized states induced by fluctuation of N concentration in the alloy. Two efficient nonradiative channels responsible for a strong thermal quenching of PL intensity were identified by the integrated PL intensity. Although the origin of these nonradiative centers is not clearly understood at present, they are probably related to defects in the GaAsSbN induced by the N presence.

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