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Localization effects on recombination dynamics in InAs/InP self-assembled quantum wires emitting at 1.5 μm

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We have studied the temperature dependence of the photoluminescence of a single layer of InAs/InP(001) self-assembled quantum wires emitting at 1.5 μm. The non-radiative mechanisms responsible for the quenching of the emission band have been identified. The exciton dynamics has been investigated using time resolved photoluminescence measurements. The results have been explained through the interplay between free excitons and localized states (arising from size fluctuations in the quantum wires). © 2011 American Institute of Physics. [doi:10.1063/1.3660260]

I. INTRODUCTION

InAs/InP(001) self-assembled quantum wires (QWRs) have received considerable attention in the last years due to their strong luminescence bands at spectral windows relevant for fiber based optical telecommunications.1 The fine control of Stranski-Krastanov epitaxial methods allows tailoring the QWRs height and composition, and thus their emission energy, making possible their integration in novel devices.2,3

Moreover, the QWR area density can be reduced sufficiently to enable the optical investigation of isolated nano-structures.4 One of the most promising semiconductor technologies arises from the combination of the light confinement characteristics of photonic crystals (PhCs) and the light emission capabilities of semiconductor nanostructures. The spontaneous and stimulated emission properties of quantum wells (QWs), quantum wires, and quantum dots (QDs) embedded in PhC microcavities have been reported.5 As a result, the use of PhC technology merged with nanostructures has allowed a great enhancement of the electro-optical properties of semiconductor based devices like larger integration capacity, higher modulation rates, lower threshold laser operation, etc. From the fabrication point of view, embedding nano-structures into a PhC micro-cavity usually requires modifications of the grown process since the epitaxy structure must be adapted to the cavity design. For example, in the case of self-assembled InAs/InP QWRs requires the deposition of a wide film of InGaAs before the InP slab.

In addition to these technological details, the exciton dynamics of the quantum emitters embedded in a photonic device should be precisely known for optimizing the device capabilities. In the case of QWRs, the radiative lifetime of an ideal 1D nano-structure is theoretically proposed to vary as the square root of the temperature.6 Such behavior has been experimentally observed in V-groove GaAs QWRs presenting lifetimes around 300 ps at low temperatures (T < 60 K).7 However, in many other experiments, the exciton recombination takes usually more than one nanosecond and deviates severely from the ideal behavior. These deviations are usually attributed to morphological and compositional fluctuations at the QWR interfaces and thus to confinement disorder and localization effects. In these cases, it can be demonstrated that, the exciton population is not thermalized and the photoluminescence (PL) decay transients are strongly affected by the presence of localized states.8 Meanwhile, at high enough temperatures, the ideal square-root-like behavior can be partly recovered at the same time the weakly localized excitons are thermally promoted to the free exciton state.

In the present work, we study the recombination dynamics of InAs QWRs emitting around λ = 1500 nm and embedded in a λ/2 InP slab. Our goal is to establish which aspects might be relevant for the control of spontaneous and stimulated emission in such systems. Indeed, the slab was grown on top of a 700-nm-thick InGaAs sacrificial layer in order to be used in future photonic crystal devices. On one hand, we show that the emission energy and optical quality of the QWRs have been not affected by the deposition of the sacrificial layer. On the other hand, we determine the radiative lifetime of our QWRs and the role played by exciton localization centers.

II. SAMPLES AND EXPERIMENTAL SETUP

The sample was grown by solid source molecular beam epitaxy (MBE) on InP(001). The InAs nanostructures were formed by the Stranski-Krastanov method depositing 1.7 monolayers (MLs) of InAs at a substrate temperature of 515 °C with a growth rate of 0.1 ML/s. Due to the anisotropic character of the elastic relaxation in this system, QWRs arrays are naturally arranged along the [1-10] crystal direction as explained elsewhere.9 Figure 1 shows an atomic force microscopy (AFM) image of an uncapped sample which exhibits a typical QWR width and height of 11 and 3.2 nm forming quasi-periodic structures (with pitch period around 18 nm). The QWRs are located at the center of a 237-nm-thick InP slab which was deposited onto a 700-nm-thick In53Ga47As sacrificial layer grown on InP (001) substrate.

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The sample was held in the cold finger of a closed-cycle cryostat operating between 12 and 300 K. The PL experiments were carried out exciting the sample just below the InP absorption band edge by means of a 980 nm pulsed laser diode with a repetition rate of 40 MHz and pulse width around 100 ps. The excitation light was focused using a 100 mm focal lens (1 in. diameter) arriving at the sample surface with an incidence angle close to 30° with a spot size of 12 μm. The emitted light was collected by means of a large focal length microscope objective, dispersed by a 0.5 m focal length spectrograph, and detected with an InGaAs Peltier cooled photodiode array in order to obtain the PL spectra. For the PL transient acquisition, the optical signal was dispersed by the same spectrograph but detected by an InGaAs avalanche photodiode. Decay curves were recorded using standard time correlated single photon counting techniques with an overall time resolution around 50-100 ps, depending on the optical intensity and the integration time.

III. RESULTS AND DISCUSSION

Figure 2(a) shows the PL spectrum of our QWRs at 30 and 12 K. The PL is studied at 30 K in order to compare with the results previously published by Alen et al. At this temperature the PL consists of a broad band centered at 1460 nm (0.85 eV). Such emission band can be decomposed into several Gaussian subbands, each one corresponding to the emission of QWRs with different heights. In the present sample, four QWR height families are identified (labeled from P1 to P4) which appear superimposed with the background emission of the InGaAs sacrificial layer. At 12 K (grey shadowed spectrum), the sacrificial layer exhibits an intense emission between 0.77 and 0.79 eV, accordingly with the Indium content of the InGaAs alloy, which rapidly disappears with the temperature increase giving rise to the low energy tail observed at 30 K. The emission energy difference between two consecutive families is consistent with a height change of 1 ML. In fact, we can attribute the emission peaks P1-P4 to QWRs with heights of 8, 9, 10, and 11 ML, as done in previous works on InAs QWRs grown on InP buffers. This observation confirms that the optical and the structural properties of the present sample are not affected by the presence of the InGaAs sacrificial layer below the QWRs. About the integrated optical intensity, all the peaks are not affected by temperature in the same way. Indeed, the behavior of each family during the temperature evolution is related to different loss mechanisms. This fact is illustrated in Fig. 2(b) where the integrated intensities of each component have been represented. While the PL intensity of P2 and P1 stays constant or even increases slightly below 70 K, the PL intensity of families P3 and P4 exhibit a continuous decrease from very low temperatures. The data of P3 and P4 fit with an expression containing two Boltzmann-type quenching mechanisms:

$$I(T) = \frac{I(0)}{1 + \tau_d [G_1 \exp\left(\frac{E_1}{kT}\right) + G_2 \exp\left(\frac{E_2}{kT}\right)]},$$

FIG. 1. (Color online) AFM image of an uncapped sample of InAs/InP self-assembled QWRs.

FIG. 2. (Color online) (a) The PL spectra of the QWRs emission at 12 K (grey solid filling) and 30 K (sparse red line filling). (b) Arrhenius plots corresponding to the families P1-P4, the experimental values obtained for P2, P3, and P4 are fitted to expression (1) and for P1 to expression (2), respectively.
Eq. (1), we can find activation the energies represented in Table I. Below 100 K, the weak decrease of the PL integrated intensity for P3 and P4 is associated to a non-radiative mechanism with activation energy $E_1 = 12$ meV. This carrier loss mechanism could be ascribed to the thermal emission of electrons to impurity centers at the InP barrier or at their mutual interfaces (or other kind of defects close to the QWRs). Above 100–120 K, the optical losses are determined by a second non-radiative mechanism, with activation energies in the range 200-230 meV. These values are well below the energy necessary for the thermion emission of confined excitons, but as previously reported, it would be consistent with a unipolar escape of electrons.

As aforementioned, the temperature evolution of the integrated optical intensity at families P1 and P2 is slightly different. In spite of this fact, the data of family P2 are also fitted to Eq. (1) but just considering the unipolar escape with activation energy $E_2 = 220$ meV. This is explained by a negligible contribution of the lower activation energy mechanism (i.e., $G_1 \approx 0$) as deduced from the near constant variation of the integrated intensity in the Arrhenius plot below 100 K. As a difference with respect to the other components, in family P1 it is required a carrier feeding mechanism to fit the experimental data below 100 K. For this reason, the family P1 is fitted by means of next expression:

$$I(T) = I(0)\left[1 + \frac{A}{1 + \frac{1}{\tau_d \exp(-E_2/kT)}}\right].$$  

(2)

The parameter $A$ accounts for the carrier population ratio being transferred towards the QWRs, whereas $a$ is the ratio between the injection rate of such carriers and the radiative recombination rate of excitons at family P1. The activation energy of this transfer mechanism is represented by $E_2$. The PL quenching mechanism at high temperatures is again very similar to the observed in the other families, $E_2 = 240$ meV, while for the transfer process an activation energy of $E_2 = 31$ meV is obtained. On one hand, this activation energy would not fit with the more common transfer mechanisms, as for example from smaller QWRs. On the other hand, the value of the activation energy is close to the gap between the P1 component and the PL peaks of the InGaAs sacrificial layer. For this reason and having into account that such transfer mechanism has been not observed in samples without sacrificial layer, this gain might be associated to a possible injection of carriers starting from the InGaAs layer through deep impurity levels present in the InP and resonant with the P1 component.

The time resolved experiments are essential to support the above given assumptions and hence for reaching a realistic understanding of the exciton dynamics occurring in our QWRs. As discussed in the following, due to the complex system merging localized and free states, the exciton dynamics of our QWRs will be strongly dependent on the measurement conditions: excitation power, emission energy, and temperature. The effect of the excitation power is typically observed as a reduction of the PL decay time at very high powers. In order to avoid power effects, the lifetime characterization is performed at the low power regime (600 µW of irradiated power at 980 nm wavelength, below the InP band edge). As a result, the time resolved measurements were obtained as mono-exponential transients instead of the more complex decays reported for the high power regime. For instance, in Figure 3(a), a typical PL transient is shown for QWRs of family P1 (0.827 eV) at 12 K. The decay of the optical signal is accompanied by the system response for the laser excitation. As shown in the figure, a mono-exponential decay function (red continuous line) is used to fit the experimental curve after convolution with the system response, in shadow.

The PL decay time dependence on the emission energy is illustrated in Figs. 3(b)–3(c) for two different temperatures. At 12 K, the decay time increases from 1.4 up to 2 ns in the emission energy range from 0.825 to 0.94 eV. Such tendency can be explained by taking into account the influence of electron-wavefunction spill-over effect: in smaller QWRs (higher emission energies), the electron wavefunction is extended out of the QWR (through the InP barrier), while the hole-wavefunction is mainly confined at the QWR, see Ref. 16. As a result, it is produced a noticeable reduction of the electron-hole wavefunction overlap that leads to an increase of the exciton lifetime (as reported for QWs and QDs (Ref. 17)). Since at low temperatures, the localized excitons dominate over the free ones, we can conclude the spillover effect applies for localized electrons too. At higher temperatures, the decays become a bit slower. For instance at 80 K, the lifetimes vary from 1.7 to 2.6 ns, see Figure 3(c). In this figure, we can distinguish two different behaviors: at the low energy region of the 80 K spectrum (coinciding with families P1 and P2), the decay time is still increased with the emission energy. On the other hand, above 0.85 eV (families P3 and P4), the lifetime decreases with the emission energy. Consistently with the Arrhenius plots, this lifetime reduction is associated the non-radiative mechanisms.

In order to present a better description of the exciton dynamics, the lifetime evolution with temperature has been studied from 12 to 200 K, see Fig. 4. The results point out that the different behavior of families P1 and P2 with respect to P3 and P4 is mainly produced by the loss mechanisms previously identified which mainly affect families P3 and P4. But the square root dependence on temperature expected for ideal 1D QWRs is not reproduced by any family. Indeed, the behavior shown by families P1–P3 contains qualitative coincidences with works studying localization effects in nano-structures. Among these works, we have selected the model proposed by Lomascolo et al., since through this model, the lifetime evolution with temperature is described

<table>
<thead>
<tr>
<th>Component</th>
<th>$I_0$ (a.u.)</th>
<th>$\tau_d$ (ns)</th>
<th>$G_1$ (ns$^{-1}$)</th>
<th>$E_1$ (meV)</th>
<th>$G_2$ (ns$^{-1}$)</th>
<th>$E_2$ (meV)</th>
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<tr>
<td>P1</td>
<td>4900</td>
<td>1.56</td>
<td>-</td>
<td>-</td>
<td>$4 \times 10^6$</td>
<td>240</td>
</tr>
<tr>
<td>P2</td>
<td>11750</td>
<td>1.56</td>
<td>-</td>
<td>-</td>
<td>$10^6$</td>
<td>220</td>
</tr>
<tr>
<td>P3</td>
<td>9960</td>
<td>1.64</td>
<td>1.4</td>
<td>11.7</td>
<td>$25 \times 10^5$</td>
<td>231</td>
</tr>
<tr>
<td>P4</td>
<td>1470</td>
<td>1.93</td>
<td>4.4</td>
<td>11.5</td>
<td>$&gt;10^6$</td>
<td>$&gt;200$</td>
</tr>
</tbody>
</table>
by means of parameters representative of our self-assembled QWRs. In fact, fitting the data in Fig. 4 to the next expression we are going to estimate the localization energy and time of localized excitons. The localization energy and the effective linear density of localization sites are represented as a function of the emission energy (red spheres) and the corresponding PL spectrum (black continuous line) recorded at 12 K (b) and 80 K (c).

In this expression, $\tau_{\text{rad}}(T)$ is the radiative recombination time, $\tau_0$ is the intrinsic radiative lifetime, and $\tau_L$ is the lifetime of localized excitons. The localization energy and the effective linear density of localization sites are represented by $E_L$ and $N_D$, respectively. $E_0$ is the energy related to the center-of-mass exciton wavenumber $K_0$. For InP, $K_0 = 1.3 \times 10^7 \text{m}^{-1}$ (considered emission wavelength of 1500 nm and InP refractive index equal to 3.1) and $M$ the total exciton mass (we have used the value given by Andreani et al. for InGaAs QWs, $M = 0.18 m_0$ (Ref. 21)).

Given that $M$ can be slightly different for excitons in InAs QWRs (due to a different valence band mixing), our results should be taken for a realistic, but semi-quantitative discussion. In the case of families, P1 and P2, we observe an increase of the PL decay time by more than a factor 2, as shown in Figs. 4(a)–4(b). Above 140–160 K, the PL decay decreases quickly because of the unipolar escape of electrons to the InP barrier. In contrast, we observe a PL decay time practically constant up to 50–60 K for family P4 and a weak increase up to 110 K for family P3, Figs. 4(c)–4(d). From these results, we can conclude that for families P1 and P2, the PL decay time is mainly radiative. In contrast, to fit the results from families P3 and P4 to Eq. (3) the non-radiative effects must be taken into account:

$$\frac{1}{\tau_d} = \frac{1}{\tau_{\text{rad}}} + \frac{1}{\tau_{\text{nr}}},$$

$$I_{\text{PL}}(T) = I(0) \frac{\tau_d(T)}{\tau_{\text{rad}}(T)},$$

where $\tau_{\text{nr}}$ stands for the non-radiative recombination time associated to the electron loss at low temperatures and $I_{\text{PL}}(T)/I(0)$ accounts for the PL intensity decrease. After calculating the radiative lifetimes, the best fitting parameters to Eq. (3) can be qualitatively compared (Table II). As expected, the values obtained for $\tau_0$ are larger than the values reported for GaAs V-grooved QWRs (see for example, Ref. 22) which is indicative of the different wavefunction for confined excitons. The lifetimes obtained for localized excitons ($\tau_L$) are practically the values of the PL decay times measured at 12 K consistently with their larger contribution at low temperatures. The value of $\tau_L$ increases from 1.5 to 1.9 ns for families from P1 to P4, respectively. As aforementioned, this behavior is attributed to the spill-over effect of the electron wavefunction, demonstrating this way that, it occurs for both free and localized excitons. The exciton localization energies deduced for our QWRs, $E_L \approx 8-16 \text{meV}$ (Table II), suggest that localization effects would be considerably larger than those reported in the GaAs/AlGaAs system. This result can be compared with structural characterization studies: many elongated QWR defects sometimes more than 500 nm long (zones where the height is different than the average) are typically measured by AFM characterization. Along the QWRs, there are also observed width fluctuations extending 100–150 nm in average. By considering the emission energy differences between the different families, we can estimate localization energies in the order of 30 meV for height fluctuations around 1 ML, while we expect localization energies close to 12 meV considering the inhomogeneous broadening produced by the wire width fluctuations. Let notice that the localization is produced if the dimensions of the defect are comparable with the Bohr radius (smaller than 40 nm for bulk InAs), in such a way that the magnitude of
the fluctuations determines the localization energy. In this sense, the elongated height fluctuations observed by AFM can be considered as independent QWRs given the finite extension of the exciton along the wire axis. In contrast, even the largest width fluctuations observed, i.e., 150 nm, could be responsible of certain localization phenomena. The density of localization centers deduced in the present work (listed in Table II) varies from 7 to 11 $\text{nm}^{-1}$ for families P1-P3 (even with an important relative error), which suggests a separation between defects around of 90–140 nm, in agreement with the AFM data. From this result, we can conclude that exciton localization effects can be mainly ascribed to wire width fluctuations.

The optical properties and exciton recombination dynamics in InAs QWRs has been discussed in the present sample bearing in mind its role as active medium in future photonic devices. In this sense, the presence of a sacrificial layer below the QWR layer does not affect the emission properties of the QWRs, as compared to previous published results when they are grown onto a thick InP buffer. Particularly, the observed PL quenching is characterized by a low loss behavior at intermediate temperatures (below 140 K) for families P1 and P2, which is important for laser design. In addition, the determination of the linear density of exciton localization centers suggests a moderate density of defects and the recombination dynamics is dominated by free excitons above 40–50 K. In this way, we could set the optimum temperature for the operation of devices based in QWRs belonging to families P1 and P2 in the range of 50–120 K. Above 120 K, the spontaneous emission rate would be affected by the main carrier loss mechanism of the QWRs, associated to the thermal escape of carriers out of the QWRs. On the other hand, the mentioned optimum operation temperature range would be more difficult to be identified in QWRs of families P3 and P4 due to an additional non-radiative mechanism observed even at low temperatures.

### IV. CONCLUSIONS

In the present work, we have studied the exciton recombination dynamics of self-assembled InAs QWRs embedded into a $\lambda/2$ thick InP layer (the central emission of the QWRs takes place at around 1450 nm at 12 K) deposited onto an InGaAs sacrificial film, a promising epitaxy for photonic crystal based devices. Two electron loss mechanisms have been identified as responsible of the PL quenching at intermediate and high temperatures under both low excitation power and resonant conditions. The TRPL results gives us further information about exciton dynamics, which is determined by excitons localized in QWR size fluctuations at the lowest temperatures. When increasing the lattice temperature the radiative exciton lifetime increases according to the expected thermal equilibrium between free and localized excitons.

### TABLE II. Best fitting parameters of TRPL measurements to Eq. (3).

<table>
<thead>
<tr>
<th>Component</th>
<th>$\tau_1$ (ns)</th>
<th>$\tau_0$ (ns)</th>
<th>$E_L$ (meV)</th>
<th>$N_D$ ($\text{nm}^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>P1</td>
<td>1.48</td>
<td>0.7</td>
<td>16.3</td>
<td>7 ± 5</td>
</tr>
<tr>
<td>P2</td>
<td>1.59</td>
<td>0.7</td>
<td>12.3</td>
<td>11 ± 8</td>
</tr>
<tr>
<td>P3</td>
<td>1.66</td>
<td>0.7</td>
<td>9.3</td>
<td>7 ± 5</td>
</tr>
<tr>
<td>P4</td>
<td>1.85</td>
<td>0.6</td>
<td>8.5</td>
<td>2 ± 5</td>
</tr>
</tbody>
</table>

FIG. 4. (Color online) Temperature dependence of the PL decay time for families P1-P4: (a)-(d). Continuous lines stand for direct fits of the experimental data to the Lomascolo’s Model. In the case of results for PL decay times for families P3-P4, the fit is done on radiative lifetimes. The excitation power in the experiment was kept constant with temperature at 600 $\mu$W of irradiated power.
exciton populations. The localization energies obtained here suggest a major contribution of the wire width fluctuations to the exciton localization dominating the exciton recombination at low temperatures. When increasing temperature, free excitons become important due to the thermal ionization of localized excitons. As a result, some parameters of the excitonic dynamics in InAs/InP QWRs have been obtained, some of them key for the application of this system as active medium in photonic devices working at the telecommunication windows.

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