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Competitive carrier interactions influencing the emission dynamics of GaAsSb-capped InAs quantum dots

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The optical properties of InAs/GaAs quantum dots capped with a GaAsSb quantum well are investigated by means of power-dependent and time-resolved photoluminescence. The structure exhibits the coexistence of a type-I ground state and few type-II excited states, the latter characterized by a simultaneous carrier density shift of the peak position and wavelength-dependent carrier lifetimes. Complex emission dynamics are observed under a high-power excitation regime, with the different states undergoing shifts during specific phases of the measurement. These features are satisfactorily explained in terms of band structure and energy level modifications induced by two competitive carrier interactions inside the structure. © 2012 American Institute of Physics. [http://dx.doi.org/10.1063/1.4769431]

Semiconductor quantum dots (QDs)1 have been extensively investigated in the past decade, and because of their unique physical properties arising from the quantum confinement of carriers in all three spatial dimensions, they are now widely employed for the realization of a number of optoelectronics devices. In particular, high-quality type-I In(Ga)As/GaAs QDs, where both electrons and holes are confined within the dot, have been used as the active material for near-infrared lasers in the 1.3 μm telecommunication window.2,3

Recently, In(Ga)As/GaAs QDs capped with InGaAs or GaAsSb quantum well (QW) have created interest. The larger QW lattice constant compared to GaAs reduces the strain induced in the QDs during the capping process, preserving the shape, and reducing the decomposition of the dots.4–6 Moreover, contrary to other strain-relieving layers, GaAsSb QW with 14% Sb keeps the confining potential of the QDs and consequently leads to optimized emission parameters for the calculations were taken from Ref.13. The optical matrix elements of all possible radiative recombinations involving the first two hole levels and the

where the structure is expected to exhibit not only the most enhanced luminescence characteristics but also unique physical phenomena. The study was performed at low temperature (10 K) by means of power-dependent photoluminescence (PL) and time-resolved PL (TRPL) measurements. PL experiments were performed using a standard setup equipped with a 660 nm CW semiconductor laser, a 0.5 m monochromator, an InGaAs APD detector, and a lock-in amplifier. TRPL experiments were performed employing a 780 nm, 75.6 MHz, 300 fs mode-locked Ti:sapphire laser as the excitation source, and a streak camera as the detection system. The temporal resolution of the experiment was ~20 ps. The band structure of the QDs was calculated by means of an 8-band strain-dependent k·p model (see Ref. 12, and references therein for details). The QDs were simulated as truncated pyramids (16 nm x 16 nm in base size and 4 nm in height) containing 90% In and capped with a GaAsSb QW of appropriate thickness (6 nm) and composition (14% Sb). The material parameters for the calculations were taken from Ref. 13.

Figure 1 shows the calculated band structure. In the conduction band the QD forms a deep confining potential (~0.41 eV) with a small number of confined electron levels. The valence band is more complicated because the energy difference between QD and QW valence band edges is small, less than 30 meV. The two lowest hole levels are shown in the band diagram. The first, double-degenerate, hole level (H_QD) lies in the QD ~40 meV below the QD band edge, while the second hole level (H_QW) is in the QW just ~2 meV below H_QD. H_QD and H_QW wave-functions, depicted in the two insets of Fig. 1, were also calculated. H_QD is mainly confined inside the QD, although the tails of its wave-function penetrate the surrounding QW. For H_QW the situation is reversed, its wave-function is mostly confined in the QW and it slightly penetrates the QD at the sides of the pyramid. The optical matrix elements of all possible radiative recombination channels involving the first two hole levels and the

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without a reversal, the HQD and HQW hole wave-function set. In the case of a slight change in valence band offset, the distribution in dot size and dot/well composition may result in a change or even a reversal of the valence band offset, the inhomogeneous broadening of the real structure. Because of the small size of the valence band offset, the distribution in dot size and dot/well composition may result in a change or even a reversal of the valence band offset. In the case of a slight change in valence band offset without a reversal, the HQD and HQW hole wave-function shapes remain largely unaltered, leading to similar optical matrix elements and transitions as those calculated. In the case of a reversal of the band offset, there will no longer be a dot-like wave-function, and the GS hole wave-function will look similar to HQW. Consequently, it will have similar optical matrix elements, and there will be no significant radiative recombination between it and E0, i.e., these QDs will not contribute to the observed GS emission. Therefore, the effect of the inhomogeneous broadening will not dramatically modify the calculated radiative transition channels.

The character of the observed peaks is also analyzed by using TRPL measurements under a similar power density regime. The decay profiles for the three distinct peaks are shown in the inset A of Fig. 2, while the decay times (colored open symbols) presented in Fig. 2 are calculated as a function of the wavelength by fitting the corresponding decay profiles with a single exponential function. For the GS, the decay times do not significantly change with wavelength, confirming the type-I character of this emission. The average GS carrier lifetime is 1.26 ns, and has a standard deviation of 0.26 ns. The character of the observed peaks is also analyzed by using TRPL measurements under a similar power density regime. The decay profiles for the three distinct peaks are shown in the inset A of Fig. 2, while the decay times (colored open symbols) presented in Fig. 2 are calculated as a function of the wavelength by fitting the corresponding decay profiles with a single exponential function. For the GS, the decay times do not significantly change with wavelength, confirming the type-I character of this emission. The average GS carrier lifetime is 1.26 ns, and has a standard deviation of 0.26 ns.

### Table I.

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<thead>
<tr>
<th>Levels</th>
<th>H₀D</th>
<th>H₀W</th>
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<td>E₀</td>
<td>33.75</td>
<td>0.001</td>
</tr>
<tr>
<td>E₁</td>
<td>0.006</td>
<td>24.02</td>
</tr>
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deviation which is comparable with the temporal resolution of the experiment. This value is consistent with the decay times measured at low temperature for conventional InAs/GaAs QDs. A different behaviour is observed for the two ESs, where the carrier lifetimes measured at the short-wavelength side of each peak are much shorter than those measured at the long-wavelength side. Such strong wavelength-dependent emission dynamics represents, together with the red-shift of the peak position with decreasing excitation powers, another characteristic of type-II band aligned heterostructures. After the photo-generation of e-h pairs inside the structure, the spectrum is blue-shifted and, due to the strong attraction between the electrons in the dot and the holes in the well, the overlap between the carrier wave-functions is at its maximum. The associated emission dynamics (the decay time is inversely proportional to the square of this wave-function overlap) is therefore fast. Following radiative recombination, the carrier density decreases, the spectrum undergoes a red-shift, the wave-function overlap is reduced, and consequently the emission dynamics become slower. In this particular case, the decay times vary from 220 ps to 460 ps for ES1 and from 60 ps to 150 ps for ES2. The high-energy electron levels in the QD efficiently thermalize to the GS. The ESs decay times are therefore much faster, regardless their type-II character, than that of the type-I GS.

Figure 3 shows the emission dynamics for the high-power density regime \( (P = \sim 3 \text{ MW/cm}^2) \) by means of a streak image. The position of the maximum of the intensity for the three states is shown by the black lines. Immediately after the laser pulse \( (t = 0) \), when the carrier density in the structure is the highest, the GS emission peak is red-shifted by \( \sim 11 \text{ meV} \) with respect to its few-carrier position. This shift was not observed in Fig. 2 because of the much lower power density employed in the PL experiment. The relaxation of the emission toward its final wavelength of 1203 nm occurs during the first \( \sim 1.1 \text{ ns} \), after that time the peak position remains constant until the end of the decay \( (t = 4 \text{ ns}) \). Contrary to the GS, the temporal evolution of the ES1 emission peak is not monotonic. It experiences an initial blue-shift for the first \( \sim 0.25 \text{ ns} \), then it reverses direction and a red-shift is observed until the emission is no longer visible after \( \sim 1.5 \text{ ns} \). ES2 behaves similarly to ES1, even though the initial blue-shift is not that clear from the streak image. Additional information on the emission dynamics can be obtained by extracting the decay profiles for the three distinct peaks seen in the streak image. Each decay profile, presented in the inset of Fig. 3, shows common features. After the initial fast capture of the e-h pairs generated by the laser pulse (represented by the steep intensity increase occurring within the temporal resolution of the experiment), the decay profiles exhibit a smooth rise and, only after the maximum of the intensity is reached, an exponential decay. The duration of this rise is \( \sim 0.7 \text{ ns} \) for the GS and \( \sim 0.15 \text{ ns} \) for the ES1.

The various phases exhibited by the emission dynamics can be satisfactorily explained by considering the modifications to the band structure induced by the potentials arising from the interactions between the carriers generated inside the structure under different power density regimes. The two band structures in Figs. 4(b) and 4(c) schematically show these modifications, together with the related change in energy levels of electrons and holes, for low-power and high-power density, respectively. The unperturbed band structure, previously depicted in Fig. 1, is presented again in Fig. 4(a) for comparison. When the material is excited under low-power density, the different densities of states between the QD and the QW mean that, following GS occupation, the holes begin to fill the well states, while the electrons continue to fill the ESs within the dot. Since the repulsion between the confined electrons represents the major contribution to the carrier interactions, the Coulomb potential is centred on the dot. As a result both electron and hole, QD states experience similar shifts toward higher energies, resulting in little change of the overall GS recombination energy. However, the hole states in the QW experience a significant increase in their confinement energy which, together with a smaller band-edge shift, leads to an increase in the separation between the QW hole and QD electron levels. This causes the increase of the ESs emission energy [represented by the orange arrow in Fig. 4(b)], while the GS emission energy [red arrow in Fig. 4(b)] remains approximately constant.

![Figure 3](image3.png)

**FIG. 3.** Streak image [representing the logarithm of the emission intensity in color scale as a function of wavelength (energy) and time] measured at \( T = 10 \text{ K} \) under high-power density regime. The black lines show the temporal evolution of the peak position for the three observed features. The decay profiles corresponding to each separate emission are depicted in the inset.

![Figure 4](image4.png)

**FIG. 4.** Schematic profile of (a) unperturbed band structure, (b) perturbed band structure under low-power density, and (c) perturbed band structure under high-power density. Orange arrows indicate an increase, while brown arrows indicate a decrease of optical transition energy in comparison with the unperturbed case.
Under the high-power density regime, a much higher number of e-h pairs are created inside the structure than the number of available electrons states within the dots. Therefore, after the dots are completely full, since the QW acts as a potential barrier for the carriers in conduction band, excess electrons begin to fill the bulk states in the wetting layer underneath the QDs, while the majority of the holes are still captured by the GaAsSb well. Hence, two charged carrier layers with opposite sign are formed above and below the dots resulting in an internal electric field across the structure. This field, acting similarly to the quantum-confined Stark effect, causes a reduction of the recombination energy for all the states, as depicted by the brown arrows in Fig. 4(c). This is the physical origin of the shift observed in the streak image immediately after the excitation pulse (t = 0). From this time onwards, radiative recombination depletes the carrier population in the structure, the electric field decreases and the emissions shift toward shorter wavelengths. After this initial blue-shift, once the strength of the electric field becomes comparable with the magnitude of the Coulomb interactions, the emission dynamics become governed by the superimposition of the two competitive effects. With elapsing time, due to the progressive reduction of the external electric field, the Coulomb interactions become dominant, and both ESs red-shift according to their type-II behaviour. On the other hand, the GS emission energy, which is not affected by the Coulomb interactions, undergoes only the initial blue-shift during the early stage of the decay.

The complicated shape of the decay profiles in the inset of Fig. 3 cannot be explained simply in terms of QD saturation, where the relaxation of carriers from higher energy states maintains a constant GS population. This cannot replicate the slow intensity rise after the fast carrier capture and the fact that the intensity maximum is reached after a significant time delay. Instead, these features are a consequence of the suppression of the spontaneous emission of the low-energy states due to the occupation of the high-energy ones. The large number of carriers in the high-energy levels in the valence band does not participate in the radiative processes but exert an inhibition on the recombination rate for the low-energy transitions. This also explains why the time duration of the rise is different between the GS and the ES1, with the former clearly showing the most pronounced effect since it is the lowest energy state in the structure. Moreover, the exponential decay for a general state N starts only when the population of the state N + 1 (and all the states energetically above it) is—because of radiative recombination—significantly reduced. This occurs after ~1.85 ns for the GS and after ~0.55 ns for the ES1.

In conclusion, we have studied the optical emission properties of a single layer of InAs QDs capped by a GaAs$_{0.86}$Sb$_{0.14}$ QW. This specific content of Sb was chosen for the type-I GS transition the peak position and the decay time are invariant. Under a high-power density regime, the character of the optical transitions is conserved. However, the structure exhibited complex dynamics, which are explained in terms of band structure and energy level modifications induced by two distinct and competitive potentials created by the carrier interactions. In such structures, due to the type-II character of the ESs, their lasing threshold is expected to be greater than that of conventional type-I QDs. Therefore, the employment of GaAsSb capping layer in InAs/GaAs QD-based lasers should block the lasing from high-energy states, resulting in a stable GS lasing even at high currents.

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