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Lateral interdot coupling among dense ensemble of InAs quantum dots grown on InP substrate observed at cryogenic temperatures

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Abstract. The lateral interdot coupling is investigated in high density (∼10¹¹ cm⁻²) self-assembled InAs quantum dots (QDs) grown on an InP substrate. Two types of structures are selected for this study, in which QDs are embedded into an InAlAs matrix, forming nearly twice stronger confinement for an electron and a hole than expected for an InAlGaAs counterpart. Resonantly injected low carrier population in these families of QDs gives very different spectral and temporal response in the temperature range of 5-30 K. While InAs/InAlGaAs QDs show monotonic temperature quench of photoluminescence (PL), the process seems to be ineffective in the family of InAs/InAlAs dots. Moreover, the PL decay traces for InAs/InAlGaAs QDs reveal a two-exponential decay as compared to a mono-exponential one observed for InAs/InAlAs dots. While a short decay component of ≤1.9 ns has been attributed to recombination of an electron-hole pair confined in the dot, the long one of >2.4 ns, observed exclusively for InAs/InAlGaAs QDs, is attributed to recombination of spatially separated electron-hole pairs formed due to carrier exchange between adjacent dots.

1. Introduction

The presence of lateral coupling among high surface density self-assembled quantum dots (QDs) in the absence of external electric field has been studied for many years, in the view of the appropriate description of the temperature activated carrier transfer process. At high temperatures, such processes can be well described with a carrier activation mechanism enhanced by a large density of phonon states that allows efficient carrier redistribution via higher order QDs states or the wetting layer (WL). Low-temperature processes, however, are still far for being well understood. Negligible phonon population at cryogenic temperatures makes above mentioned carrier transfer mechanism very inefficient. One can imagine that in such circumstances QD confined carriers can still be shared among closely spaced QDs either by: (i) resonant coupling of fundamental states, or (ii) coupling between the fundamental state in one dot to an excited...
state in the adjacent one. In both cases, the resonance condition is constrained to the thermal energy in the system.[3] In the ensemble of self-assembled QDs grown within the Stranski-Krastanow mode, the resonant coupling described in the first case is very unlikely, since local strain conditions prevent the nucleation of two identical dots in the close proximity that is an essential requirement to observe the effect. A very interesting and more probable is to observe the second process. First, more pronounced penetration of a carrier wave function into the barrier for an excited state of a dot increases the chance to be coupled to the adjacent dot. Second, among the inhomogeneous ensemble of dots one can find two non-identical QDs close to each other that can obey the second condition for coupling.

To look for the phenomenon of the lateral interdot carrier coupling at cryogenic temperatures the attention has been focused on InAs QDs grown on an InP(001) substrate, devoted for the laser applications in the third telecom spectral window (1.55 μm). The application target of these dots implicates their high surface density, reaching roughly $10^{11}$ cm$^{-2}$. Since the coupling condition, among many other parameters, depends on the barrier material, two structures are selected where QDs are embedded either in an InAlAs or InGaAlAs barrier. The first barrier provides nearly twice stronger carrier confinement in the dot than the second one. The experimental methodology is based on spectroscopic tools, including photoluminescence (PL), photoluminescence excitation (PLE), photoreflectance (PR), and time-resolved photoluminescence (TRPL).

2. Experimental details and methodology

Investigated QD structures were grown by molecular beam epitaxy on (001)InP substrates. Two structures were taken into account where InAs QDs formed by self assembly are embedded into the In$_{0.52}$Al$_{0.48}$As or In$_{0.53}$Ga$_{0.23}$Al$_{0.24}$As matrix, lattice matched to InP. Slightly different thermodynamics of the QDs growth between two structures results in different dimensions and surface coverage. A typical in-plain size of InAs/InGaAlAs QDs is $\sim 12$ nm$\times\sim 24$ nm and the height is $\sim 3$ nm, whereas InAs/InAlAs QDs are almost twice bigger in the plain, having the size of $\sim 40$ nm$\times\sim 70$ nm and $\sim 3$ nm in height. While the InAs/InGaAlAs QDs have a surface density of $\sim 1\times 10^{11}$ cm$^{-2}$, the InAs/InAlAs QDs have only slightly smaller density of $\sim 7\times 10^{10}$ cm$^{-2}$. Further information about the growth conditions are in Ref.[4, 5].

For all the experiments, the QDs structures where held in an variable temperature He-flow cryostat, operating in the range of 5-300 K, with the accuracy of $\leq 1$ K. For PL, TRPL, and PLE experiments, the structures were excited by a train of 160-fs long pulses, at a repetition frequency of 76 MHz, generated by an optical parametric oscillator with wavelength tunability between 1.1 μm and 1.6 μm. Subsequent PL emission was dispersed by a 0.3-m-focal-length monochromator and detected either by a liquid-nitrogen-cooled NIR-type streak camera or an InGaAs linear array detector. Details of the PR experiment are described in Ref.[6].

3. Results and discussion

Spectral response of investigated QD structures is presented in Fig. 1. The PR experiment revealed a spectral feature at $\sim 1.42$ eV attributed to the interband absorption in the InP substrate. The second PR transition at $\sim 1.12$ eV is observed only for the InAs/InGaAlAs QD structure and related to optical absorption in the In$_{0.53}$Ga$_{0.23}$Al$_{0.24}$As matrix. Respective optical transition for the In$_{0.52}$Al$_{0.48}$As matrix is expected at $\sim 1.56$ eV (not shown). The absorption edge of the wetting layer is observed in the PLE experiments at $\sim 0.98$ eV and $\sim 1.02$ eV for QD structures with a quaternary and ternary barrier, respectively. The QD emission band is centered at $\sim 0.85$ eV for InAs/InGaAlAs QDs, and at $\sim 0.81$ eV for InAs/InAlAs QDs. Spectral broadening of the PL band ($\Delta E$), obtained at the same excitation conditions, is $\sim 28$ meV for QDs with a quaternary barrier, and is twice smaller than $\Delta E \approx 56$ meV for QDs in the ternary matrix. Although $\Delta E$ generally reflects inhomogeneities within the QDs ensemble related to
Figure 1. Photoluminescence, photoluminescence excitation, and photoreflectance spectra measured for InAs/In$_{0.53}$Ga$_{0.23}$Al$_{0.24}$As and InAs/In$_{0.52}$Al$_{0.48}$As QD structures. InP substrate, InGaAlAs barrier, and the wetting layer related optical transitions are indicated by arrows.

Figure 2. Photoluminescence quench registered for InAs/In$_{0.53}$Ga$_{0.23}$Al$_{0.24}$As QDs (black squares) and InAs/In$_{0.52}$Al$_{0.48}$As QDs (red circles) at the resonant excitation condition at 0.918 meV and photoexcitation of less than 1 e-h pair per QD.

Figure 3 shows examples of PL decay traces measured at $T=30$ K for investigated QD structures at the same excitation conditions. One can see that both profiles are qualitatively different. The PL decay for the InAs/InAlAs QD structure can be well approximated by a mono-exponential process, however, the PL decay profile for InAs/InGaAlAs can be viewed as at least bi-exponential, composed of a fast and a slow component. The fast component of $1.4\pm0.2$ ns decay time is attributed to the e-h recombination at the QD ground state, the strain, size and chemical content distribution, however, narrowing of the PL emission may also indicate a selection process due to the presence of interdot lateral carrier transfer.

To evaluate the hypothesis concerning carrier exchange between adjacent QDs and to exclude the WL mobility edge or other 3D states one can carry out the experiment where electron-hole (e-h) pairs are photo-injected directly at QDs states. Fig. 2 shows temperature dependent PL quench measured at low excitation condition ($\ll$1 e-h pair/dot), with the photon energy of $\sim$0.918 eV, settled below the WL mobility edge. At high temperatures, exceeding 30 K, PL quenches registered for both structures behave similarly, with one exclusion seen between 30 K and 60 K for InAs/InAlAs QDs that will not be discussed here. One can focus on the low temperature range of 5-30 K, where thermal excitation of e-h pairs to QD excited states or the WL is less efficient, if not negligible. In this range, the PL quench for InAs/InAlAs QDs is not observed at all, however, it is well visible for QDs in the quaternary matrix. This observation can be related to at least two processes: (i) carrier transfer to optically inactive states either located within a neighboring QD or in its vicinity (tail of the WL density of states), (ii) an electron or a hole within an e-h pair is spatially separated in adjacent dots, owing to quantum tunneling effect, without loosing the Coulomb correlation. Both scenarios are very interesting and have not been widely investigated in the InAs/InP material system. One can try to take a look at the second process. An e-h pair, spatially separated between two adjacent dots, should have smaller oscillator strength of the optical transition, translating into long radiative decay time. Therefore, the PL dynamics should be contributed not only by a common e-h recombination but also by the proces postulated above.

Figure 3 shows examples of PL decay traces measured at $T=30$ K for investigated QD structures at the same excitation conditions. One can see that both profiles are qualitatively different. The PL decay for the InAs/InGaAlAs QD structure can be well approximated by a mono-exponential process, however, the PL decay profile for InAs/InGaAlAs can be viewed as at least bi-exponential, composed of a fast and a slow component. The fast component of $1.4\pm0.2$ ns decay time is attributed to the e-h recombination at the QD ground state, the
Figure 3. (a), (c) Examples of TRPL traces registered at 0.821 eV and T=30 K for InAs/InGaAlAs QDs and InAs/InAlAs QDs, respectively. (b) The value of PL decay time parameter estimated for fast ($\tau_f$ - black squares) and slow ($\tau_s$ - black triangles) component of TRPL traces at various temperatures for InAs/InGaAlAs QDs. Black stars indicate the amplitude ratio between fast and slow component. (d) The value of PL decay time parameter for the InAs/InAlAs QDs. Excitation at 0.918 eV.

equivalent process in the InAs/InAlAs QDs has the time constant of 1.9±0.2 ns.[7] Derivation of the slow component by the fitting procedure is quite difficult, nevertheless, the related time constant exceeds 2.4 ns. The slow component can be related to the recombination process of a spatially indirect e-h pair. In the considered temperature range, all components weakly depend on temperature. However, for the InAs/InGaAlAs QDs, the ratio between amplitudes of a slow and a fast component should increase if the carrier exchange process is temperature driven. Indeed, this behavior is observed in the amplitude ratio, as presented in Fig. 3(b).

4. Conclusions
The issue of lateral interdot coupling in a dense ensemble of InAs QDs embedded into different matrix material has been studied by spectroscopic methods at cryogenic temperatures. A striking difference in the spectral and temporal response between InAs/In$_{0.52}$Ga$_{0.48}$As and InAs/In$_{0.53}$Ga$_{0.23}$Al$_{0.24}$As QDs have been observed and attributed to the presence or absence of an interdot coupling controlled by confinement parameters. Strong temperature driven PL quench in InAs/In$_{0.53}$Ga$_{0.23}$Al$_{0.24}$As QDs and observation of the long-lasting component of the PL decay have suggested the presence of spatially indirect e-h pairs due to coupling between adjacent QDs.

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