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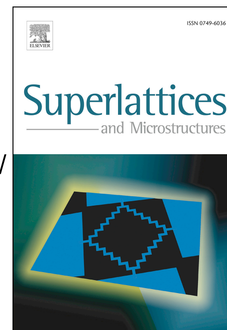

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Probing the carrier transfer processes in a self-assembled system with $In_{0.3}Ga_{0.7}As$/GaAs quantum dots by photoluminescence excitation spectroscopy

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In this report we present experimental studies on the energy transfer between the wetting layer and single large elongated $In_{0.3}Ga_{0.7}As$/GaAs quantum dots. We obtain insight into the electronic and optical properties of $In_{0.3}Ga_{0.7}As$/GaAs quantum dots by probing their confined electronic states via photoluminescence excitation spectroscopy on the single dot level. We demonstrate that the energy separation between the states of a quantum dot and the wetting layer states affects the carrier transfer efficiency - reduced transfer efficiency is observed for smaller dots with higher indium content. We also discuss the effects of the excited states and the trapping of carriers on confinement potential fluctuations of the wetting layer. Eventually, the transfer of charge carriers from localized wetting layer states to a single quantum dot is evidenced in temperature-dependent photoluminescence excitation spectroscopy.

keywords: InGaAs, quantum dot, wetting layer, photoluminescence excitation, energy transfer

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1. Introduction

Self-assembled quantum dots (QDs) have very successfully established their position in modern science and many photonic applications. They provide unique possibilities of confining charge carriers [1], studying their interaction with electromagnetic field in the regime of cavity quantum electrodynamics and non-classical light sources [2], or exploring the coupling between carriers trapped in different dots [3]. QDs have also found a quick way to real-world applications, like telecommunication lasers [4], optical amplifiers [5], solar cells [6] or light-emitting diodes [7]. One particular and important feature of QDs is their in-plane extension - usually the dots are intentionally fabricated to be highly symmetric. Indeed, many realizations of InGaAs/GaAs dots with high In content result in symmetric shape dots. However, ways to fabricate QDs with substantial in-plane asymmetry have been already demonstrated, for instance, by utilizing: (i) a growth-temperature-induced transformation to elongated InGaAs QDs [8,9]; (ii) low-indium-content and low-strain QDs grown at low-growth rate [10]; (iii) different sources of As [11], (iv) overgrowth of InAs QDs [12]; (v) growth on high-index-oriented GaAs substrate [13]; (vi) growth of InGaAs/GaAs quantum dot chains [14,15]; (vii) growth of strongly elongated InAs QDs on InP substrate [16–18]. These elongated nanostructures have shown to differ from their symmetrical kin with emission shifted to longer wavelengths [18], distinct polarization properties [19] or larger oscillator strength due to the increased exciton coherence volume [20]. While the elongation of InAs/InP QDs is naturally achieved during molecular beam epitaxy [16–18], the growth of asymmetric and laterally extended InGaAs/GaAs QDs with large oscillator strength is more challenging [8–10]. The successful realization of lowly strained and hence enlarged InGaAs/GaAs QDs led to the first demonstration of strong coupling between excitons confined in such a quantum dot and discrete modes of electromagnetic field in a three-dimensional microcavity [2]. This has been followed by further studies of this unique type of dots demonstrating, for instance, magnetic field control of the exciton oscillator strength [19,21].

In general, a decrease of QD planar symmetry results usually in strong enhancement of one of the linear polarization directions in surface emission (along the dot elongation) [19,22]. However, surprisingly these enlarged InGaAs/GaAs QDs have shown to be weakly sensitive to the quantum dot asymmetry providing almost no polarization dependence on the nanostructure shape [23]. This is a result of the nonlinear piezoelectric field's anisotropy, which overwhelms the influence of the confinement potential symmetry, making such QDs ideal candidates for applications requiring polarization independence. Similarly, the exciton fine structure splitting, usually increased for asymmetric structures, has also been found to be unexpectedly low (of the order of 5 μeV) [23] which is an important feature for dots considered as sources of entangled photon pairs [24].

Quantum dot properties are defined not only by their shape, composition and strain but are also strongly influenced by the dot's immediate surroundings, especially the wetting layer (WL). It has the form of a thin quantum well formed during the QDs' self-assembly and plays an important role in the energy transfer between confined states of various dots, or QDs and the remaining parts of the structure. Indeed, temperature-driven carrier transfer from elongated InGaAs/GaAs QDs to the wetting layer has been suggested as the main path of carrier loss in these dots, with activation energies in the range of 15 – 30 meV [25]. The inverse process of carrier trapping is even more interesting for it shows the possible channels of feeding the QD states with carriers.

Against this background, the present report is focused on the energy transfer from states of the wetting layer to states of a single large and elongated $In_{0.3}Ga_{0.7}As$/GaAs quantum dot. We present the experimental evidence of such a transfer and we show how the efficiency of this process depends on the energy separation between the specific QD and the WL. Moreover, we discuss excited states and the influence of the carriers' localization in the wetting layer on these transfer processes.

2. Material and methods

The investigated $In_{0.3}Ga_{0.7}As$ quantum dots were grown by solid-source molecular beam epitaxy on undoped GaAs substrate. A QD layer with nominal thickness of 4.5 nm was deposited on a 300-nm-thick GaAs buffer layer, where 2.7 nm of the nominal thickness is wasted to form the wetting layer, and the remaining material constituted self-assembled quantum dots [26]. The indium content of 30% results in low lattice mismatch (2%) between the deposited InGaAs layer and the GaAs matrix, and the QD self-assembly takes place in very low strain conditions. This leads to the surface coverage in the range of 6-9 $\times$ 10^9 dots/cm^2. The $In_{0.3}Ga_{0.7}As$ quantum dots are elongated in $[1\bar{1}0]$ direction and the lateral dimensions are in the range of 50-80 nm and 20-30 nm, whereas the QDs height is of the order of a few nanometers. Further growth details are provided elsewhere [10]. For single dot study the sample was patterned by lithography and etching to form mesas of sub-micrometer sizes to limit the number of QDs probed in the optical experiment.

The energy transfer in the structure was examined by photoluminescence excitation spectroscopy (PLE) on single dot level. This technique has become an important tool for the investigation of energy transfer processes in quantum dot structures [27–30]. Recently, its application was even extended to QDs emitting in the more challenging UV spectral range [31,32]. The experimental setup was equipped with a tunable external-cavity diode laser in Littrow configuration as an excitation source, offering narrow spectral linewidth of the laser lines ($\ll$ 1 μeV). The sample was mounted in a microscopy-type He-flow cryostat for measurements at cryogenic temperatures. Both the excitation and the signal collection were done through a microscope objective (numerical aperture: 0.4), with an excitation and observation spot on the sample surface of about 2 μm in diameter. The emission from the structure was dispersed by a 0.55 m focal-length monochromator and detected with a nitrogen-cooled InGaAs linear

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array. During the PLE measurement the output power of the tunable laser was maintained at a constant level to provide the same excitation conditions for all the laser wavelengths.

3. Results and discussion

Figure 1 shows a PLE map for single $In_{0.3}Ga_{0.7}As/GaAs$ QDs. The resolution of the map is limited by the excitation step of 0.2 nm. The excitation power density was maintained on a moderate level to populate the lowest excitonic states in QDs only. An exemplary photoluminescence (PL) spectrum, for excitation energy marked with the horizontal dashed line, is presented on the top panel with a few single dot emission lines clearly visible. As an example, the intensity of QD5 line (marked by the vertical dashed line), making up a PLE spectrum, is plotted on the right panel. The strong PLE intensity increase is observed for excitation energies close to 1.35 eV, corresponding to the wetting layer ground state energy [25]. The PLE signal enhancement shows directly the presence of the energy transfer from the wetting layer to $In_{0.3}Ga_{0.7}As/GaAs$ quantum dot states at low temperature (5 K). The sharp feature at 1.337 eV, which is equivalent to the difference between excitation and emission energy of 36 meV is consistent with LO phonon energy in the GaAs matrix. It can be also seen as a bright trail in the bottom-right corner of the PLE map, following the excitation laser line. The laser exciting the structure is scattered in an inelastic process, where one LO phonon is emitted (Stokes Raman scattering). It results in an observation of much weaker laser line redshifted by 36 meV from the exciting line energy. As it can be seen in Fig. 1, this scattered line is also shifting when the excitation energy is changed. When it crosses the QD5 emission line it creates apparent intensity enhancement clearly seen on the right panel of Fig. 1 - a result of these two signal intensities overlay. Besides the phonon-related feature, the PLE spectrum shows only very low absorption below the wetting layer edge, indicating that in $In_{0.3}Ga_{0.7}As/GaAs$ QD structures the effect of the continuum of states, resulting from the crossover of

wetting layer 2-dimensional states and 0-dimensional quantum dot states, is rather weak if compared to standard self-assembled InGaAs QDs with higher In content [33].

The comparison of PLE spectra for different $In_{0.3}Ga_{0.7}As$ quantum dot lines QD1 – QD5 is presented in Fig. 2. The main evident difference between the excitation spectra of these dots is the intensity change in the wetting-layer-related part of the spectra. The PLE signal intensity can be described as $I_{\mathrm{PLE}} = I_{\mathrm{ex}}\eta_{\mathrm{abs}}\eta_{\mathrm{rel}}\eta_{\mathrm{em}}$, where I_{ex} is the intensity of the excitation beam and the remaining elements represents probabilities of absorption (η_{abs}), relaxation/transfer to the emitting state (η_{rel}) and emission (η_{em}). The excitation intensity was kept constant for all the investigated QDs. Similarly, the absorption probability, and thus η_{abs}, in a 2.7-nm-thick wetting layer can be assumed as almost invariant in the experiment. It is clearly seen that the PLE signal intensity decreases for quantum dots emitting at lower energies, where the latter usually is attributed to dots of increased size. However, in case of the investigated $In_{0.3}Ga_{0.7}As$/GaAs QDs it has been proven that lower emission energy is associated with smaller dots but with higher In content [23]. The efficiency of the energy transfer from the wetting layer (η_{rel}) can be influenced by the need of dissipating larger energy to feed the In-richer QDs, resulting in the decreased PLE intensity in the WL region. The larger energy difference between WL and In-rich QD ground states engages more acoustic-phonon and carrier-carrier scattering processes, resulting in a less efficient energy transfer to these dots. Additionally, carrier transfer to smaller dots can be deteriorated by the respectively lower carrier capture cross-section. However, the emission probability (η_{em}) can also differ between QDs due to the oscillator strength deviations, because the radiative lifetime of asymmetric $In_{0.3}Ga_{0.7}As$/GaAs QDs depends on the emission energy [34]. For In-richer quantum dots the radiative lifetime is increased, indicating lower oscillator strength in this confinement regime, in accordance with smaller emission probability. Therefore, the observed decay of the PLE signal in the

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WL absorption region can result from the less efficient carrier transfer to the lower energy (and smaller) dots and lower oscillator strength values of the In-richer QDs.

Some of the investigated $In_{0.3}Ga_{0.7}As/GaAs$ QDs revealed more complex PLE spectra. Figure 3 shows a PLE map at even lower excitation power density (by one order of magnitude) with two QD-related lines being visible (QD_A: E = 1.3008 eV, QD_B: E = 1.3023 eV). These QD emission lines show additional intensity enhancements below the wetting layer ground state energy (the intensity increase in the bottom right corner of the map arises from the background of the used external cavity laser and it should be ignored as an experimental artifact). The PLE spectra for these two QD emission lines taken at 5 K are presented in Fig. 4. The QD_A line shows two PLE enhancement peaks in the vicinity of the wetting layer ground state energy (1.346 eV and 1.348 eV), whereas the QD_B exhibits two enhancements at 1.334 eV and 1.341 eV, i.e. clearly below the WL energy. The difference between the excitation and emission energy values for these two lines from QD_B is 33 meV and 38 meV, respectively. To explain the origin of these two features laying below the WL ground state energy, an absorption spectrum of a single $In_{0.3}Ga_{0.7}As/GaAs$ QD has been simulated and is presented in Fig. 5. The calculation was based on a realistic lens-shaped dot geometry (25 nm width, 80 nm length and 3 nm height) placed on a 2.7-nm-thick wetting layer (and the nominal of 30% indium) and was performed within the 8-band $\mathbf{k}\cdot\mathbf{p}$ model [35]. Strain and corresponding piezoelectric field have been also included in these calculations. To take into account the experimental conditions, the line broadening (0.5 meV) has been added, which simulates the combined effect of the spectral resolution and spectral diffusion. Figure 5 shows all the states confined within the simulated QD. In the energy range of 33-38 meV above the dot ground state there are two apparent maxima which correspond to the highest states confined within the simulated $In_{0.3}Ga_{0.7}As/GaAs$ quantum dot. Thus, the peaks observed in the PLE spectrum of line QD_B can be related to the QD excited states. Moreover, states confined within the similar energy

range can be found in dots with different geometries and In compositions. However, depending on the details not every single QD presents similarly rich spectrum of confined states, therefore, such excited-state-related features in the PLE spectra could only be seen for a few of the investigated dots . The detailed theoretical analysis of the states confined within a single $In_{0.3}Ga_{0.7}As/GaAs$ quantum dot together with the influence of the dot geometry on its energetic structure can be found elsewhere [36].

The maxima of the QD_A line in PLE spectra of Fig. 4 do not match the energies of expected QD excited states nor optical phonon energy values, but they fall into the range of quasi-zero-dimensional structures formed of the fluctuations in the wetting layer (possible thickness or content fluctuations driven by the indium segregation and clustering effects). Structures with asymmetric $In_{0.3}Ga_{0.7}As/GaAs$ quantum dots have already shown fingerprints of such carrier localization within the WL. As shown in a previous report, the increase of temperature from 5 K to 30 K resulted in the decrease in intensity of single emission lines in the WL spectral region, which corresponds to the exciton localization energy of the order of 2-3 meV [20]. To see if the features in Fig. 4 can be linked to the WL-localized states, the PLE spectra were measured at even higher temperature of 40 K, corresponding to the thermal energy of 3.3 meV (also shown in Fig. 4.) – in the range where delocalization of carriers/excitons from these shallow traps is expected. Indeed, the peaks close to the wetting layer spectral region in the PLE spectrum of the QD_A line are diminished at 40 K, whereas the excited-states-related features in the PLE spectrum of the QD_B line are almost preserved at this higher temperature (only redshifted by approx. 1 meV due to the temperature-induced energy bandgap change).

With the increased temperature there is also a second change evident in both the PLE spectra – the WL spectral region increases in the intensity. As it is expected, the temperature-induced delocalization makes supplying the QD with carriers easier [37]. The carrier delocalization is followed by their redistribution within the wetting layer and dot emission fed by the carriers from WL states becomes

enhanced. On the other hand, the transfer from WL-localized states to QD states takes place between the states of zero-dimensional character and separated spatially, thus the probability of this process is rather low and it was observed for some of the investigated QDs only, which is in agreement with a spatially inhomogeneous distribution of WL potential fluctuations. The transfer will be possible only when the dot is positioned in the vicinity of the WL potential fluctuation.

4. Conclusions

By performing single dot photoluminescence excitation spectroscopy we demonstrated the energy transfer from the wetting layer to quantum-dot-states for lowly strained $In_{0.3}Ga_{0.7}As/GaAs$ self-assembled QDs with atypical morphology. The efficiency of this process was shown to depend on the quantum dot size and In content and was found to be lower for small dots with high indium content. This behavior can be explained by a combination of several factors: lower carrier capture cross-section of smaller dots, their lower oscillator strength values and larger energy dissipation while feeding these dots. We also discussed the appearance of the resonance features in PLE related to the QD excited states together with a fingerprint of the energy transfer to the quantum dot from the 0D-like states for carriers localized on wetting layer potential fluctuations.

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Figure captions

FIG. 1. PLE map (linear scale) of single $In_{0.3}Ga_{0.7}As/GaAs$ quantum dots at low temperature (5 K). The top panel shows the PL spectrum for the excitation energy marked with the horizontal dashed line. The panel on the right side presents a PLE spectrum for the detection energy marked with the vertical dashed line showing a broad wetting-layer-related absorption band.

FIG. 2. Comparison of single $In_{0.3}Ga_{0.7}As/GaAs$ quantum dot PLE spectra for different QD lines QD1 – QD5 at low temperature (5 K).

FIG. 3. PLE map (linear scale) of single $In_{0.3}Ga_{0.7}As/GaAs$ quantum dots at low temperature (5 K). The top panel shows the PL spectrum for the highest excitation energy (1.355 eV).

FIG. 4. PLE spectra of single $In_{0.3}Ga_{0.7}As/GaAs$ quantum dots for two different temperatures (5 K and 40 K).

FIG. 5. Simulated absorption spectrum of a single $In_{0.3}Ga_{0.7}As/GaAs$ quantum dot of 25 nm width, 80 nm length and with a height of 3 nm.

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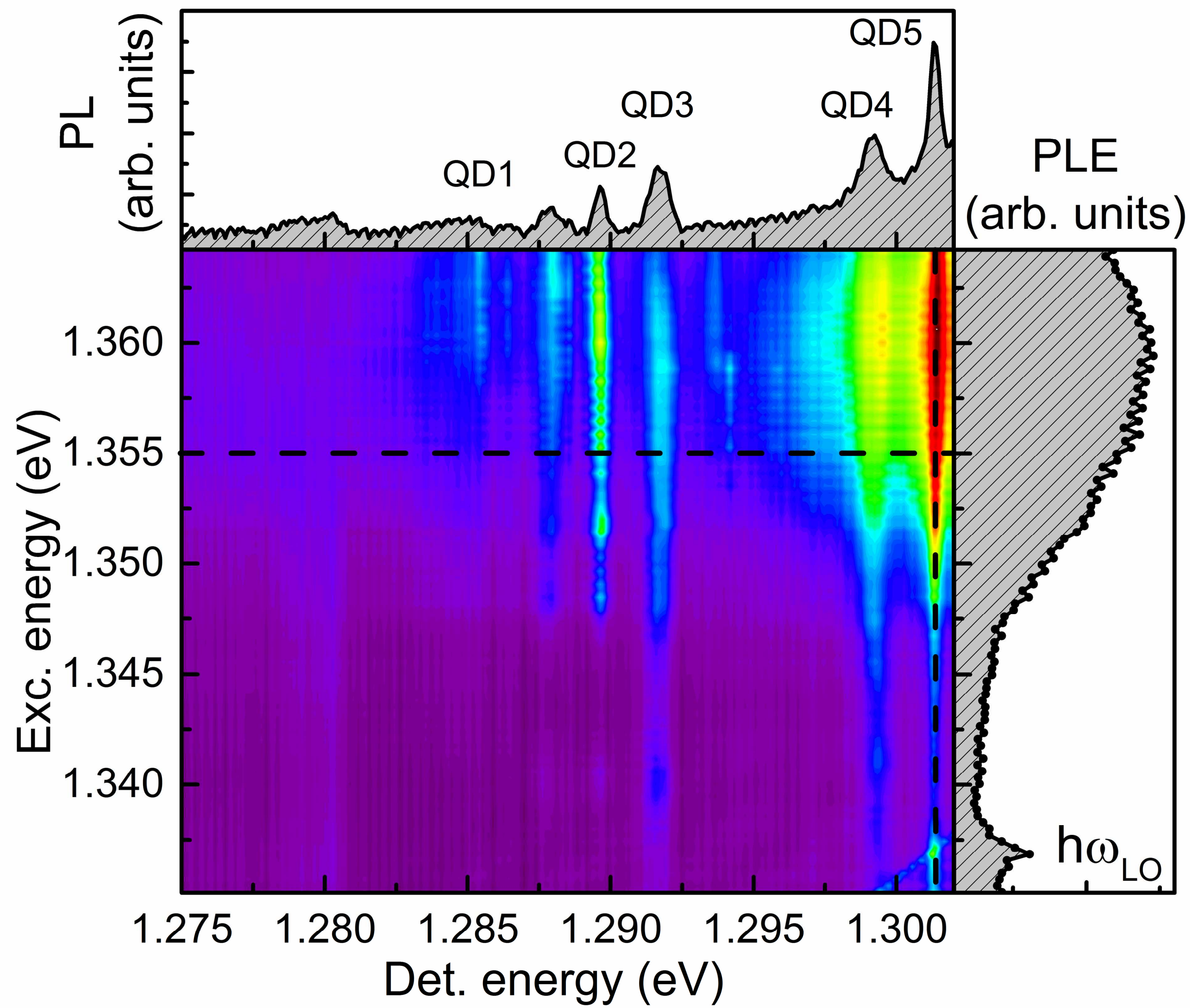

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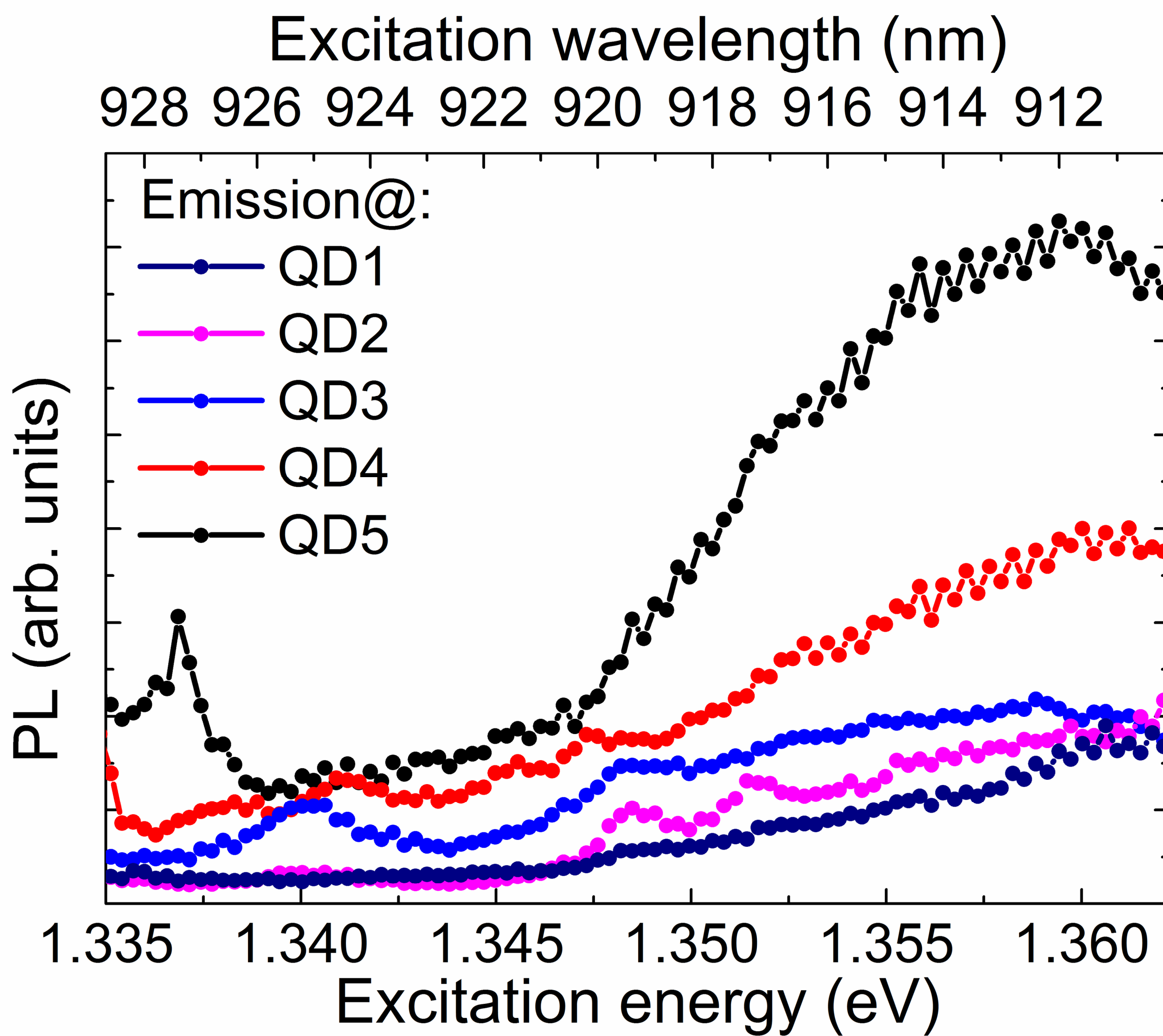

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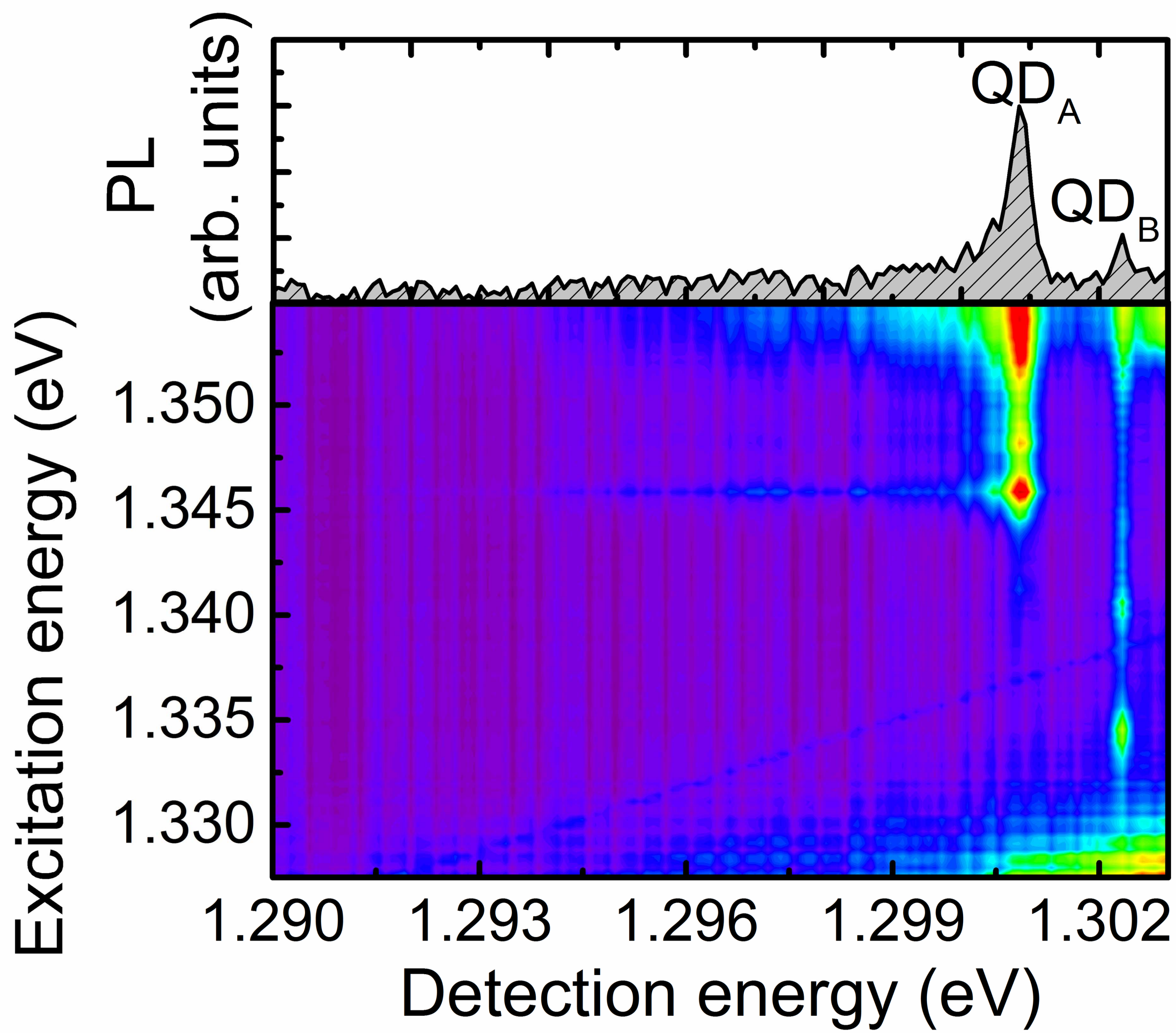

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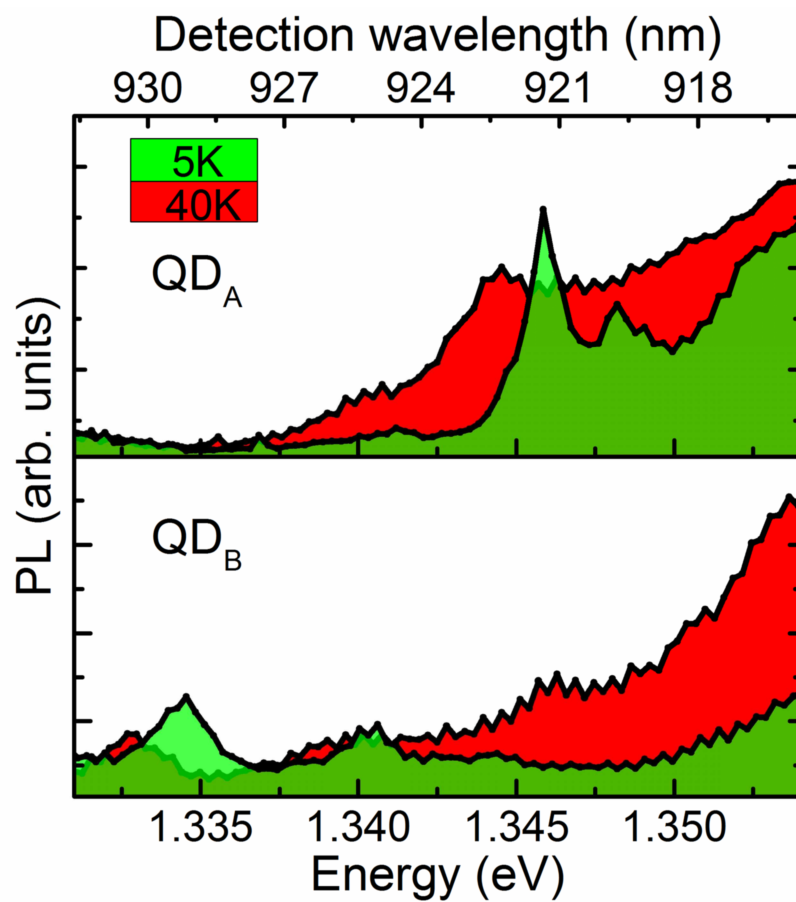

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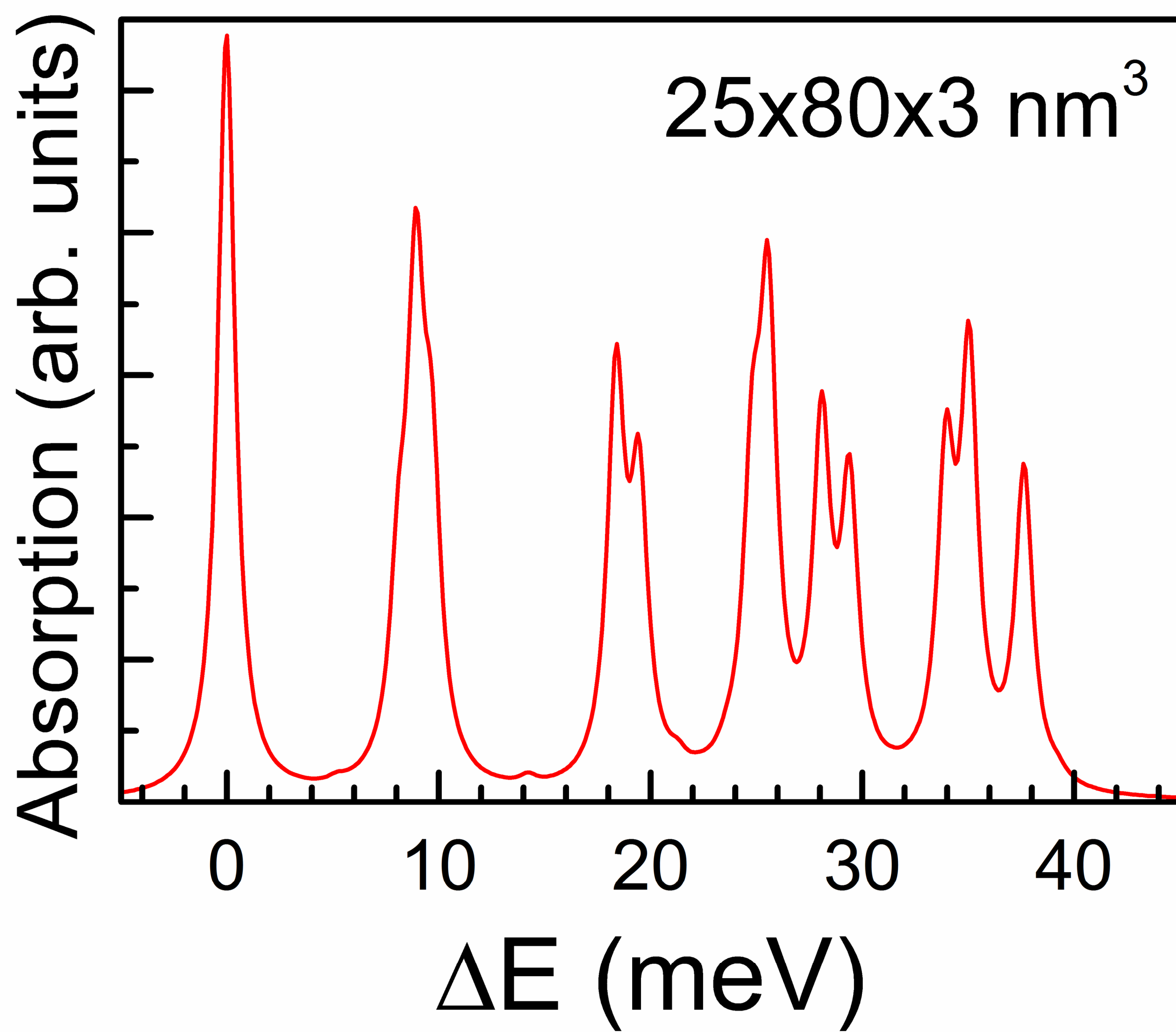

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- Transfer of carriers in a self-assembled system with lowly-strained, large and elongated $In_{0.3}Ga_{0.7}As$/GaAs dots has been probed by photoluminescence excitation spectroscopy
- Energy separation between $In_{0.3}Ga_{0.7}As$/GaAs quantum dot states and the wetting layer states affects the transfer efficiency - reduced transfer efficiency is observed for smaller dots with higher indium content
- Direct transfer of charge carriers from localized wetting layer states to a single quantum dot has been observed for the first time

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