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Excitation transfer in self-organized asymmetric quantum dot pairs

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Excitation transfer processes within self-organized quantum dot (QD) pairs in bilayer InAs/GaAs QD samples are investigated. QDs in samples with a 1.74-ML InAs seed layer and a 2.00 ML InAs second layer are found to self-organize in pairs of unequal sized QDs with clearly discernible ground-state transition energy. Photoluminescence (PL) and PL excitation results for such asymmetric QD pairs provide evidence for non-resonant energy transfer from the smaller QDs in the seed layer to the larger QDs in the second layer. Variations in the optical behavior as a function of the spacer thickness and composition are attributed to the barrier-dependent tunnel probability. Tunneling times down to 20 ps (36 ML GaAs spacer) are estimated, depending exponentially on the GaAs spacer thickness. [S0163-1829(98)52640-9]

The formation of nanoscale coherent islands in highly strained semiconductor epitaxy has been extensively studied as a means to generate optically active quantum dots (QDs).[1] One important aspect of such Stranski-Krastanow QDs is strain-driven vertical self-organization[2] of island stacks in multilayered samples, which allows for an increased density of high-quality QDs and opens new design possibilities in coupled quantum structures. Nevertheless, no systematic investigation of excitation transfer processes in such close-packed QD systems has been reported yet.

First indications for strong electronic coupling as well as excitation transfer processes of stacked InAs/GaAs QDs have been reported for samples with constant deposition amount for each layer.[3–6] A systematic lowering of the QD ground-state transition energy observed in photoluminescence (PL) spectra of samples with increasing number of InAs layers as well as decreasing spacer thickness ($\lesssim$20 ML) has been taken as an indication for the formation of coupled QD states.[3,4] Although stacked QDs are found to behave like uncoupled ones for thicker spacers ($\gtrsim$36 ML), excitation transfer processes have been proposed to explain excitation density dependent PL spectra.[6]

In this paper, we present structural and optical results for bilayer samples with differing InAs deposition amounts in the first and the second layer. This variable deposition amount approach leads to vertically stacked pairs of unequal sized QDs in which the density of the QDs in the upper layer can be controlled by that of the first layer whereas their average size can be made larger by a higher deposition amount.[7] The ground-state transition energies of the two can be clearly distinguished in the PL spectra. Based on the electronic structure of the constituents we name such a system an ‘‘asymmetric QD pair’’ (AQDP) in analogy with the extensive work on *asymmetric* double quantum-well (QW) systems.[8–11] The PL and PL excitation (PLE) results for InAs/GaAs AQDPs in such bilayer samples demonstrate energy transfer processes and allow estimation of the nonresonant carrier tunneling time.

The investigated samples with a 1.74-ML InAs seed layer and a 2.00-ML InAs second layer were grown by molecular-beam epitaxy on semi-insulating GaAs(001) substrates. InAs was deposited at 500 °C, an As_4 partial pressure of 6×10^{-6} Torr, and a growth rate of 0.22 ML s^{-1} as described in Refs. 2 and 12. For a first set of samples 36-, 45-, or 54-ML-thick GaAs spacers were grown at 400 °C using migration-enhanced epitaxy (MEE).[12] For a second set of samples, part of the GaAs spacer was replaced with $Al_{0.21}Ga_{0.79}As$ which, being almost lattice matched to GaAs, preserves the surface strain distribution. After 22-ML MEE GaAs spacer growth, the temperature was increased to 610 °C for the deposition of 7-, 16-, or 25-ML $Al_{0.21}Ga_{0.79}As$, a 90-s growth interruption, the deposition of 7-ML GaAs, and a second 90-s growth interruption, before the temperature was decreased to 500 °C for the growth of the second InAs layer. Finally, a 170-ML MEE GaAs cap was grown at 400 °C. The samples were structurally characterized using cross-sectional transmission electron microscopy (XTEM). PL was studied at 6.2 K in a continuous-flow He cryostat using an Ar^+ laser for GaAs excitation, a Ti-Sapphire laser for wetting layer (WL) excitation and PLE spectra, and a cooled Ge diode in conjunction with a 0.85-m double grating monochromator for detection.

The formation of AQDPs in bilayer InAs/GaAs samples with different InAs deposition amounts in the seed and second layer is illustrated schematically in Fig. 1(a).[7] For the 1.74/2.00 ML deposition combination the seed and second layers provide, respectively, the smaller (SQD) and larger (LQD) QDs. Figures 1(b) and 1(c) depict XTEM images of samples with 36-ML spacer without and with [as indicated in Fig. 1(a)] a 7-ML $Al_{0.21}Ga_{0.79}As$ insertion, respectively. The investigated samples show a high degree of vertical island correlation ($\geqslant$95%) and the island density, controlled by the seed layer,[7] is consistent with that typical for a single layer 1.74-ML InAs sample.[13] The lower $GaAs/Al_{0.21}Ga_{0.79}As$ interface appears flat in Fig. 1(c) in agreement with earlier results,[12] showing that for the employed growth conditions a 22-ML MEE-grown GaAs spacer completely buries the InAs islands in the seed layer and planarizes the growth front. Finally, the PL results (Fig. 2) indicate that the subsequent growth of the remaining spacer at 610 °C does not significantly affect the islands in the seed layer.

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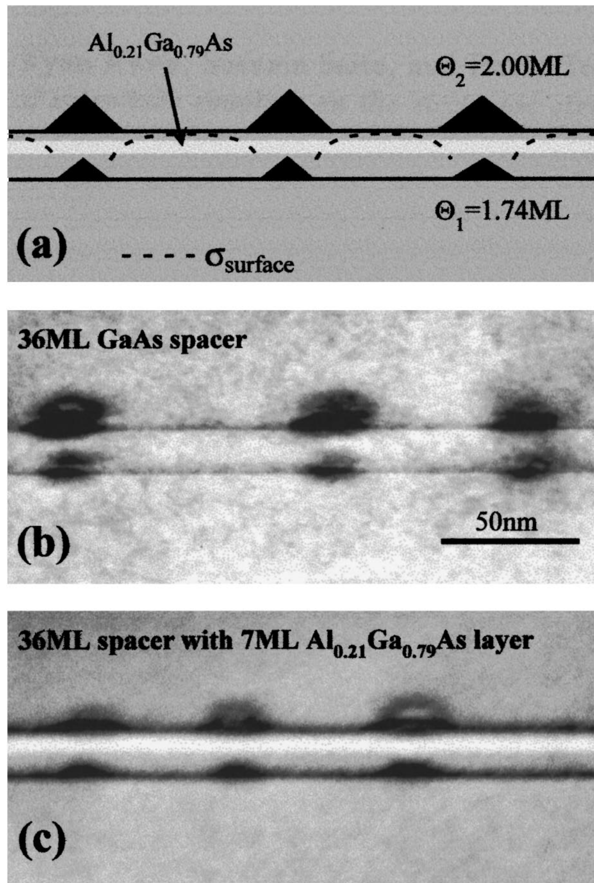

FIG. 1. (a) Schematic of AQDPs formed in bilayer InAs/GaAs vertically stacked QD samples with variable deposition amount (Ref. 7). (b) and (c) show cross-sectional $g=(200)$ dark-field TEM images obtained for 1.74/2.00 ML bilayer samples with 36-ML spacer without and with a 7-ML $Al_{0.21}Ga_{0.79}As$ layer, respectively.

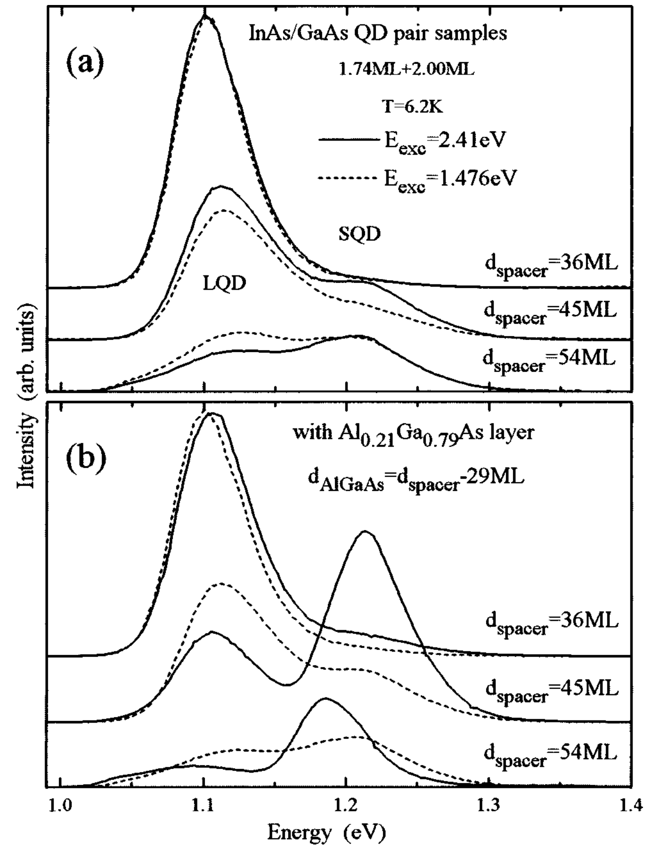

FIG. 2. Low-temperature PL spectra for 1.74/2.00 ML bilayer samples with various spacer thicknesses without (a) and with (b) $Al_{0.21}Ga_{0.79}As$ layer. PL was excited in the GaAs barrier at 2.41 eV (full lines) or the WL at 1.476 eV (dashed lines).

Figure 2(a) compares PL spectra of 1.74/2.00 ML InAs deposition AQDP samples with 36-, 45-, and 54-ML GaAs spacers. The PL spectrum of the 36 ML sample shows a single, almost Gaussian, peak at 1.102 eV having a full width at half maximum of 58 meV. The peak energy is ~100 meV lower than that of QDs in single layer samples[12,14] with 1.74- or 2.00 ML InAs deposition grown at the same conditions, suggesting the luminescence to originate from the LQDs in the second layer. With increasing spacer thickness an additional PL peak becomes evident on the high-energy side at ~1.21 eV, resulting in a clear double peak structure for the 54 ML spacer sample. The high-energy peak coincides with the QD PL peak observed for single layer samples with 1.74 ML InAs deposition.[12] Figure 2(a) shows the transition energy of LQDs to increase (~15 meV) and that of SQDs to decrease (~10 meV) with increasing spacer thickness. Although, this might be a consequence of the decreasing electronic and/or strain coupling in the AQDPs, we cannot exclude changes of the average island sizes in the samples due to statistical variations of the QD density in the first layer,[7] which might vary by about $\pm 50\ \mu m^{-2}$.[13]

The apparent increase in the PL yield of the SQDs with increasing spacer thickness is attributed to the decreasing electronic coupling and, consequently, decreasing excitation transfer probability in the AQDPs. Replacing part of the GaAs spacer with the wider band gap $Al_{0.21}Ga_{0.79}As$ further increases the tunnel barrier and favors PL from SQDs as seen in Fig. 2(b). Additionally, a comparison of PL spectra excited in the GaAs barrier (solid lines) and the WL (dashed lines)[15] shows GaAs barrier excitation to favor SQDs. Since the WL properties are not affected in the stacking process,[6] we assume for the WL excitation case the excitation rates for SQDs and LQDs to be the same. For GaAs barrier excitation case electron-hole pairs are predominantly generated on the substrate side of the InAs layers and, thus, favor excitation of SQDs in the seed layer. This effect becomes more pronounced for samples with an $Al_{0.21}Ga_{0.79}As$ layer, causing carriers generated on the substrate (cap) side to excite SQDs (LQDs). From the cap thickness and the exciton diffusion length in GaAs we estimate a ratio of 5:1 for the excitation rates of SQDs and LQDs in such samples.

Figure 3 compares PLE spectra in the WL region detected on the ground-state PL of SQDs and LQDs for some of the investigated samples. For samples with pure GaAs spacer the WL resonance of the first and second InAs layers are indistinguishable at 1.455 eV, a value typical for single layer samples.[14] The intensity of the WL resonance in the PLE spectra of SQDs decreases with decreasing spacer thickness due to increasing excitation transfer to LQDs. Samples with $Al_{0.21}Ga_{0.79}As$ layer (denoted here as WL^{Al}) show an additional resonance shifted by ~15 meV towards higher energies. The WL^{Al} resonance is attributed to the second-layer WL that is supposedly affected by the $Al_{0.21}Ga_{0.79}As$ layer

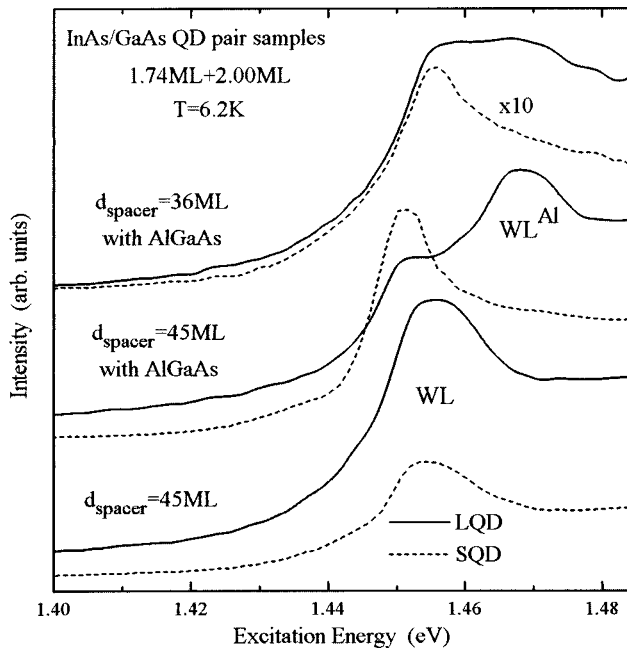

FIG. 3. Low-temperature PLE spectra detected on the maximum of the SQD and LQD PL peaks showing the WL resonances.

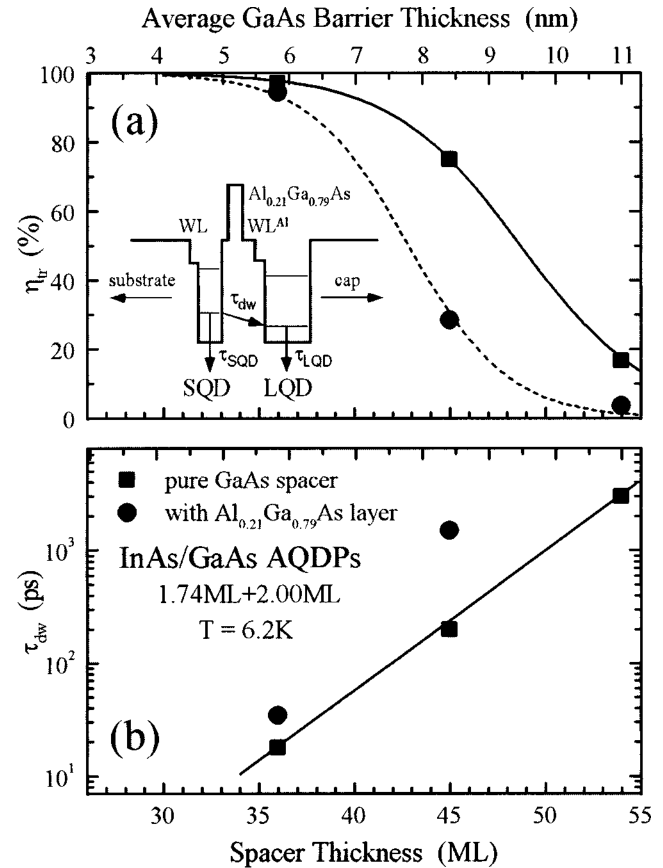

FIG. 4. Excitation transfer yield η_{tr} (a) and dwell time τ_{dw} (b) for AQDPs as a function of the spacer thickness. The average tunnel barrier thickness is derived assuming an average SQD height of 4.5 nm (see the text). The inset shows a schematic conduction-band energy diagram for AQDPs with an $Al_{0.21}Ga_{0.79}As$ layer.

being separated by 7 ML of GaAs from the WL. The different energy of the WL resonance in the first and second layer provides a means to distinguish the corresponding excitation processes. As shown in Fig. 3, the WL of the seed InAs layer (~1.455 eV) is observed for both SQD and LQD excitation, but its intensity in the spectra of LQDs decreases with increasing spacer thickness. In contrast, the WL^{Al} resonance appears only in the PLE spectra of LQDs, indicating unidirectional excitation transfer from SQDs to LQDs. Obviously, the carrier dynamics in the WLs is dominated by intralayer carrier capture into localized QD states, being faster than 5 ps.[16]

The inset of Fig. 4 gives a schematic conduction-band energy diagram for the 1.74/2.00 ML AQDP structures with $Al_{0.21}Ga_{0.79}As$ layer. In the following we assume intralayer and intradot carrier relaxation to be faster than interlayer excitation transfer. Thus, excitation transfer originates in the ground state of SQDs with the dwell time denoted τ_{dw}. The ground state lifetimes of SQDs and LQDs are denoted τ_{SQD} and τ_{LQD}, respectively. In this simple model, the yield (η_{tr}) of excitation transfer from SQDs to LQDs is given by $\eta_{tr} = \tau_{SQD}/(\tau_{SQD}+\tau_{dw})$. Figure 4(a) shows the dependence of the excitation transfer yield η_{tr} on the spacer thickness as estimated from the ratio of the integrated PL intensities of SQDs and LQDs in the spectra shown in Fig. 2, assuming equal or, in the case of GaAs excitation of samples with $Al_{0.21}Ga_{0.79}As$ layer, a 5:1 ratio of the excitation rates for SQDs and LQDs. The η_{tr} decreases with increasing spacer thickness as well as the addition of an $Al_{0.21}Ga_{0.79}As$ layer for the same overall spacer thickness. The excitation transfer probability decreases with increasing width and height of the tunnel barrier.

A QD ground-state lifetime of ~600 ps has been determined for 1.74-ML InAs single layer as well as constant deposition amount multilayer samples grown at the same conditions.[6] Figure 4(b) depicts on a semilogarithmic scale the dwell time τ_{dw}, derived using η_{tr} as defined previously and assuming τ_{SQD}=600 ps, versus the spacer thickness. For a pure GaAs spacer τ_{dw} increases exponentially from ~20 ps (36 ML spacer) to ~3000 ps (54-ML spacer) with increasing spacer thickness. The larger relative increase of τ_{dw} upon inclusion of the $Al_{0.21}Ga_{0.79}As$ layer for the 45 ML as compared to the 36 ML spacer sample is attributed to its increased width of 16 ML compared to 7 ML. The low η_{tr} observed for the 54 ML sample with $Al_{0.21}Ga_{0.79}As$ layer suggests excitation transfer to be negligible.

We note that for the AQDP structures the three-dimensional shape of the InAs islands makes a definition of a tunnel barrier thickness somewhat arbitrary and that the nonuniformity of the islands leads to statistical variations. We approximate an effective tunnel barrier with the average distance between the island tip in the seed layer and the second InAs layer. The upper scales of Figs. 4(a) and 4(b) give the average effective tunnel barrier thickness taking into account the average island height of ~4.5 nm determined from atomic force microscope images of uncapped single layer 1.74-ML samples.[13] The dwell times for the AQDPs are comparable to those reported for asymmetric double $GaAs/Al_xGa_{1-x}As$ quantum wells with similar barrier thickness.[10,11]

Room-temperature PL measurements (not shown) reveal for LQDs an excited-state transition at ~100 meV above the ground-state transition, nearly coinciding with the ground-

state transition of the SQDs. However, as a result of the nonuniformity of self-organized QDs the AQDPs are, in general, nonresonant. Thus we conclude that the excitation transfer between SQDs and LQDs is a nonresonant multiphonon-assisted tunneling process. The results are in qualitative agreement with recent calculations that predict[17] electron dwell times shorter than 100 ps for QD/deep defect pairs with separations less than ~10 nm. We emphasize that the actual transfer mechanism, i.e., exciton or separate transfer of electrons and holes is not clear yet. In the latter case, the estimated dwell time τ_{dw} would characterize the faster transfer mechanism, most probably that of electrons, resulting in the formation of an intermediate, spatially indirect exciton. The low recombination probability and the finite dwell time for the second carrier (the hole) effectively suppress PL from the indirect exciton state. Finally, we assumed the tunneling processes to originate in the SQD ground state. This assumption becomes questionable for the 36-ML spacer, for which the estimated dwell time of ~20 ps is shorter than intradot relaxation on a 40-ps time scale.[6] In this case, the estimated dwell time presents only an upper limit.

We have presented structural and optical results for a new class of bilayer stacks of self-assembled QD structures that involve differing deposition amounts and lead to vertically self-organized QD pairs, whose density is controlled by the first-layer deposition amount while the average size of the second-layer dots is controlled by its deposition amount.[7] Such ''asymmetric'' QD pairs enable studies that provide insight into excitation transfer processes in self-organized QD stacks. Controlling the spacer thickness within the limits imposed by the need for vertical self-organization[2] allows tuning the carrier dynamics in the SQDs from the recombination-limited to the excitation transfer-limited regime. The estimated dwell times range from ~3000 ps for 54 ML down to the few 10 ps region for 36-ML GaAs spacer. The results suggest that lateral energy transfer in an InAs/GaAs QD layer can be observed for areal densities in excess of $\sim 2\times10^{11}$ cm^{-2} and that asymmetric QD pairs may provide a means to circumvent the slowed-down carrier relaxation in single QDs.

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