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State filling dependent luminescence in hybrid tunnel coupled dot–well structures

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A strong dependence of quantum dot (QD)–quantum well (QW) tunnel coupling on the energy band alignment is established in hybrid InAs/GaAs–InGaAs/GaAs dot–well structures by changing the QW composition to shift the QW energy through the QD wetting layer (WL) energy. Due to this coupling a rapid carrier transfer from the QW to the QD excited states takes place. As a result, the QW photoluminescence (PL) completely quenches at low excitation intensities. The threshold intensities for the appearance of the QW PL strongly depend on the relative position of the QW excitonic energy with respect to the WL ground state and the QD ground state energies. These intensities decrease by orders of magnitude as the energy of the QW increases to approach that of the WL due to the increased efficiency for carrier tunneling into the WL states as compared to the less dense QD states below the QW energy.

Introduction

The unique optical and electronic properties of quantum dots (QDs) make them suitable for the study of fundamental physical problems and applications in a range of novel opto-electronic devices. Recent techniques of QD growth allow for the fabrication of single QDs, QD aggregates (molecules), and dense arrays of uniform QDs. Such state-of-the-art nanoscale structures allow for the exploration of many challenging problems in various areas of physics: quantum electrodynamics of solid state materials by placing semiconductor QDs inside a photonic crystal, micropillar or microdisk resonator, spintronics through the observation of pure dephasing of a QD spin qubit interacting with a nuclear spin bath, quantum optics using QDs as efficient nanoscopic light emitters, and quantum information processing. Many high-performance devices have been developed recently based on QDs: QD hetero-structure lasers, high speed nanophotonic devices, lasers for optical communication, etc. QD systems have been further developed using various hybrid structures, such as QD bioconjugates for imaging, labeling and sensing, integrated organic and inorganic materials at the nanometer scale for hybrid light-emitting diodes, and colloidal QDs in hybrid organic–inorganic structures. Recently, hybrid systems composed of QDs brought in close contact with a quantum well (QW) are of particular interest, because they combine the advantages of both the zero-dimensional (0D) QD and the two-dimensional (2D) QW structures. As a result, such 0D–2D systems have been used in photonic crystals, infrared detectors, and tunneling-injection lasers, for example.

Ideally, single QDs have discrete atomic-like energy spectra and delta function-like density-of-states. In reality, electrons in self-assembled QDs are strongly coupled to phonon excitations, which significantly alters the electronic and optical density of states as well as the carrier relaxation dynamics. Also, self-assembled QDs form naturally on top of and only after the formation of a highly strained 2D wetting layer (WL) of the same material, and they are normally capped by a crystalline layer. They are therefore embedded in a matrix of a crystalline medium. Coupling with this surrounding as well as immediate coupling between neighboring QDs significantly modify the QD properties also. Besides the discrete atomic-like electron states, complementary QD states can arise due to a coherent hybridization of localized QD states and extended continuum states through electron/hole tunneling or photon exchange. Also, variations in the confinement potential in the surrounding of the QD may result in the appearance of localized electronic states with large oscillator strength regardless of the WL thickness and even of inter-diffusion. A strong 0D–2D hybridization is expected for the QD and WL hole states giving rise to 2D-to-0D crossed transitions in photoluminescence (PL) which have indeed been detected experimentally. Such complicated energy structure and density of states generated by embedded QDs provide numerous possible channels for trapping carriers from...
The surrounding into QDs. Also, the embedding matrix affects the escape dynamics of carriers from the QD and therefore influences the quantum efficiency. A careful design of the QD environment and of the hybridization of the environment with the QD states is therefore needed in order to optimize their optical properties. Recently, it was demonstrated that QW–QD hybrid structures with thin barriers exhibit a particularly strong dot–well coupling which is tunable and can greatly enhance the carrier collection efficiency of the QD layer. By independently varying the thickness and height of the barrier layer between the dots and the well it was possible to control the strength of the dot–well coupling and, hence, the carrier flux from the QW to the QD layer. In this paper we demonstrate how this flux depends on the energy band alignment in the QW–QD hybrid structures composed of InAs QDs and an In$_x$Ga$_{1-x}$As QW layer separated by a thin GaAs barrier. Varying the composition of the QW layer allows us to tune the QW energy between the lowest QD state and the WL energies. We show a strong dependence of the net QD and QW PL and PL excitation (PLE) response on the relative alignment of the quantum well, quantum dot and wetting layer optical transition energies.

**Experimental**

The QW–QD samples were grown by molecular beam epitaxy on semi-insulating GaAs (001) substrates. Each sample includes a 15 nm thick In$_x$Ga$_{1-x}$As QW with $x$ ranging from 0.07 to 0.20, a GaAs spacer of thickness either 4 nm or 5 nm, and a layer of self-assembled QDs formed by the deposition of 2 ML of InAs on the GaAs spacer. For optical measurements, samples were covered with a 50 nm GaAs cap. A set of reference samples were also grown, one with a single QD layer only and a set with a single QW layer each for each of the different QW compositions, all grown, one with a single QD layer only and a set with a single QW layer each for each of the different QW compositions, all using the same growth conditions as the QW–QD nanostructures. A morphological and structural analysis of the hybrid samples was carried out using a FEI Titan 80–300 Transmission Electron Microscope (TEM). Fig. 1a shows a multi-beam bright field TEM image of the structure with its schematic representation in Fig. 1b. It is clearly seen that the interfaces between the different layers are sharp and well defined, showing no signs of any contamination. The diffraction contrast (dark in the image) directly below the dots does not extend beyond the spacer layer and does not penetrate into the well. This implies that the growth-induced strain profile is limited to the QD/WL/barrier interface and affects the QW only weakly. The TEM analysis indicates that the QDs have an average diameter of 20 nm and a height of 5 nm with an areal density of $\sim$10$^{10}$ cm$^{-2}$.

The PL measurements were performed in a variable temperature, closed-cycle, helium cryostat using the 532 nm line from a Nd:YAG laser for excitation. The laser spot diameter was $\sim$20 µm and the optical excitation power was in the range of $\sim$10$^{-7}$ to 10$^2$ mW. The PL signal from the sample was dispersed by a monochromator and detected by a liquid nitrogen-cooled InGaAs photodiode detector array. The PLE measurements were carried out using a tunable Ti:sapphire laser. Time-resolved PL measurements were performed using 2 ps pulses at $\lambda = 750$ nm from a Ti:sapphire laser. For detection, a monochromator and a synchroscan streak camera equipped with an infrared-enhanced S1 photocathode were used. The overall time resolution of the set-up was $\sim$15 ps.

**Results and discussion**

Fig. 2 shows low temperature ($T = 10$ K) PL spectra measured under increasing excitation intensity, $I_{ex}$, for the sample with $x = 0.15$ and $d_{sp} = 4$ nm. At the lowest excitation intensity $I_{ex} = 1.5 \times 10^{-6} I_0$, $I_0 = 1000$ W cm$^{-2}$, the QD exciton ground state is found at $E_{QD}^{ex}$ $\sim$ 1.10 eV and is fit well by a single Gaussian with a full width at half-maximum (FWHM) of $\Gamma_0 = 32$ meV. Such a small linewidth reflects the highly homogeneous size distribution of the QD ensemble. With increasing $I_{ex}$, additional PL bands emerge at $\lambda_{QD}^{ex}$ ($i = 1, 2, 3, \ldots$), corresponding to the QD exciton excited states resulting from state filling at higher excitation intensities. The maxima of these additional bands are separated by an energy of $\sim$66 meV with correspondingly narrow linewidths, $\Gamma_i = 34 \pm 3$ meV. In addition to the observed QD

![Fig. 1](image1.png) (a) Multi-beam bright field TEM image of the InAs/GaAs QD–In$_x$Ga$_{1-x}$As/GaAs QW structure with an indium composition of $x = 0.18$ and a spacer layer thickness of $d_{sp} = 4$ nm and (b) its schematic clearly defining different regions. The diffraction contrast directly below the dots (dark color) is the result of strain from the dots affecting the surrounding lattice.

![Fig. 2](image2.png) Low temperature ($T = 10$ K) PL spectra measured at different excitation intensities in the InAs/GaAs QD–In$_{0.15}$Ga$_{0.85}$As/GaAs QW structure with $d_{sp} = 4$ nm. The spectra are vertically shifted for clarity. $I_0 = 1000$ W cm$^{-2}$.
excited states, a strong PL band reflecting QW heavy hole exciton emission develops at $E_{QW}^X = 1.347$ eV.

We observe an important correlation between the QW and the QD emission in Fig. 2. The QW excitonic emission appears only if all QD states at energies below $E_{QW}^X$ are filled at high $I_x$. At low excitation power no QW PL signal is detected. This implies that the tunneling-induced dot–well coupling in these thin spacer-layer samples ($d_{sp} = 4$ nm) is sufficiently strong to fully suppress the QW PL. Due to this coupling the carriers effectively escape from QW states into the energetically lower-lying QD states quenching the QW emission at low excitation power. Raising the excitation intensity, $I_x$, increasingly fills the lower-lying QD states. This state-filling reduces the carrier transfer probability, thus populating the QW long enough to observe its exciton state.

In order to test this simple picture of the QW–QD system we examine the system as the energy of the QW ground state approaches the energy of the InAs WL. Here, we choose samples with QW composition of $x = 0.07, 0.10, 0.15, 0.18$ and a spacer thickness of $d_{sp} = 4$ nm. Using the results of Fig. 2 and ref. 29, the energy band alignment in these hybrid structures can be presented as shown schematically in Fig. 3 where the excitonic transition energies of the lowest QW sub-band are shown for the four hybrid samples relative to the energies of QD excited states. Carrier transfer from QW states to QD excited states is shown with thick horizontal arrows, whereas the radiative transitions are shown as thin wavy arrows. One can see from Fig. 3 that a decrease in $x$ increases the QW exciton energy $E_{QW}^X$. In this case the number of QD states with an energy of less than $E_{QW}^X$ increases. Thus, the excitation intensity needed to saturate these states and observe a noticeable QW PL signal also has to be increased.

Fig. 3 shows the PL spectra of these four samples along with that of the reference QD sample measured at $I_x = 10I_0$. The bottom inset of Fig. 4 demonstrates the normalized PL signal measured at low excitation intensity for the set of reference QWs. It can be seen from Fig. 3 that for $x = 0.07$ and 0.10 the emission energy of the QW, $E_{QW}^X$, is above the four lower emission energies of the QDs, while for $x = 0.15$ and 0.18 the QW emission is at an energy below that of the 4th QD excited state. At the same time, the intensity of the QW emission for $x = 0.07$ and 0.10 is significantly lower than the intensity for $x = 0.15$ and 0.18 measured under the same excitation conditions. These observed tendencies are also present for the sample set with $d_{sp} = 5$ nm. Fig. 5 shows the PL spectra of two representative hybrid samples from this set with $x = 0.15$ and 0.20, along with the reference QD sample. For $x = 0.20$, $E_{QW}^X$ is above the energy of only the 2nd excited QD state, $E_{QD}^2$, whereas for $x = 0.15$, $E_{QW}^X$ exceeds the energy of the 3rd excited QD state, $E_{QD}^3$. In comparison with the $d_{sp} = 4$ nm series, the QW intensity in these samples becomes significant at much lower excitation intensity. However, we again observed that the QW PL is significantly reduced if $E_{QW}^X$ exceeds the energy of a subsequent excited QD state.

In general, the PL spectra of all hybrid samples at energies below $E_{QW}^X$ are very similar to those of the reference QD sample in the respective energy region (Fig. 4 and 5). We also find that the amplitude of the GaAs free exciton PL band ($\lambda_{GaAs} = 824$ nm) remains constant for all samples studied. This observation indicates that the incorporation of the QW into the QD samples does not introduce new trapping centers in the hybrid structures in comparison with the reference sample. Such centers could...
rapid thermalization, generate under non-resonant continuous wave (cw) excitation into the nanostructures. Now, we present a simplified description of the QW ground state. These carriers further relax fulfilling the

\[ I_{\text{ex}} = \frac{N_{\text{QW}}(I_{\text{ex}})}{N_{\text{QW}}(I_{\text{ex}})} \]

and

\[ N_{\text{QW}}(I_{\text{ex}}) = N_{\text{R}}^{\text{QW}} + N_{\text{NR}}^{\text{QW}} + N_{\text{QW}}^{\text{QW}}. \]

Here, \( N_{\text{QW}}^{\text{QW}} \) is the number of carriers recombining radiatively per unit time in the QW layer and contributing to the QW PL signal \( N_{\text{PL}}^{\text{QW}} \), \( N_{\text{NR}}^{\text{QW}} \) denotes the non-radiative recombination rate in the QW; and \( N_{\text{QW}}^{\text{QW}} \) is the flux of carriers tunneling from QW states into the lower-lying QD states. From eqn (1) we can derive the QW steady-state population \( n_{\text{st}}^{\text{QW}}(I_{\text{ex}}) \) as

\[ n_{\text{st}}^{\text{QW}}(I_{\text{ex}}) = N_{\text{QW}}^{\text{QW}}(I_{\text{ex}})(\gamma_{\text{R}} + \gamma_{\text{NR}} + \gamma_{\text{T}})^{-1} \]

where \( \gamma_{\text{R}} \), \( \gamma_{\text{NR}} \), and \( \gamma_{\text{T}} \) are the radiative, non-radiative, and carrier transfer rates, respectively. In this model we chose for simplicity to neglect the back-transfer of carriers from QD states to the QW as the intra-dot carrier relaxation dynamics is on a (sub-) picosecond time scale,\(^2\) much faster than any of the rates defined in eqn (2). Defining the QW PL intensity as

\[ I_{\text{PL}}^{\text{QW}}(I_{\text{ex}}) = \beta N_{\text{RK}}^{\text{QW}}, \]

where \( \beta \) is a factor which depends on the actual experimental arrangement and using eqn (1) for \( N_{\text{QW}}^{\text{QW}} \), we have

\[ I_{\text{PL}}^{\text{QW}}(I_{\text{ex}}) = \beta(N^{\text{QW}}(I_{\text{ex}}) - N_{\text{QW}}^{\text{QW}} - N_{\text{NR}}^{\text{QW}}). \]

The \( N_{\text{T}}^{\text{QW}} \) term in eqn (3), which describes the carrier flux from QW to QD states, can be represented in a form that explicitly takes into account the QD state filling.

\[ N_{\text{T}}^{\text{QW}}(I_{\text{ex}}) = \frac{n_{\text{R}}^{\text{QW}}(I_{\text{ex}}) \sum_{i=0}^{g}(N_{i} - n_{i}(I_{\text{ex}}))/\tau_{i}^{\text{QW}}}{\tau_{i}^{\text{QW}}/C_{0}} \]

where \( n_{i}(I_{\text{ex}}) \), \( N_{i} \), and \( \tau_{i}^{\text{QW}} \) are the population, degeneration, and tunneling time from the QW to the \( i \)th QD state, and \( \Omega \) denotes the highest index, \( i \), for which the QD excited state energy, \( E^{\text{QD}}_{\text{i}} \), lies below the energy of the QW, \( E^{\text{QW}}_{\text{X}} \). We assume that the tunneling time, \( \tau_{i}^{\text{QW}} \), is only weakly dependent on the index \( i \).

Even though state-dependent tunneling dynamics are clearly expected, they will have only a limited effect on the QD state-filling and therefore on the intensity-dependent onset of QW emission, the main scope of the present model. By taking the degeneracy to be of the form \( N_{i} = 2(i + 1) \), one can find the number of QD states (the capacity of the QD system) that are filled by tunneling from the QW layer,

\[ \sum_{i=0}^{g} N_{i} = (\Omega + 1)(\Omega + 2) \]

Using eqn (3) and (5) we can derive the threshold excitation intensities over which the QW PL for various positions of \( E^{\text{QW}}_{\text{X}} \) with respect to the QD excited states is non-zero.

\[ I_{\text{PL}}^{\text{QW}}(I_{\text{ex}}) = \beta\left(N^{\text{QW}}(I_{\text{ex}}) - N_{\text{QW}}^{\text{QW}} - N_{\text{NR}}^{\text{QW}}\right) \]

\[ = \alpha(I_{\text{ex}})[A(I_{\text{ex}}) - (\Omega + 1)(\Omega + 2)] \]

with

\[ \alpha(I_{\text{ex}}) = \beta n_{\text{R}}^{\text{QW}}(I_{\text{ex}})/\tau_{i}^{\text{QW}} \]

and

\[ A(I_{\text{ex}}) = \tau_{i}^{\text{QW}}(N^{\text{QW}}(I_{\text{ex}}) - N_{\text{NR}}^{\text{QW}})n_{\text{st}}^{\text{QW}}(I_{\text{ex}}). \]

In this model, a finite QW emission occurs if the term in brackets in eqn (6), \( A(I_{\text{ex}}) - (\Omega + 1)(\Omega + 2) \), is positive. This introduces a threshold intensity for QW PL, which is defined largely by \( \Omega \), i.e., the total number of electronic states in the quantum dot below the energy of the QW. Eqn (6) contains two parameters at a fixed \( I_{\text{ex}} \) and can be used for fitting the data presented in Fig. 4 and 5. Fig. 4a shows the dependence of \( I_{\text{PL}}^{\text{QW}} \) on \( \Omega \) for the special case of \( d_{\text{ex}} = 4 \) nm and \( I_{\text{ex}} = 10I_{\text{ex}} \), i.e., when the QD excited states are substantially filled. The results of this fit support the plausibility of the model based on the carrier transfer from the QW to the QD layer affected
by QD state filling limited tunneling. We note that the $x = 0.07$ sample is of particular interest with the highest QW energy. As can be seen from its PL spectrum (Fig. 4), the QW exciton energy, $E_{\text{QW}} = 1.422$ eV, is very close to the energy of the exciton transition in the WL of the reference QD sample, $E_{\text{WL}}^{\text{QD}} = 1.427$ eV. The light-hole (lh) exciton transition energy of the WL in the reference QD sample is found at $E_{\text{WL}}^{\text{QD}} = 1.485$ eV. For thin barriers ($d_{\text{ep}} = 4$ nm), the small energy difference between the excitonic transitions in the QW and the WL results in an efficient tunnel coupling between these two systems which form an asymmetric double well potential.

Tangible evidence for this direct tunnel coupling between the WL and the QW is obtained from PLE spectra shown in Fig. 6. All spectra are recorded at $T = 10$ K with a very low excitation intensity of $I_{\text{ex}} = 1.5 \times 10^{-6} I_0$. The detection wavelength is set to the maximum of the QD ground state emission at $\lambda_0^{\text{QD}}$ in each sample. All spectra show similarly strong peaks at the 818 nm exciton resonance of the GaAs buffer layer. This indicates that the carrier trapping from the GaAs buffer into the QDs is not drastically affected by the insertion of the various QW layers. The PLE spectrum taken on the reference QD sample shows clear resonances from the hh and lh exciton absorptions at 861 nm and 833 nm, respectively. The PLE spectra of the hybrid structures display clear peaks at the ground and first excited exciton resonances of the InGaAs QW. As expected, these resonance energies, $E_{\text{QW}}^{\text{QD}}$ and $E_{\text{QW}}^{\text{QD}}$, the two lowest energy peaks marked with arrows in sample #1, red-shift when increasing $x$. We observe little change in width or amplitude of the QW resonances in samples #1 to #3. However, in sample #4, we find a more complicated spectrum of peaks, indicated by the arrows on the sample #4 curve in Fig. 6. Here, the energies of the WL hh transition and the QW ground state exciton resonance are almost coincident. We interpret the multiple peaks found within this energy range as a quantum mechanical anti-crossing between these two states. The energy splitting of this anti-crossing is ~6 to 8 meV, indicating electron tunneling times of a few hundred femtoseconds between the WL and QW states in this sample. The observation of such an anticrossing would imply that the corresponding QW and WL electronic states become strongly hybridized, suggesting that this WL–QW double layer essentially acts as a “super-wetting layer” which efficiently traps carriers in either the GaAs barrier, the QW or the WL. Subsequently, the carriers are then injected into the QD states allowing for recombination. This apparent hybridization makes this sample interesting for time-resolved experiments aimed at tracking the microscopic carrier relaxation dynamics in the tunnel-coupled hybrid nanostructures in real time.

To confirm our assumption that the peak splitting seen around 1.433 eV (865 nm) for sample #4 in Fig. 6 indeed reflects a hybridization between the QW and WL states, we have performed simulations of the relevant electronic states and optical transitions in our samples using the nextnano simulation software. We used an effective mass approximation for a hybrid system comprising a 0.4 nm InAs WL, a 4.0 nm spacer and a strained 14 nm InGaAs QW layer with variable indium composition. The choice of WL and spacer layer matches X-ray diffraction analysis of our sample structure and was found to give good agreement with our optical measurements.

The energies of the three lowest electron states and the nine lowest hole states returned from these simulations are plotted as a function of indium composition in Fig. 7. The plot reveals distinct anticrossings between the highest energy heavy hole state confined in the wetting layer (at 1.52 eV) and the different heavy hole state of the strained QW layers. The splitting is on the order of 2–3 meV. Fig. 7 also reveals clear anticrossings between the lowest energy electron state in the wetting layer (at 2.95 eV) and the ground and first excited confined electron state in the quantum well layers. The anticrossing between the WL state and the QW ground state occurs for a composition of $x \approx 0.055$ and a splitting of ~15 meV, is predicted. A similar anticrossing between the WL state and the first excited QW state occurs for $x = 0.105$, showing a splitting energy of 25 meV. These anticrossings occur very near the indium compositions for which we find peak splittings in the experimental PLE data. Also, the magnitude of the predicted splitting between the coupled electron states matches well with experiment for the two higher energy peaks, whereas two lower energy peaks for sample #4 might be interpreted as being primarily due to the much smaller hole level splittings. This provides additional evidence that the experimentally observed PLE splittings reflect anticrossings due to level hybridization between coupled QW and WL states.

For a quantitative interpretation of our experimental PLE spectra, we have simulated the absorption coefficients of the corresponding optical interband transitions. These results suggest the following assignment of the main transitions seen in Fig. 6. For the reference QD sample, the lowest energy peaks are the e1–hh1-transition between electron and heavy hole states and...
the e1–lh1-transition between electron and light hole states all localized in the WL. For sample #1, $x = 0.18$, the lower energy dominant peaks are the e1–hh1 and e2–hh2 transitions predominantly localized in the QW. Simulations indicate that the third lowest peak (at 1.41 eV in Fig. 6) is the result of the lowest light-hole transition, e1–lh1, which is localized in the QW. The first transition involving electron states in the WL appears at ~1.44 eV. This full spectrum for sample #1 is only weakly affected by carrier tunneling and hybridization between the WL and the QW. The same holds true for sample #2 ($x = 0.15$) where only a slight blue-shift is apparent as compared to sample #1, resulting from the decreased In composition. Similarly, for sample #3, $x = 0.10$, the lowest energy peak again corresponds to the QW-like e1–hh1 transition. The next higher peak, however, contains contributions from the e2–hh2 and e2–hh3 transitions and is strongly affected by the coupling between the excited QW and the WL conduction band states. The effect of the QW–WL hybridization is most pronounced for sample #4, $x = 0.07$. Here, the QW and WL conduction band ground states are strongly hybridized, resulting in four low-energy peaks (e1–hh1, e1–hh2, e2–hh1 and e2–hh2), marked by arrows in Fig. 6. For all transitions, the involved hybridized electron wavefunction is delocalized between the WL and QW regions. We take the fact that modifications of the PLE spectrum due to electron tunneling between WL and QW are predicted for exactly those transitions in the experimental spectrum (Fig. 6) which show a splitting into multiple peaks as clear and convincing evidence for a tunneling-induced hybridization of the QW and WL states in sample #4. Even though the trends seen in the transitions of Fig. 7 match our data in relative energy and appropriate alloy concentrations extremely well, due to the simplicity of our model and the resolution of our data, a quantitative comparison between the theoretically predicted transition energies and the peaks in the PLE data of Fig. 6 is beyond the scope of this paper.

The results of a preliminary time resolved PL measurement carried out on these hybrid samples are shown in Fig. 8. Here, the samples are excited at a non-resonant wavelength of 750 nm, i.e., above the energy of the GaAs barrier. The PL transients are then recorded with a streak camera, detecting at the maximum of the QW emission band, ranging from 930 nm for sample #1 to 863 nm for sample #4. All measurements are recorded at $T = 10$ K and at an average laser power of $8 \mu$W, corresponding to a peak excitation intensity $I_p = 1.3 \times 10^4$ W cm$^{-2}$ which was chosen sufficiently low in order to insure that the state-filling effects do not affect the relaxation dynamics. The low power regime is determined by successively reducing the excitation intensity until single exponential transients with intensity-independent decay times are observed. At higher excitation intensities, distinctly non-exponential PL transients reflecting the delayed decay of the QW PL due to QD state-filling are found (not shown). The transients were fit by using a simple monoexponential decay model, convoluted with a 15 ps instrument response function. The quality of these fits supports the validity of this model. The inset of Fig. 8 shows the dependence of the PL decay times on the QW ground state exciton energy $E_{exc}$ in the hybrid samples. The WL decay time is shown by an empty circle. The vertical bars give an experimental accuracy.

Fig. 7 Effective mass calculations of the three lowest energy electron states (e1–e3) and nine lowest heavy hole (hh1–hh7) and light hole (lh1–lh2) states in the WL–QW region of the hybrid QD–QW system. The simulations are performed for a 14 nm thick In$_{x}$Ga$_{1-x}$As/GaAs QW separated by a 4 nm GaAs barrier from a 0.4 nm thick InAs WL as a function of Indium composition $x$. Clear anticrossings between the WL and QW conduction band states are seen for $x = 0.06$ and 0.10 for the ground and first excited QW states respectively. The low energy optically allowed transitions are marked for $x = 0$ (reference QD), $x = 0.07$ (sample #4), $x = 0.10$ (sample #3) and $x = 0.18$ (sample #1) as arrows.

Fig. 8 PL transients measured at the maximum of the QW PL band in the hybrid samples. For comparison, the instrument response function recorded with scattered laser light is also shown. The inset shows the dependence of the PL decay times on the QW ground state exciton energy $E_{exc}$ in the hybrid samples. The WL decay time is shown by an empty circle. The vertical bars give an experimental accuracy.
reference QD sample, with a decay time of $\tau_{\text{QW}}$. The data are shown together with the transient from the WL in the reference QD sample, with a decay time of $\tau_{\text{WL,refQD}} = 30\,\text{ps}$. We see here that the decay times decrease from 70 ps (#1) to 28 ps (#4) as $E_{X}^{\text{QW}}$ tends towards $E_{X}^{\text{NL}}$, ultimately approaching the same decay time of WL for the case of sample #4 where we presume a strong WL–QW hybridization. The rapid PL decay for sample #4 can indicate that the measurement mainly probes PL from the WL or that the carrier transfer between QW and WL is faster than the WL to QD transfer which limits the PL decay time in this sample. Since the maximum amplitude of the time-resolved PL signal increases by more than a factor of two when decreasing $x$ from 0.18 to 0.07, we expect a significant contribution of QW emission to the PL of sample #4 and thus conclude that the QW-to-WL carrier transfer is much faster than the WL PL decay time of 30 ps. This is fully consistent with the WL–QW hybridization deduced from the PLE data of Fig. 6 which show peak splittings of several meV and therefore indicate sub-ps electron tunneling dynamics between WL and QW ground states. For the samples with higher indium concentrations, the tunneling dynamics between the WL and the QW and subsequent capture by the QD states becomes slower, resulting in an increase in the PL decay times.

Evidently, the time resolution of such streak camera measurements with non-resonant excitation is limited by the finite instrument response time (20 ps here) but apparently also by the rather slow carrier relaxation dynamics from the GaAs barrier into WL and QD states. This clearly calls for experiments with selective resonant excitation of the WL, QW and QD states to further elucidate the microscopic carrier tunneling dynamics in such QW–QD hybrid structures. Our PLE results, specifically the observed WL–QW hybridization in Fig. 6, suggest that such resonantly excited experiments should be performed with a time resolution of at least 100 fs and may reveal evidence for coherent tunneling dynamics even in ensemble measurements. The results of these studies will be published separately. The carrier relaxation dynamics probed in such ensemble studies is likely to be affected by the inhomogeneous broadening of the QD ensemble as well as the unavoidable disorder effects from the QW and the WL. It may therefore be interesting to complement such resonant ensemble measurements with time-resolved measurements of single QDs within this system.34,35

**Conclusions**

In summary, we have observed a strong coupling in hybrid InAs/ GaAs–In$_{x}$Ga$_{1-x}$As/GaAs dot–well structures which varies in strength as the QW excitonic emission energy varies through the QD excited states by changing the indium composition of the quantum well. As a result of a rapid carrier tunneling from the InGaAs QW to the QD excited states, the QW PL completely quenches at low excitation intensities. With increasing excitation intensity, the QD excited states emerge and subsequently fill, followed by the QW emission. The threshold intensities at which the appearance of the QW PL takes place depend strongly on the position of the QW excitonic energy, $E_{X}^{\text{QW}}$, within the range of energies bound by the WL and the QD ground state. These intensities grow by orders of magnitude when the QW energy approaches the WL energy which is a result of having to fill a greater number of available QD excited states at lower energies than the QW energy. Our PLE experiments give preliminary evidence for a hybridization of the QW and WL states when tuning both into resonance. Ultrafast coherent tunneling dynamics on a 100 fs time scale are anticipated as a result of this hybridization, effectively transforming the coupled WL–QW layers into a reservoir with a greatly enhanced density of states for injecting cold carriers into quantum dots. A more detailed analysis of the microscopic carrier dynamics in such hybrid structures is currently underway.

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**Notes and references**