Impact of carrier redistribution on the photoluminescence of CdTe self-assembled quantum dot ensembles

S. Mackowski*
Department of Physics, University of Cincinnati, Cincinnati, Ohio 45221-0011, USA

G. Prechtl and W. Heiss
Institut für Halbleiter-und Festkörperphysik, Universität Linz, 4040 Linz, Austria

F. V. Kyrychenko
Department of Physics, University of Florida, Gainesville, Florida 32611-8440, USA

G. Karczewski and J. Kossut
Institute of Physics, Polish Academy of Sciences, 02-668 Warszawa, Poland

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We report on the photoluminescence (PL) measurements of two structurally different CdTe self-assembled quantum dot (QD) samples. These include CdTe QD's connected with the uniform two-dimensional wetting layer (WL) and fully developed CdTe QD's that are isolated from each other. Both temperature and excitation power dependencies found for the ensemble of fully developed QD's reflect that of individual QD's. In this case, no signature of intradot carrier redistribution is observed even at 120 K. In contrast, both continuous-wave and time-resolved PL show the presence of strong thermally induced carrier redistribution for the structure, where the dots are connected by a uniform two-dimensional WL. In particular, three distinct temperature regions are identified. At temperatures below 30 K, dots in the ensemble are isolated from each other: the emission occurs only from QD's occupied initially by both electron and hole. When temperature increases ($T>30$ K), the carriers from QD's are thermally activated into the uniform WL and are subsequently captured by other QD's. Finally, above 100 K the nonradiative recombination in the WL prevents the capture of thermally activated carriers. These results, while providing a thorough understanding of the optical properties of QD structures, give also important information about structural complexity of epitaxially grown II-VI semiconductor QD's.

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I. INTRODUCTION

The optical properties of excitons confined to semiconductor self-assembled quantum dots (QD's) have been studied very intensively for almost a decade.[1] Among a variety of different techniques, photoluminescence (PL) measurements performed both on single QD's[2–10] and on large QD ensembles[11–20] have provided a detailed understanding of how the zero-dimensional confinement in QD's influences the properties of excitons. For instance, it has been demonstrated that, similar to semiconductor quantum wells (QW's), the change of the QD size and of the average distance between QD's may alter the energies of electronic levels and the interaction between neighboring QD's.[4,10,20,21] These results show that morphology of the QD system plays an important role in determining the optical properties of a particular quantum structure.

Most of the studies have been focused on III-V semiconductor self-assembled QD's, such as $In_xGa_{1-x}As$ QD's embedded in a GaAs matrix, where QD's are formed during the epitaxial growth through a Stranski-Krastanov mode.[13] In this case, as displayed in Fig. 1(a), above a certain critical thickness of $In_xGa_{1-x}As$ layer, a coherent transformation from two-dimensional (2D) growth to three-dimensional (3D) growth takes place. One of the signatures of the Stranski-Krastanov transition is the presence of a uniform 2D WL that *physically* connects all the QD's in the ensemble [see Fig. 1(a)]. It is important to note that when increasing the nominal thickness of $In_xGa_{1-x}As$, the QD's get larger but the thickness of the WL remains constant.[13] As a result, at elevated temperatures, transfer of carriers between neighboring QD's could take place via the uniform WL. It has been shown that thermally induced carrier redistribution strongly modifies the temperature behavior of PL emission of such a QD structure.[22–26]

In contrast, for QD's made of II-VI semiconductor compounds [such as CdSe/ZnSe (Refs. 16, 27, and 28) or CdTe/ZnTe (Refs. 2, 29, and 30)], the mechanism of the dot formation differs from the Stranski-Krastanov mode. As it is shown in Fig. 1(b), in this case, the growth starts from randomly distributed 2D platelets, which serve as precursors for the QD growth.[16] This unique behavior has been ascribed to stronger diffusion of II-VI materials as compared to III-V compounds.[16] Recent results of scanning transmission electron microscopy (TEM) accompanied by micro-PL measurements show that both 2D-like and 3D-like objects exist simultaneously even if only 0.7 monolayers (ML's) of CdSe is deposited on the ZnSe substrate.[16] Remarkably, at this stage of the growth, no signature of a uniform 2D WL is seen. When adding more CdSe, the average thickness of these 2D platelets increases and, at the same time, the 3D QD's are getting larger. Interestingly, however, in the case of II-VI

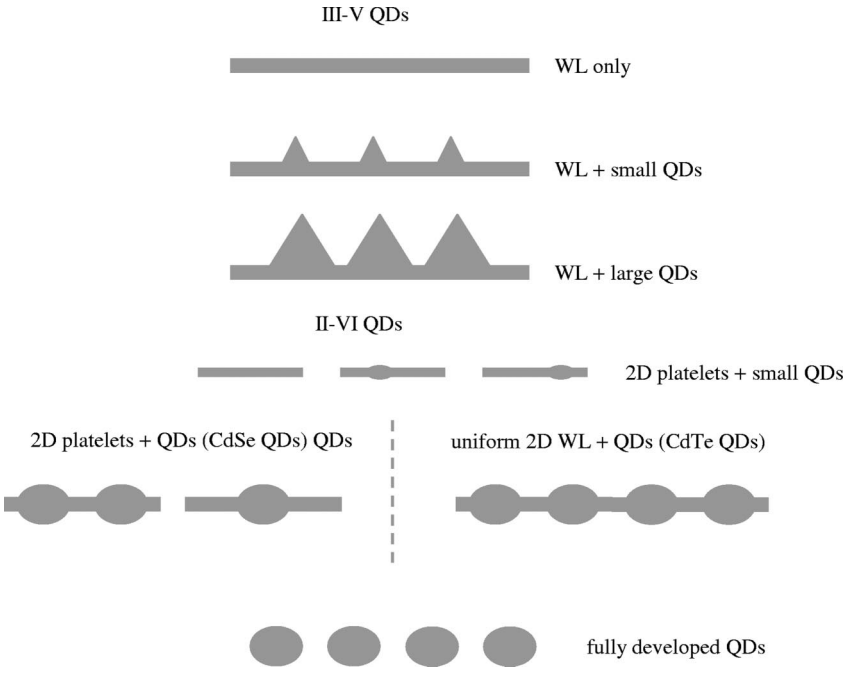

FIG. 1. (a) Schematic diagram of the Stranski-Krastanov growth mode typical for III-V self-assembled QD's. The growth starts from the WL formation, then small QD's are formed, and with further increase of the amount of deposited QD material the dots get larger. (b) Schematic diagram of QD formation observed for II-VI semiconductors: the growth starts from 2D platelets on which small dots are formed. Then two scenarios are possible as the growth continues: either the platelets with QD's remain isolated (left) or they form a uniform 2D WL (right). As the amount of the deposited QD material increases these both scenarios end with large isolated QD's.

semiconductors it is also possible that by changing the growth conditions, these 2D platelets may eventually form a uniform 2D layer that connects all the QD's in the ensemble. The formation of a 2D wetting layer has been indeed observed for CdTe QD's grown on a ZnTe substrate.[29] With a further increase of the amount of the QD material both scenarios end up with an ensemble of fully developed 3D QD's that are isolated from each other, with no uniform WL present in such a structure. This variety of qualitatively different morphologies characteristic for II-VI self-assembled QD's offers many possibilities for studying the influence of the structural properties on the mechanisms of carrier transfer between neighboring QD's.

In this work we report on optical properties of CdTe self-assembled QD's grown with a different nominal CdTe coverage. We show that depending on the amount of QD material deposited, the dots are either isolated from each other or connected by the uniform 2D WL.

For the QD sample with a nominal CdTe thickness of 4 ML we find that the PL energy dependence on the temperature obtained for a QD ensemble is identical with the one measured for single CdTe QD. Regardless of the temperature and excitation power, the emission of this sample reflects the distribution of ground-state energy within the QD ensemble. In addition, the measured decay time of the exciton recombination in these QD's is constant up to 100 K, and it is independent of the excitation power. This is the evidence that the carrier transfer between dots is inhibited and therefore we argue there is no uniform WL formed in this structure. In other words, the PL emission of this QD ensemble is determined solely by the properties of individual noninteracting QD's, in agreement with the growth scheme presented above.

On the other hand, when the nominal thickness of the CdTe QD layer is equal to 2 ML, both the temperature and the excitation power dependencies of the PL emission show drastically different behavior. In this case we observe three distinct temperature regions. At low temperatures (below 35 K) the line shape of the QD emission and the exciton decay time do not depend on the excitation power. Therefore, only the dots that are *initially* populated by electrons and holes contribute to the emission. In other words, any redistribution of carriers between QD's is inhibited. In contrast, in the temperature range between 35 and 100 K as the excitation power decreases, the maximum of the QD emission shifts towards lower energies. At the same time the linewidth of the QD emission gets narrower. Furthermore, for the weaker excitation powers the decay time of exciton recombination in QD's is much longer. We interpret these observations as a result of thermally induced carrier redistribution between different QD's. The carrier transfer between the QD's occurs via a 2D WL that connects the dots. It is important to note that although in this temperature range the emission of QD's depends strongly on the excitation power, both the linewidth and the energy of the 2D WL emission are unchanged, as is the decay time. Finally, as the temperature increases further, the redistribution of carriers between QD's via the WL is suppressed due to enhanced nonradiative recombination in the WL itself. As a result, in this temperature range, we find only a very weak power dependence of both energy and PL linewidth of the QD emission. A simple model that takes into account thermal activation of carriers originally localized in QD's into the WL and their subsequent capture by another dot reproduces these experimental observations well. These results of PL spectroscopy provide a detailed description of the interdot carrier dynamics in CdTe QD's connected with a uniform WL. Moreover, these measurements enable one to describe by optical means the global morphology of the studied QD system.

II. SAMPLES AND EXPERIMENTAL DETAILS

Two CdTe/ZnTe QD samples with qualitatively different morphology are studied in this work. They were grown by

molecular beam epitaxy on (100)-oriented GaAs substrates by deposition of 2 and 4 ML of CdTe on top of ZnTe buffer layers. The substrate temperature was equal to 280 °C. The first sample consists of CdTe QD's superimposed on a 2D WL, while the second one contains only fully developed isolated QD's. From the TEM images we estimated the typical size (diameter) of the CdTe QD's to be 2–4 nm,[29,31] while their density approaches 10^{12} cm^{-2}. Further details of the growth as well as the optical characterization of these samples can be found elsewhere.[29,31]

Continuous-wave PL studies of the large QD ensembles were carried out in two modes. First, for a constant temperature of the sample, the excitation power dependence of the emission was measured. Second, the temperature dependence of the PL was studied for a constant excitation power. In these experiments the 457-nm line of an argon-ion laser was used for excitation with a spot size of 200 μm in diameter. The PL signal was detected by a liquid-nitrogen-cooled CCD camera after being dispersed by a 0.25-m monochromator (grating 1800/mm).

The PL measurements of single CdTe QD's (micro-PL) were performed with the laser focused down to a spot size of 1 μm in diameter by the use of a microscope objective. The sample was mounted in a continuous-flow helium cryostat and the temperature was changed from 5 to 60 K. The emission was collected through the same objective, dispersed by a double 1-m monochromator (gratings 2400/mm) and detected by a CCD camera. The spectral resolution of the setup was about 50 μeV.

Recombination dynamics of excitons in the CdTe QD's were studied through time-resolved PL by means of time-correlated single-photon counting. A frequency-doubled Ti: sapphire laser with 6-ps pulses and a repetition rate of 82 MHz at a wavelength of 370 nm was used as an excitation source. The signal was dispersed by a 0.25-m monochromator and detected by a fast microchannel plate photomultiplier tube. The overall temporal resolution of the setup was about 60 ps.

III. LOW-TEMPERATURE PHOTOLUMINESCENCE CHARACTERIZATION OF QUANTUM DOTS

In Fig. 2 we show PL spectra measured for both QD samples at $T=4.2$ K. In spite of the same material combination (CdTe/ZnTe) the spectra of these two QD structures are *qualitatively* different. The emission of the 2-ML CdTe QD sample (squares) features two peaks. The high-energy emission is narrower and more intense while the low-energy emission is much broader and somewhat weaker. In micro-PL spectra, when the size of the excitation spot is reduced down to 1 μm in diameter, the low-energy line splits into the series of ultranarrow individual emission lines with a typical linewidth of 0.1 meV.[31] Therefore, we assign the low-energy emission to exciton recombination in CdTe QD's. In contrast, the micro-PL spectrum of the high-energy line does not feature any structure, the overall shape is unchanged, and it remains smooth. We ascribe this high-energy line to the 2D WL emission. As shown in the following, for this QD structure we observe a strong thermally induced redistribution of carriers between QD's through this uniform WL.

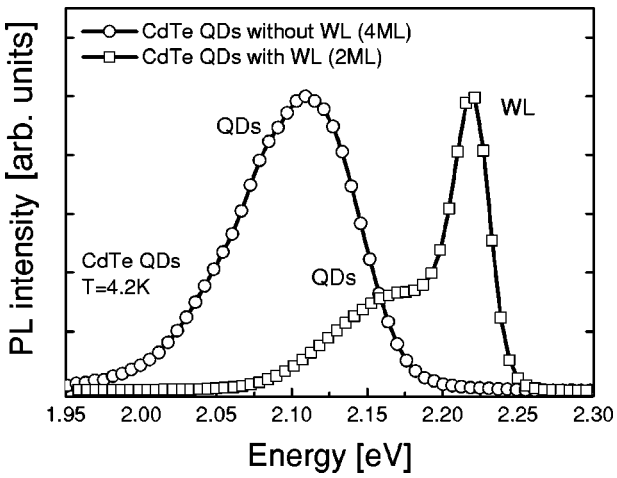

FIG. 2. Low-temperature PL spectra of the structures containing CdTe QD's with the nominal thickness of 2 ML (squares) and 4 ML (circles). The emission ascribed to QD's and to the WL is indicated.

On the other hand, for the 4-ML CdTe QD's the PL spectrum (circles) consists only of a single and relatively broad (~70 meV in the full width at half maximum) emission. Clearly, we observe no spectral signature of the well-resolved WL emission for this sample. In addition, the results of micro-PL show that this whole PL line is composed of ultranarrow emission lines ascribable to exciton recombination in single QD's.[31] This suggests that the 4-ML CdTe QD sample contains only fully developed QD's, which are not connected by the uniform 2D WL. Indeed, recent results of PL excitation spectroscopy performed on this sample have shown that there is no 2D continuum of states between QD emission and the ZnTe barrier.[32] The absence of the WL should prevent any lateral transfer of carriers between QD's, so this QD ensemble should behave as a set of individual *isolated* QD's. Indeed, the results of power- and temperature-dependent PL measurements presented in the next section support this interpretation.

IV. EXPERIMENTAL RESULTS: ENSEMBLE OF QUANTUM DOTS WITHOUT THE WETTING LAYER

In this section the PL properties of the 4-ML CdTe QD sample are discussed. Since no spectroscopic evidence of carrier redistribution is found for this sample, the obtained results are interpreted as a direct proof that this sample consists only of QD's that are *not connected* with a uniform 2D wetting layer. This conclusion is consistent with the scheme of the epitaxial growth of II-VI QD's presented above.

The results of continuous-wave PL obtained for the QD sample with a nominal CdTe thickness of 4 ML are summarized in Fig. 3, where we compare the temperature dependencies of the PL energy obtained for a single QD[31] and for the large QD ensemble. For the latter, the data measured at three different excitation powers are shown. Apart from experimentally determined energies (points) we also plot the line representing a temperature-induced shift of the CdTe (Ref. 33) band-gap energy. The relation $E(T)$, scaled in or-

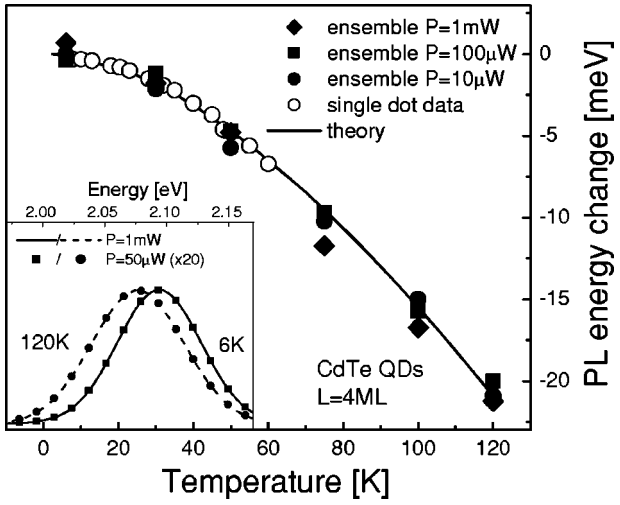

FIG. 3. Comparison between the temperature dependencies of the PL emission energy for a QD ensemble (solid points) measured at three different excitation powers and of single CdTe QD's (open circles). The line represents the CdTe energy band gap shrinkage induced by the temperature. In the inset the normalized PL spectra measured at 6 and 120 K for two values of the excitation power are shown. The lines (points) correspond to excitation power equal to 1 mW (0.05 mW, multiplied by 20).

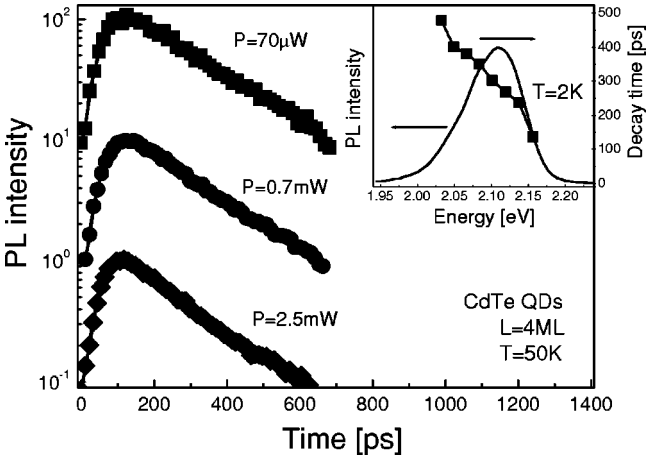

FIG. 4. Temporal behavior of the PL intensity measured at $T=50$ K for the CdTe QD's with the nominal thickness of 4 ML. The transients obtained for three different excitation powers are shown. In the inset the detection energy dependence of the exciton recombination time at $T=2$ K is presented together with the PL spectrum.

der to account for the quantum confinement in QD's, describes the experimental data very well. In the inset to Fig. 3 the actual PL spectra of the QD ensemble measured at 6 and 120 K normalized to their maximum intensities are presented. The solid lines (open symbols) correspond to excitation power of 1 mW (0.05 mW, multiplied by 20). As one can see, the shape of the PL emission does not depend on the excitation power even above 100 K. This observation shows that in this temperature range the exciton recombination occurs *only* from the dots that are initially occupied by both electron and hole and no transfer of the carriers between different QD's takes place. In other words, for all excitation powers and temperatures the measured PL spectrum reflects the energy distribution in the QD ensemble. The agreement between the PL energy of a single CdTe QD and that of the QD ensemble demonstrates further that the optical properties of the ensemble are determined by the properties of single and isolated QD's.

The results of time-resolved PL performed for this sample are presented in Fig. 4. As shown in the inset, at $T=2$ K the decay time of exciton recombination (τ_R) increases for lower emission energies, as observed in other QD systems,[34,35] probably due to acoustic-phonon-assisted tunneling between QD's with different energies.[36] The observed τ_R dependence on the emission energy could be also ascribed to the presence of excited states in the QD's. Indeed, as it has been shown for a number of QD systems, the decay time of the excited states is usually much faster than that of the ground state.[1] However, in the case of these QD's the PL excitation measurements of single QD's have revealed very intense and narrow lines ascribed to excited states in the energy range of about 120 meV.[32] Therefore, due to such a large energy splitting [larger than the linewidth of the QD emission (see Fig. 2)], we would rather exclude the excited states as the origin of the observed energy dependence of τ_R. For the results presented in this work, it is important to note that the value of τ_R measured for the maximum of the PL spectrum is equal to 300 ps and it is insensitive to the temperature up to around 100 K. Above this value, τ_R starts to decrease due to nonradiative recombination in QD's.[37] The dominant nonradiative mechanism responsible for the decrease of τ_R and for the reduction of the PL intensity with temperature is related to thermal activation of carriers into the ZnTe barrier. In fact, the activation energy determined from the intensity quenching with temperature equals 40 meV, which approximately corresponds to the heavy-hole confinement energy in these QD's.[38] Importantly, we also find that for the temperatures below 100 K, the decay time measured for the maximum of the PL emission does not depend on the excitation power. As an example in Fig. 4 we show temporal behavior of the PL intensity measured for the maximum of the PL emission at $T=50$ K for three different excitation powers. As for the energies and linewidths of the continuous-wave PL, this τ_R insensitivity to the excitation power is observed in the whole studied temperature range.

The experimental results obtained for this QD sample under both continuous-wave and pulsed excitations demonstrate unambiguously that the PL emission of the large QD ensemble is solely determined by the properties of individual and isolated QD's. We find no signature of the thermally activated carrier redistribution between different QD's, which we ascribe to the absence of a uniform 2D WL in this QD structure. We conclude, in agreement with the model of II-VI QD growth, that the sample with the 4-ML CdTe QD layer featuring single PL line (see Fig. 2) consists only of fully developed isolated QD's. The absence of the WL in this sample is further supported by recent PL excitation measurements of this QD sample where no signature of any 2D continuum of states has been observed.[32]

V. EXPERIMENTAL RESULTS: ENSEMBLE OF QUANTUM DOTS WITH THE WETTING LAYER

In this section the optical measurements performed for the QD sample with 2 ML nominal CdTe thickness are presented. In clear contrast to the previously discussed QD structure, the results of both continuous-wave and time-resolved PL experiments obtained for this sample show the presence of strong thermally induced carrier redistribution between different QD's. This intradot carrier transfer is possible via the 2D wetting layer that connects the dots in the ensemble. A theoretical model presented in this section fully supports this interpretation.

A. Continuous-wave photoluminescence

The PL measurements of the optical properties of CdTe QD's with a nominal CdTe thickness of 2 ML were carried out in two different regimes: the excitation power was varied at a fixed temperature, and the data were taken as a function of temperature with a constant excitation power. In Fig. 5 we show on a semilogarithmic plots the PL spectra taken at constant temperature of (a) 4.2 K, (b) 35 K, and (c) 70 K as a function of excitation power. At $T=4.2$ K the shape of the spectrum (its energy and linewidth) as well as the relative intensities between QD- and WL-related emissions remain unchanged at increasing excitation power. In contrast, at $T=35$ K, the emission of the QD's changes dramatically with power. The strong impact of the excitation power, not found for the sample with the 4-ML-thick CdTe QD layer, is observed already at $T=35$ K, but it is even more pronounced at $T=70$ K. We approximated the PL spectra with the sum of two Gaussian line shapes so that the power dependencies of both the energy and the linewidth for QD-and WL-related emissions could be obtained. In Fig. 6 the maximum of the PL emission [Fig. 6(a)] and the PL linewidth [Fig. 6(b)] is plotted as a function of excitation power for four temperatures. For $T=35$ K and $T=70$ K, as the excitation power decreases, the maximum of the QD emission shifts towards lower energies, and simultaneously, the PL line gets narrower. More specifically, at $T=70$ K the PL line ascribed to the QD emission narrows from 80 to 40 meV when the excitation power is reduced by almost four decades. At the same time, the energy of the maximum of the QD-related emission shifts by 80 meV. In contrast to the QD emission, both the emission energy and the PL linewidth of the 2D WL emission show essentially no change upon the excitation power [open points in Figs. 6(a) and 6(b)]. The behavior observed for the WL emission is identical to the results of similar measurements performed on strained CdTe/ZnTe quantum wells with thicknesses between 1 and 5 ML.[39] Interestingly, when the temperature increases to 100 K, the QD PL shows almost no dependence on the excitation power, similarly at $T=4.2$ K.

As can be seen in Figs. 5(b) and 5(c), the shape of the low-energy side of the QD emission is not affected by the excitation power; the slope of all spectra is identical. This shows that the excitation power dependence observed for this sample is a result of thermal activation of the carriers

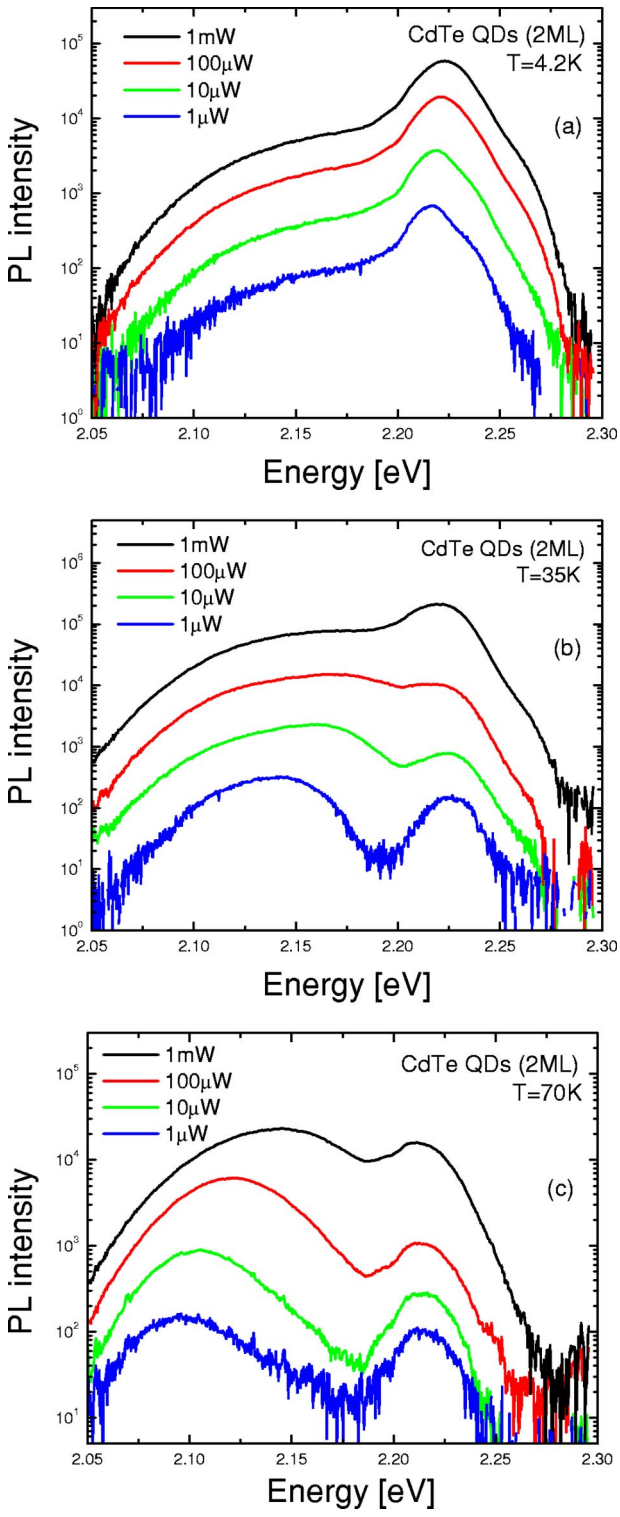

FIG. 5. Excitation-power-dependent PL spectra measured for the CdTe QD's connected with the WL. The measurements were carried out at (a) $T=4.2$ K, (b) $T=35$ K, and (c) $T=70$ K. The spectra are plotted on a semilogarithmic scale.

confined to QD's in the WL. Such a process occurs only if there exists a uniform 2D layer that connects a large number of QD's.

The impact of thermally induced carrier redistribution between QD's can also be observed in the temperature depen-

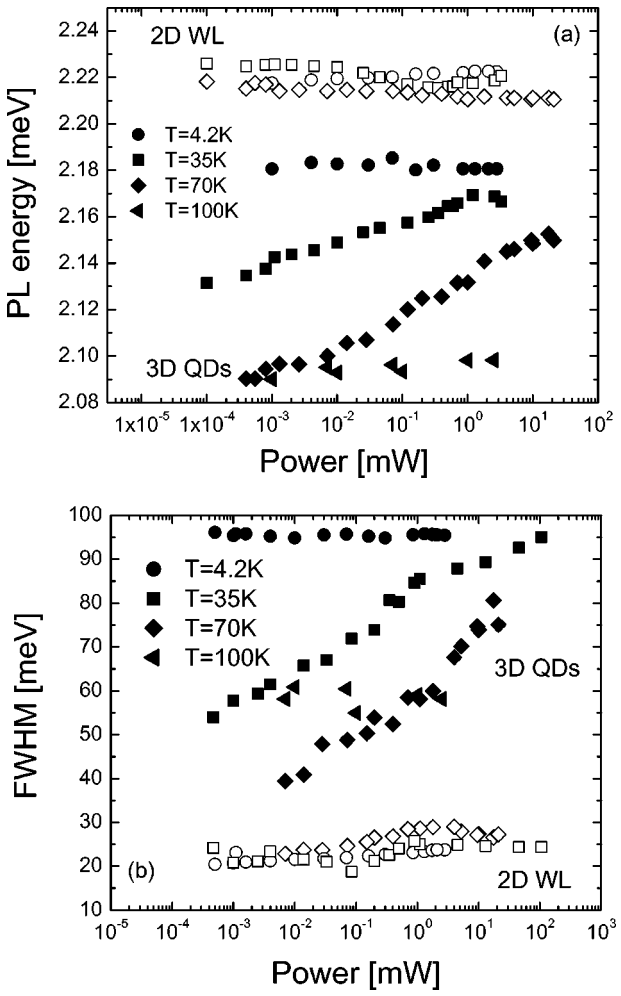

FIG. 6. (a) PL emission energy and (b) PL linewidth plotted as a function of the excitation power for both QD-related (solid points) and the WL-related (open points) emissions. The dependencies obtained at four temperature are shown.

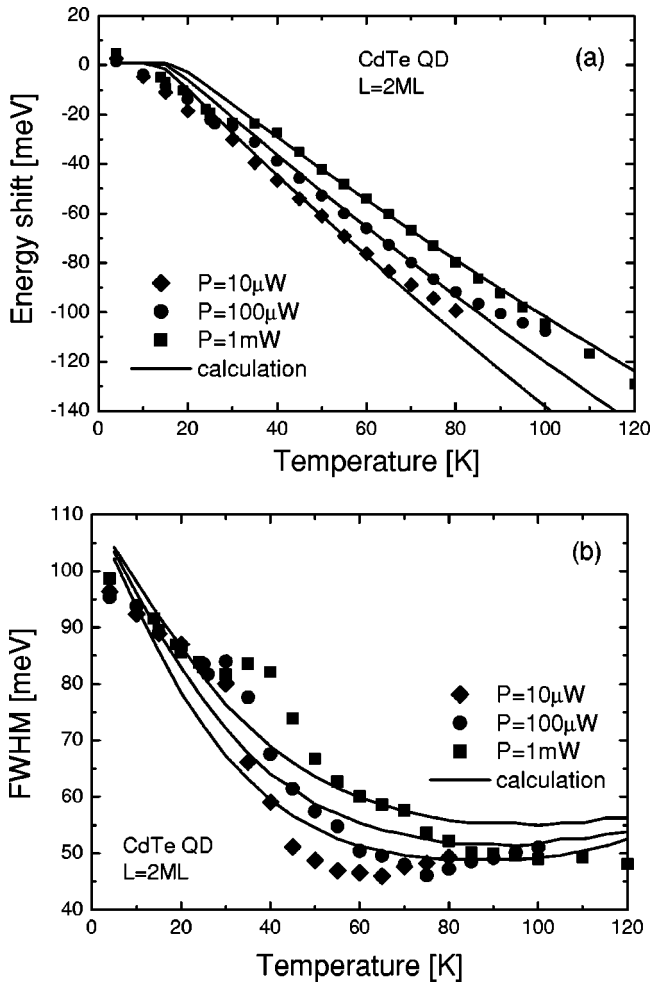

FIG. 7. (a) PL emission energy and (b) PL linewidth plotted as a function of the temperature for QD-related emission. The experimental values (solid points) were obtained for three different excitation powers, as indicated. The lines are the result of the calculation that takes into account the thermally induced carrier redistribution between QD's.

dence of the PL emission at a constant excitation power. In this case we find, similar to other reports,[22–25] that the PL energy of the QD ensemble decreases with the temperature much faster than that for single QD's (see Fig. 3).[31] In addition, the width of the PL line of the QD ensemble shows nonmonotonic behavior: it first decreases, goes through a minimum, and then starts to increase.[23] The results of these experiments are summarized in Fig. 7, where we plot (a) the PL energy and (b) the linewidth of the QD emission as a function of the temperature for several values of the excitation power. As shown in Fig. 7, we observe that the faster redshift of the PL energy and narrowing of the linewidth are enhanced for the weaker excitation power. Note that the position of the minimum in the temperature dependence of the PL linewidth [see Fig. 7(b)] occurs at a lower temperature for the weaker excitation.

The qualitative understanding of the continuous-wave PL results of this QD sample is schematically sketched in Fig. 8, where the occupation of QD potentials (short lines) is shown in six different excitation and temperature conditions. The circles correspond to the occupied QD states and the dashed stripe is the WL. The distance between this stripe and the short lines represents the energy spacing between the WL and the particular QD. The QD's are populated in time $T1$ according to the energy distribution within the ensemble. Then the redistribution of carriers may occur (at $T2$), which would change the QD occupation and therefore the subset of QD's that are observed in the emission (at $T3$).

At low temperature [Fig. 8(a)] regardless of the excitation power, the thermal activation of carriers from QD's to the WL is inhibited. The recombination takes place only in the dots that have been initially populated. Since, as we assume, the initial occupation is determined by the energy distribution of QD's, no excitation power dependence of either the PL energy or the linewidth is expected. Such a situation is indeed observed at $T=4.2$ K [see Fig. 5(a)]. As the temperature increases, the thermal activation of carriers to the WL becomes possible [Fig. 8(b)]. Then, the carriers localized by QD's with energies close to the WL energy may escape to the WL; these QD's would become empty at $T2$. If these thermally activated carriers do not recombine in the WL (either

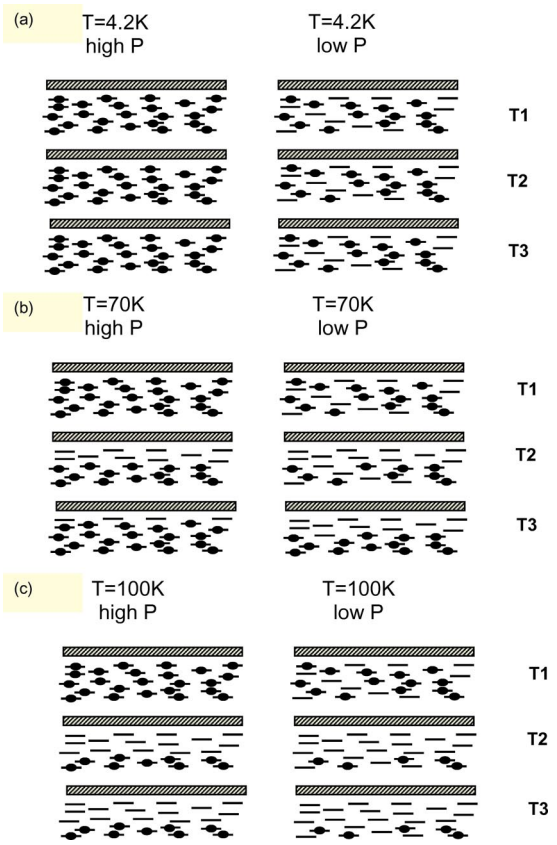

FIG. 8. Occupation of the ground states of QD's connected with the WL as a function of the excitation power at three different temperatures: (a) $T=4.2$ K, (b) $T=70$ K, and (c) $T=100$ K. The diagrams at the excitation ($T1$), redistribution ($T2$), and recombination ($T3$) times are shown.

radiatively or nonradiatively), they would be eventually captured by other dots where they recombine. In this regime, for high excitation power [Fig. 8(b), left panel] the change in the energy or the linewidth of the QD emission is small, since there are no available QD's states with low energies. In contrast, if the excitation power is low [Fig. 8(b), right panel], once the carriers are thermally activated to the WL, they are able to find a QD with much lower emission energy. As a result, for lower excitation power the recombination at time $T3$ occurs from QD's with narrower energy range. This simple picture explains the narrowing of the PL line and the large redshift of the maximum of the QD PL emission observed at $T=35$ K and $T=70$ K (see Fig. 6). The third regime, $T=100$ K, is shown in Fig. 8(c). Likewise, for the previous cases, at $T1$ the initial occupation of QD's reflects the energy distribution in the ensemble. Since the temperature is relatively high, more QD's become depopulated due to carrier thermal activation to the WL. Here, however, the recombination in the WL is much faster than the time required for these carriers to be recaptured by the QD's. As a result, they recombine (mostly nonradiatively[39]) in the WL. Under such circumstances, no significant influence of the excitation power on the PL linewidth and maximum emission energy is expected, as it is indeed observed experimentally (see Fig. 6).

B. Model

In order to describe quantitatively the results of continuous-wave PL experiments, we model the QD structure as an ensemble of strongly localized potentials connected with a WL characterized by a 2D continuum of states. This model is based on the one developed by Sanguinetti *et al.*;[24] it assumes the following:

(a) The capture of photocreated carriers from the ZnTe barrier into the QD's takes place only via the 2D WL. Analogously, we neglect a direct carrier escape out of QD's to the ZnTe barrier.

(b) The carriers in the WL could either be captured by QD's or they recombine radiatively or nonradiatively in the WL.

(c) Each QD can be populated by only one electron-hole pair, and the initial QD population is determined by the statistical distribution of QD ground-state energies within the ensemble. The energy distribution of QD's is identical to the PL spectrum measured at $T=4.2$ K.

(d) The radiative recombination time of excitons is the same for all QD's, and it does not depend on either size or shape (i.e., the emission energy) of QD's.

(e) The probability of thermal escape from QD to WL (γ) is proportional to $\exp(-\Delta E/k_B T)$, where ΔE is the difference between the energy of the QD and the energy of the WL. This value is determined experimentally and it is equal to 50 meV. It is also important to mention that it agrees perfectly with the activation energy obtained from the temperature dependence of the total intensity of QD emission.[29]

Under these assumptions, we obtain the following expressions for the number of carriers localized in the QD's (n_F) and in the WL (n_R), respectively, as normalized to the sample surface:[38]

$$\frac{dn_R}{dt} = g - n_R k_T + \int_{-\infty}^{0} n_F(E)\,\gamma(E) k_C n_W dE - \int_{-\infty}^{0} n_R[n_D(E) - n_F(E)] k_C dE = 0, \tag{1}$$

$$\frac{dn_F(E)}{dt} = -n_F(E) k_R - n_F(E)\,\gamma(E) k_C n_W + n_R[n_D(E) - n_F(E)] k_C = 0. \tag{2}$$

In these formulas g represents the generation rate for the carriers captured by the WL from the ZnTe barrier. Quantities $n_D(E)$ and $n_F(E)$ represent the density of all QD's and all occupied QD's, respectively, with energies between E and $E+dE$. k_R is the probability of the radiative recombination in the QD. The uniform 2D WL is characterized by the density of available states in the WL ($n_W \gg n_R$) and by the probability that the carrier in the WL is not captured by the QD potential (k_T). Importantly, both nonradiative and radiative processes in the WL are described by k_T. Finally, the redistribution of carriers between QD's is described by the prob-

ability that the carrier in the WL is captured by the single QD $k_C \propto |\int \Psi_{QW} \Psi_{QD} d^3r|^2$, and by the probability γ that the carrier in the dot is thermally activated into the WL.

From Eq. (2) one obtains the expression for the number of occupied QD's within the energy range $(E, E+dE)$:

$$n_F(E) = \frac{n_R n_D(E) k_C}{k_R + \gamma(E) k_C n_W + n_R k_C}. \tag{3}$$

Inserting this expression into Eq. (1), one obtains the following:

$$g - n_R k_T - n_R k_C n + \int_{-\infty}^{0} \frac{n_R k_C^2 n_D(E)[\gamma(E) n_W - n_R]}{k_R + n_R k_C + \gamma(E) n_W k_C} dE = 0. \tag{4}$$

In this calculation we describe the distribution of energy within the QD ensemble with the Gaussian function:

$$n_D(E) = \frac{n\sigma}{\sqrt{\pi}} e^{(E-E_0)^2 \sigma^2},$$

with the parameters E_0 and σ being directly determined from the low-temperature PL spectrum. In addition, the QD density and the recombination time in QD's are determined experimentally and are equal to $n = 10^4\ \mu\text{m}^{-2}$ and $\tau_R = k_R^{-1} = 300$ ps.[31] Similarly, as in Ref. 24, we set the value of $k_C = 800$ GHz. The only fitting parameters were the generation rate g and the density of states, n_w, in the WL.

The results of this fitting procedure are represented by the solid lines in Figs. 7(a) and 7(b). In the case of medium excitation power (100 μW) the values of $g = 10 k_R n_D$ and $n_W = 4.5 \times 10^{17}\ \text{cm}^{-2}$ were obtained. We note that the density of states in the WL, n_W, is of the same order of magnitude as reported in Ref. 24. Taking these values, the temperature dependencies of the PL energy and linewidth were computed for the excitation power equal to 10 μW and 1 mW. Despite the simplicity of the model, the calculated values agree well with the experimental results. The quantitative correspondence between the two data sets is observed for the energy dependence on the temperature [see Fig. 7(a)] up to $T = 70$ K, then at higher temperatures the dynamics of the system changes (recall the physical model at $T = 100$ K). On the other hand, the calculated behavior of the PL linewidth, although slightly different from the experimental results, still shows the major trends [see Fig. 7(b)]. In particular, the calculation reproduces the observation that the minimum of the PL linewidth occurs at lower temperatures for weakly excited QD ensembles. The small discrepancies may be related to the fact that we approximate all the PL spectra with Gaussian line shapes and that we have not included in the calculation the spatial dependence of the QD distribution (in the model all the dots are in the same place). It is also important to note that in the model we neglect the influence of the transitions between the bright and dark states in the QD's.[1] This stems from the fact that such a transition requires a spin flip of the exciton. Independent measurements of the spin relaxation time in this QD sample have shown that the exciton spin relaxation time [almost equal to 5 ns (Ref. 40)] in these QD's is by an order of magnitude longer than the exciton recombination time and the capture time [k_C^{-1} (see model)]. Therefore, the two latter processes determine the population of the QD's and any contribution from the spin-flip processes can be neglected.

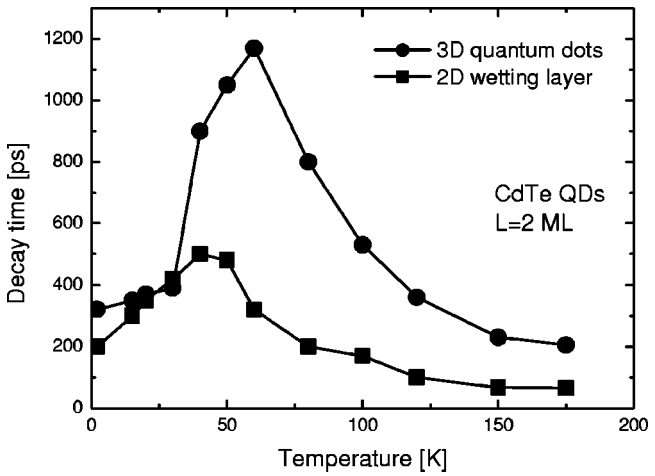

FIG. 9. Decay times of the exciton recombination measured as a function of the temperature for both QD-related (circles) and the WL-related (squares) emissions. The excitation power was kept constant through the whole temperature range.

In conclusion, the comparison between experimental and calculated results of the continuous-wave PL strongly supports the interpretation that the experimentally observed effects are mostly due to thermally induced redistribution of carriers between QD's via the uniform 2D wetting layer.

C. Time-resolved photoluminescence

The impact of carrier redistribution within the QD ensemble is also observed in the temporal behavior of the QD emission, as shown in Fig. 9, where the temperature dependence of the recombination time of the excitons confined to CdTe QD's is presented. Recall that in the case of isolated QD's (that are not connected with a 2D WL) the value of $\tau_R = 300$ ps was constant up to around 100 K. As can be seen in Fig. 9, for the CdTe QD's connected to the WL, τ_R shows strikingly different behavior.[35,41] While it seems to be roughly constant up to 30 K, above that temperature it increases and then reaches the maximum at $T = 60$ K. Further increase of the temperature results in a monotonic decrease of τ_R. We note that in the case of QD emission the decay time increases at the temperature range where the carrier redistribution starts to dominate the continuous-wave PL properties (see Fig. 7).

In order to understand this effect in more detail, we performed excitation power-dependent time-resolved PL measurements. In Fig. 10 we show the time evolution of the PL emission measured at (a) $T = 1.8$ K, (b) $T = 60$ K, and (c) $T = 100$ K for several excitation powers and for different detection energies corresponding to both QD's and WL emissions. The transient PL spectra, plotted in a semilogarithmic scale, are normalized so as to make the comparison more straightforward. In addition, in Fig. 11 we present the evaluated decay times together with the respective PL spectra. The

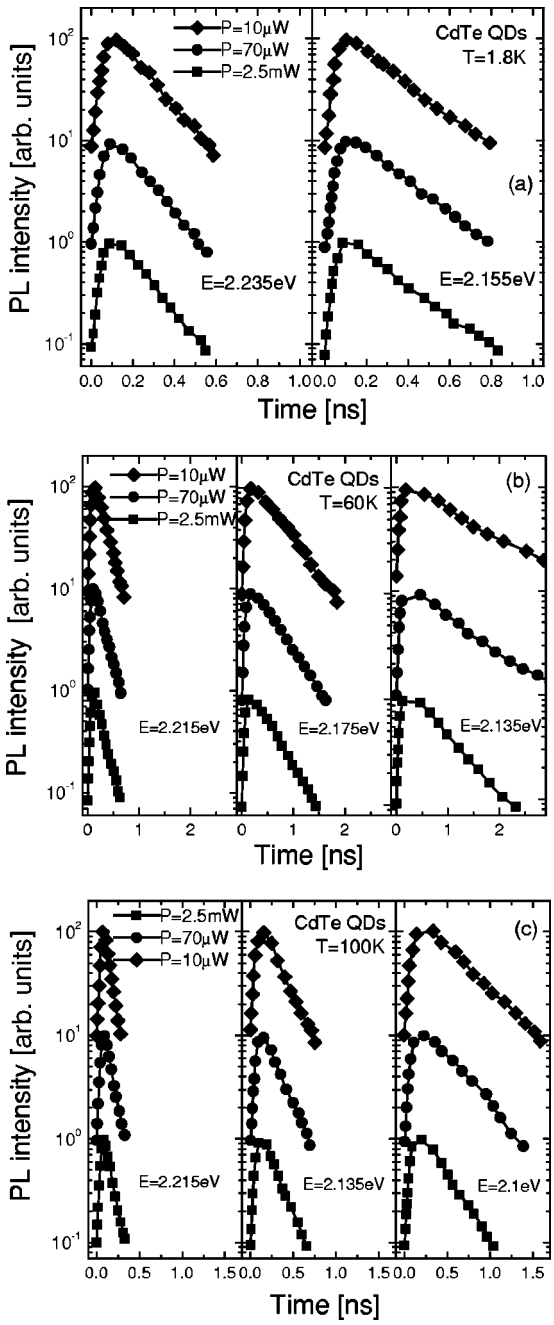

FIG. 10. Temporal behavior of the PL emission intensity measured at (a) $T=1.8$ K, (b) $T=60$ K and (c) $T=100$ K. The data were obtained for the detection energies corresponding to QD's and the WL emissions. In each case the transients were taken at several excitation power values.

spectra are normalized to the maximum of the WL emission. Noticeably, at $T=1.8$ K [Figs. 10(a) and 11(a)] the measured values of τ_R for both WL and QD emissions do not depend on the excitation power.

Distinctly different situation is seen when the temperature is raised up to 60 K [Figs. 10(b) and 11(b)]. In this case, τ_R of the QD emission strongly increases as the excitation power decreases. We point out that the increase of the decay time is present for the *whole* QD-related emission, although the largest enhancement is observed for the dots with the

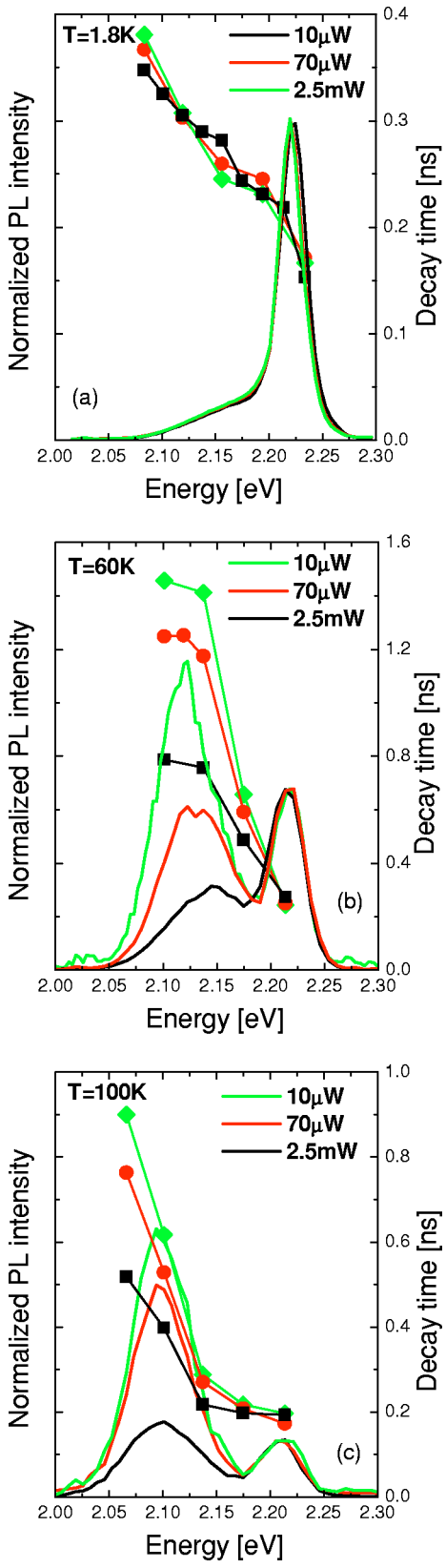

FIG. 11. Decay times of the exciton recombination evaluated from the transients shown in Fig. 10. In addition the PL spectra normalized to the maximum of the WL emission are shown.

lowest emission energies. At the same time, the value of τ_R measured for the WL is insensitive to the excitation power. Following the physical interpretation of the continuous-wave PL results (Fig. 8), we assign this strong τ_R dependence to the carrier redistribution between QD's through the WL. More specifically, the observed increase of τ_R for QD's is due to gradual ''feeding'' of QD's with the thermally activated carriers.

Importantly, the observation of thermally induced carrier redistribution requires that the time necessary for the activated carrier to find another dot is shorter than the nonradiative (or radiative) recombination time in the WL itself. The influence of the nonradiative processes in the WL on the optical properties of the QD's is indeed seen in Figs. 10(c) and 11(c), where we show the results of time-resolved PL obtained at $T=100$ K. Although still the interdot carrier redistribution via the WL induces some changes of the QD emission, the influence of this process is significantly reduced. We do not observe a strong redshift of the energy with decreasing excitation power nor narrowing PL linewidth. This shows that the energy distribution of the *luminescent* QD's does not depend on the excitation power. Therefore, the recombination in the WL prevents the recapture of the carriers that have been thermally activated from the QD's.

VI. CONCLUSIONS

To summarize, we have studied the impact of QD morphology on the carrier redistribution in the ensembles of self-assembled CdTe QD's. In the case of fully developed CdTe QD's that are not connected with a WL, the redistribution of carriers between QD's is inhibited. For this structure, where the nominal thickness of CdTe equals 4 ML, the PL properties of large QD ensembles reflect the behavior of single dots that are isolated from each other. We demonstrate that for the redistribution to occur, the presence of the uniform 2D WL is necessary, as observed for CdTe QD's formed from a CdTe layer with the thickness equal to 2 ML. The thermally induced carrier redistribution is demonstrated through detailed power- and temperature-dependent continuous-wave and time-resolved PL measurements. We find that at low temperatures, where the thermal energy of the carriers localized by the QD's is small, the initial population of the QD ensemble determines both the PL spectrum and the PL temporal behavior. As the temperature increases, the carriers are able to escape from the dots to the 2D WL. This process results in a significant narrowing of the PL linewidth as the excitation power decreases and in a strong redshift of the energy of the PL emission. Furthermore, we observe in a dramatic increase of the recombination time of the QD-localized excitons as the excitation power decreases. Finally, at high enough temperatures (seen around 100 K) the nonradiative recombination in the WL takes over, and the thermally activated carriers do not have enough time to find the QD before they recombine in the WL. The experimental results presented in this work demonstrate the significant influence of the morphology of the particular QD structure on its optical properties, specifically on the carrier dynamics within a QD ensemble.

*Author to whom correspondence should be addressed. Electronic address: seb@physics.uc.edu

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