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Influence of the temperature on the carrier capture into self-assembled InAs/GaAs quantum dots

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Photoluminescence (PL) spectroscopy and atomic-force microscopy (AFM) were used to investigate the size evolution of InAs quantum dots on GaAs(001) as a function of the amount of InAs material. Different families of islands were observed in the AFM images and unambiguously identified in the PL spectra, together with the signal of the wetting layer. PL measurements carried out at low and intermediate temperatures showed a thermal carrier redistribution among dots belonging to different families. The physical origin of this behavior is explained in terms of the different temperature dependence of the carrier-capture rate into the quantum dots. At high temperatures, an enhancement of the total PL-integrated intensity of the largest-sized quantum dots was attributed to the increase of diffusivity of the photogenerated carriers inside the wetting layer. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1568538]

I. INTRODUCTION

Self-assembled InAs/GaAs quantum dots (QDs) have been intensively investigated because they represent interesting systems for fundamental physics and became quite important to the development of devices.[1] As far as technological aspects are concerned, QD lasers made from InAs/GaAs heterostructures have now characteristics as good as quantum-well (QW) based devices.[2] These QDs are spontaneously formed during the epitaxial growth of strained layers and consist of small three-dimensional (3D) islands of strained material that start to develop on top of a two-dimensional (2D) wetting layer (WL) above a critical thickness d_c as a consequence of the accumulation of elastic energy in the epitaxial layer. The morphological structure of the InAs layer is mainly determined by the amount of InAs material (d_{InAs}) deposited on the surface,[3] but also depends on other growth parameters like the growth temperature (T_G),[4,5] V/III flux ratio,[6] InAs deposition rate,[7] as well as the thickness, temperature, and growth technique of the GaAs cap layer.[8,9]

Although several works have already been devoted to studying the fundamental properties of quantum dots, there is no consensus about the mechanisms that govern the radiative recombination processes of photogenerated carriers in temperature-dependent photoluminescence (PL) measurements. An unusual temperature dependence of the emission energy, of the full width at half maximum, and of the integrated intensity of the emission peaks related to the radiative recombination of confined carriers in QDs have already been reported.[10–15] The PL-intensity quenching with increasing temperature is commonly attributed to nonradiative recombination centers or to the thermal escape of carriers from the QDs to the wetting layer and/or GaAs barriers.[10–12] An unusual enhancement of the PL signal coming from the QDs was attributed to the gradual release of electrons from traps as the temperature was increased.[13] The tunneling of carriers between dots has also been invoked to explain the unusual reduction of the PL linewidth with increasing temperature and to justify the faster redshift of the QDs' emission energy with respect to the band gap of the bulk material constituting the QDs.[5,14]

In the present work, we carried out PL and atomic-force microscopy (AFM) measurements on QD samples grown with different InAs coverages but using a special structure that allowed the investigation of the optical and structural properties of the QDs in the same sample. We observed that quasi-three-dimensional islands were present below the usual critical thickness d_c and acted as the precursors of the usual QDs, as already observed by Heitz *et al.*[3] Such structures confine carriers and give rise to bound states with an energy lower than the one of the wetting layer. They can be detected in the PL spectra, together with the emission coming from the WL and the other families of QDs. We also pointed out the influence of the temperature on the capture rate of the photogenerated carriers by the quantum dots and clearly evidenced the carrier diffusion inside the wetting layer.

II. EXPERIMENTAL DETAILS

All the samples were grown in a Gen II MBE system on top of an epi-ready GaAs(001) substrate that was fixed on a molybdenum block with liquid indium. The structure consisted of a 0.25-μm-thick GaAs buffer followed by an InAs layer with a (nominal) thickness d_{InAs}, a 0.10-μm-thick GaAs barrier and, on the top, a second InAs layer identical to the first one. After the growth of the GaAs buffer at 570 °C, the substrate temperature was increased to 610 °C to smooth

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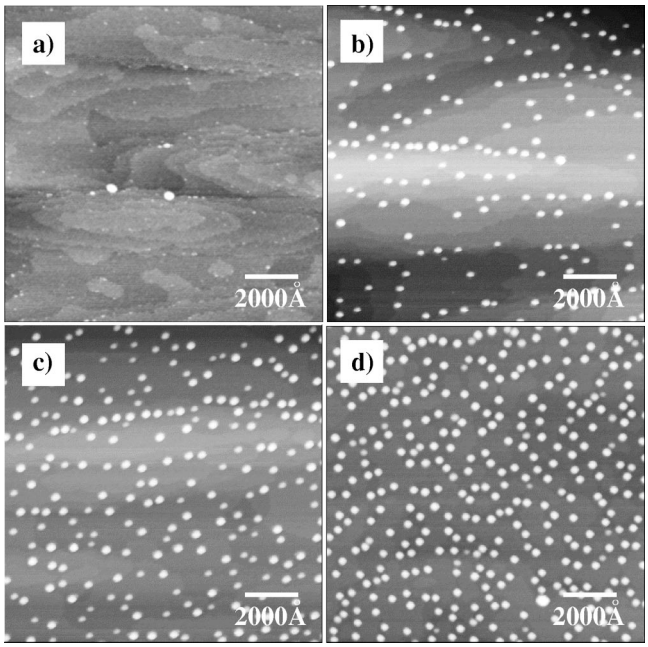

FIG. 1. 1×1 μm^2 AFM images showing the surface morphology of the top InAs layer for the samples with a coverage of (a) 1.50, (b) 1.75, (c) 2.00, and (d) 2.50 ML. The total gray scale of the respective images is 100, 200, 300, and 300 Å.

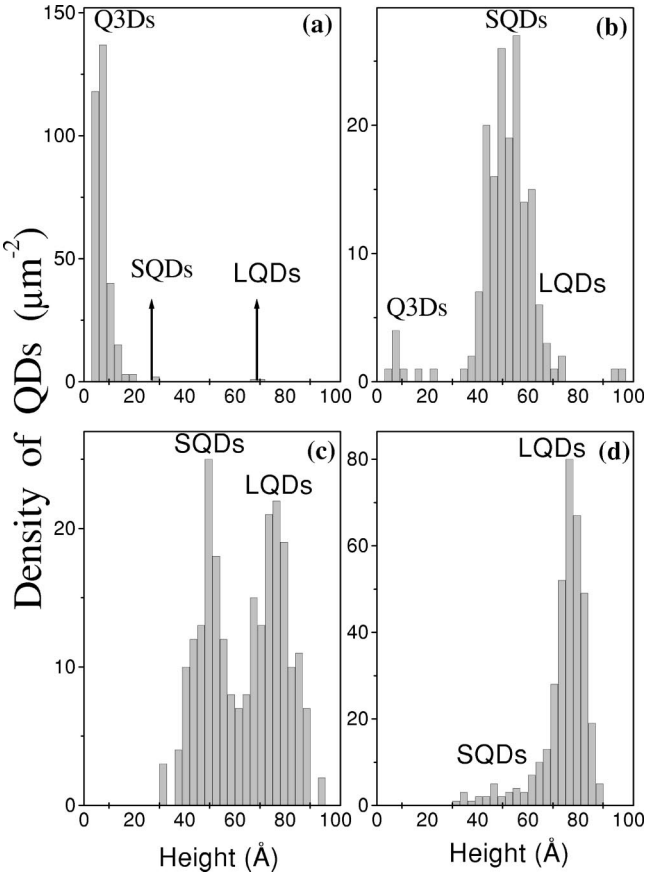

FIG. 2. Height histograms of the structures detected in the respective images of Fig. 1. The different types of families are indicated by SQDs (small QDs), LQDs (large QDs) and Q3Ds (quasi-3D clusters).

the surface and lowered to 510 °C in order to deposit the first InAs layer. The first 100 Å of the second GaAs barrier were grown at 510 °C and then the temperature was increased to 570 °C to grow the rest of the barrier (900 Å). The smoothing procedure of the GaAs surface was realized once again before the growth of the second InAs layer. A growth rate of 0.1 monolayer per second (ML/s) and 1.0 ML/s were used for the InAs and GaAs material, respectively, together with an arsenic beam-flux equivalent pressure of 7.5×10^{-6} Torr. A set of five samples was grown with a different value of d_{InAs} from sample to sample: 1.00, 1.50, 1.75, 2.00, and 2.50 ML. In the rest of the text, each sample will be designated by the nominal thickness d_{InAs} of its InAs layer.

The structural data of the quantum dots (size distribution and density) were obtained in ambient conditions with a Nanoscope IIIa AFM, from Digital, operating in contact mode with a sharpened tip located at the apex of a "V"-shaped silicon-nitride cantilever. Optimized feedback parameters and a weak interaction between the tip and the sample led to high-quality and reliable images that allowed the investigation of the QD morphology as a function of the InAs-film thickness.[16] The PL measurements were performed in an optical cryostat operating with liquid helium from 1.4 to 300 K. The samples were excited with the 5145 Å line of an argon laser at a power density of 100 W/cm^2. The luminescence signal was analyzed by a monochromator and detected by a Ge detector cooled with liquid nitrogen.

III. QUANTUM-DOT SIZE DISTRIBUTION

Figure 1 shows the AFM images recorded on the top InAs layer of four samples of the set and Fig. 2 reports the height distributions of the QDs in the respective images of Fig. 1. For the sample with $d_{\mathrm{InAs}}=1.00$ ML, the images (not shown here) showed that there were no QDs, as expected. With further InAs deposition ($d_{\mathrm{InAs}}=1.50$ ML), an accurate analysis of the image and size distribution revealed the presence of three different types of structures. The largest QDs, with an average base $\overline{b}=338$ Å, average height $\overline{h}=69$ Å, and areal density $\overline{D_a}=2$ μm^{-2} will be referred to here as *large* QDs (LQDs). A second family of QDs with the same density as the previous one, but with a smaller size ($\overline{b}=200$ Å, $\overline{h}=28$ Å) was denominated as *small* QDs (SQDs). Although both types of QDs can not be called a family at the current stage of their evolution, we shall show briefly that they will turn into two distinct families in the samples with a larger InAs thickness. In the $d_{\mathrm{InAs}}=1.50$ ML sample, these two families coexist with a much larger density ($\overline{D_a}=316$ μm^{-2}) of even smaller structures ($\overline{b}=157$ Å, $\overline{h}=7$ Å) with a height of 2–4 ML that will be called quasi-3D (Q3D) clusters, in analogy with the terminology adopted in Ref. 3.

For the samples with a larger amount of InAs material, the AFM images pointed out an increase of the density of the SQDs and LQDs and a decrease of the density of the Q3D clusters, indicating that the latter act as the precursors of the two families of larger QDs, as already observed by Heitz *et al.*[3] Indeed, in our AFM images the total QDs' density in the last sample ($d_{\mathrm{InAs}}=2.50$ ML) is roughly equal to the density of Q3D structures in sample $d_{\mathrm{InAs}}=1.50$ ML. The

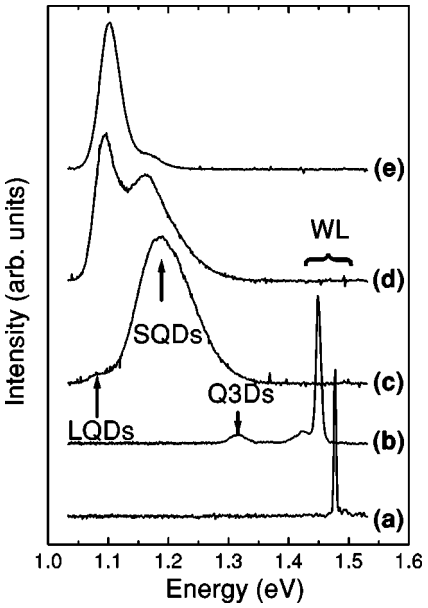

FIG. 3. 2 K PL spectra of all the samples of the set with an InAs coverage of (a) 1.00, (b) 1.50, (c) 1.75, (d) 2.00, and (e) 2.50 ML. The samples were excited with a power density of 100 W/cm^2 and a photon energy of 2.41 eV.

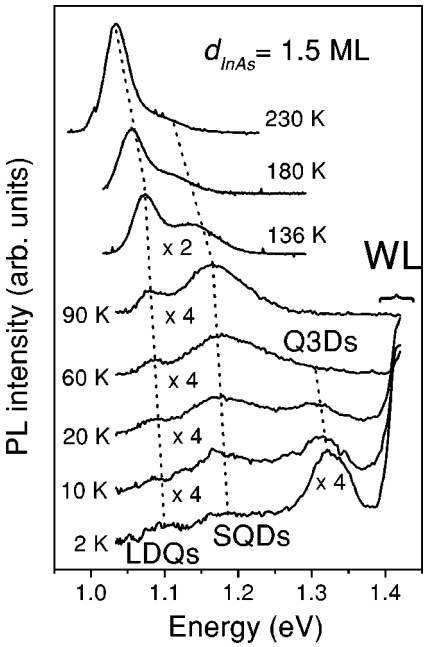

FIG. 4. Temperature-dependent PL spectra of the sample with an InAs coverage of 1.50 ML. The measurements were carried out with a power density of 10 W/cm^2 and a photon energy of 2.41 eV. The dotted lines connecting the peaks are only a guide to the eyes.

sequence of height histograms clearly shows that, initially, the Q3D structures are transformed into SQDs that, with further InAs deposition, turn into LQDs. For $d_{\mathrm{InAs}}=2.00$ ML, the two families of SQDs and LQDs coexist, as shown in Fig. 2(c), and differ only by the mean height of their members. Since the LQDs already reached their maximum allowable size, due to strain-energy considerations, they can not grow further and their size does not change during the whole evolution of the QDs until their coalescence, as already pointed out by da Silva *et al.*[16] In the last stages of their evolution ($d_{\mathrm{InAs}}=2.5$ ML), there is a significant increase of the density of the LQDs at the expenses of the SQDs as can be seen in Fig. 2(d).

Although the QDs on the bottom InAs layer are morphologically different from the ones of the top layer (because of the extra GaAs cap layer), we expect their evolution to be similar.[17] We shall show in Sec. IV that all the features of the top QD layer observed in the AFM images can be identified in the PL spectra of the buried structures as well.

IV. PL MEASUREMENTS

A PL spectrum recorded at $T=2$ K is presented in Fig. 3 for each sample of the set. The narrow peak at 1.476 eV [spectrum (a), $d_{\mathrm{InAs}}=1.00$ ML], which evolves into a wider peak at 1.450 eV with increasing InAs deposition [spectrum (b), $d_{\mathrm{InAs}}=1.50$ ML] is attributed to the recombination of the heavy-hole exciton in the WL. The redshift of the WL peak is associated with the increase of the mean width of the 2D layer with further InAs coverage. The PL spectrum of the sample with 1.50 ML of InAs also revealed two emissions around 1.42 and 1.32 eV. The nature of the 1.42 eV peak was recently ascribed in the literature to carrier localization inside In-rich islands located at the second interface of an InGaAs QW (i.e., of the InAs WL in our case) as a consequence of In segregation.[18] We attribute the 1.32 eV peak to the emission from the Q3D clusters observed in the AFM images. This peak was previously observed by Heitz *et al.*[3] in QD samples with 1.15 and 1.5 ML of InAs and also related to the presence of Q3D clusters. Since in this type of structure the confinement along the vertical direction is much stronger than laterally, they are usually considered as very thin quantum wells (with a width of 2–4 ML) whose excitonic emissions are expected to appear below the emission of the WL (also considered as a QW with a width of 1 ML) due to their larger height.

For the samples $d_{\mathrm{InAs}}\geqslant 1.75$ ML, the emissions related to the WL and the Q3D structures are no longer observed [see spectra (c)–(e)]. Increasing the InAs coverage ($d_{\mathrm{InAs}}=2.00$ ML), the emissions from the SQDs and LQDs [(spectrum (d)] have comparable intensities due to their similar areal density [Fig. 2(c)]. When the density of the SQDs decreases with further InAs coverage ($d_{\mathrm{InAs}}=2.50$ ML), only the emission of the LQDs is observed [spectrum (e)] since the areal density of the LQDs is several times larger than that of the SQDs.

PL measurements on the sample $d_{\mathrm{InAs}}=1.50$ ML as a function of temperature exhibited a very interesting behavior and will be discussed now. The emissions related to the LQDs and SQDs were clearly observed together with the signal coming from the WL and Q3D islands when the optical measurements were performed as a function of temperature with a lower excitation power ($P_{\mathrm{exc}}=10$ W/cm^2) than in Fig. 3. These PL spectra are shown in Fig. 4 where it is possible to observe that the peaks related to the LQDs and SQDs are already well defined and appear in the same spectral region as the ones observed in the samples with a higher InAs coverage. Only the tail of the intense WL peak is shown in order to better visualize the weaker emissions com-

ing from the other structures. We can see that, with increasing temperature, there is a quenching of the Q3D clusters and WL emissions, meanwhile the emissions from the LQDs and SQDs become the most pronounced features. Although the areal density of the LQDs and SQDs are similar in this sample ($\overline{D}_a = 2\ \mu m^{-2}$) and much smaller than the one of the Q3D clusters, in the temperature range $20 \leqslant T \leqslant 136$ K the peak ascribed to the SQD emissions is more intense than the other ones, whereas beyond $T = 136$ K the LQD peak becomes dominant in the PL spectra.

It is clear that, when the temperature is increased, there is a gradual transfer of the PL intensity from the high-energy to the lower-energy peaks, indicating that the largest-sized QDs become the main recombination path. This behavior reflects the influence of temperature on the capture rate of the photogenerated carriers by the quantum dots and can be explained as follows. The capture process consists of two essential parts: the transport of carriers (a real spatial transfer of carrier) to the close proximity of a QD and their energy relaxation from the fundamental state of the WL to the lowest discrete energy level in the InAs QD. If the incident radiation has an energy larger than the band gap of the GaAs material, essentially all the carriers are generated into the thick GaAs barriers surrounding the InAs layers. However, we observe that the emission from the thin WL is several orders of magnitude larger than the GaAs emission, meaning that there is a strong diffusion of the carriers from the GaAs barriers to the WL. Once inside the WL, the excess energy of the carriers is released either by a fast (few ps) emission of optical phonons or by a much slower emission of acoustic phonons.[19] During this relaxation time, the carriers are able to move freely over a long distance within the WL, will most probably pass in the near vicinity of a strong capturing center (a QD or a Q3D structure) and, in order to be trapped, will be obliged to relax in energy. Since the main temperature-dependent contribution to the capture process is due to phonon scattering, the capture probability by the QDs will be inversely proportional to the number of phonons involved in the relaxation mechanism. Thus, at low temperature, the larger QDs will have a lower capture rate due to the larger energy difference between the onset of the WL and the QD transition energies. Indeed, time-resolved PL studies showed that larger InAs QDs have a much larger rise time (156 ps)[20] for the ground state luminescence than small QDs (20 ps),[21] showing the faster capture rate of the carriers from the WL into the smaller QDs. Although these data were obtained in different experimental conditions, a large QD will always have a longer rise time than a smaller QD when they are simultaneously probed in the same sample (as we did here) because the rise time is mostly related to the carrier diffusion within the WL and to the energy relaxation toward the lowest discrete level of the QD. Since the latter phenomenon is a multiple-phonon process, a large QD will need more phonons than a smaller one because the difference between the WL energy level and the lowest level of the QD is larger.

With increasing temperature, there is a competition between the energy loss of the carriers by phonon emission and the energy gain by phonon absorption. So, for the same reasons that led the smaller QDs to have a higher capture efficiency at low temperature, there will now be a stronger escape of the carriers from the smallest QDs with increasing temperature. In this way, when the sample temperature is raised, the capture process of carriers by the smallest structures (Q3D islands) is no longer efficient, yielding a gradual transfer of the PL intensity from the highest-energy peaks to the lower-energy ones, as observed in our optical data. The fact that the integral of the PL intensity remains almost constant from 2 to 136 K together with the observation that, at high temperature, the LQD peak becomes dominant in the PL spectra (despite the very low areal density of those structures) confirm that the transfer of carriers between different families of QDs originates mainly from the different temperature dependency of the carrier-capture probability of the confining structures, instead of other thermally activated processes. For instance, it is important to point out that, in this sample, the carrier tunneling between individual quantum dots does have a minor contribution to the transfer process because the average distance between neighboring QDs was estimated to be about 600 Å. In such conditions, resonant and assisted interdot tunneling (by static defects or lattice vibrations) become very ineffective due to the small overlap between the envelope functions of spatially distant dots.[22]

For temperatures above 136 K, an anomalous increase of the integrated PL intensity by a factor of 4 was observed instead of the usual reduction of the optical signal resulting from the nonradiative transitions that are more pronounced at high temperatures. This peculiar behavior is a direct manifestation of the diffusion enhancement of the photogenerated carriers inside the WL. At such temperatures, the escape of the carriers out of the Q3D structures is so strong that they can only recombine through the QDs. Although the areal density of those structures is very low, the carriers can now diffuse over very long distances and encounter new LQD and SQD structures that were not accessible at lower temperature. This enhancement of the effective diffusivity of the carriers in the WL with temperature thus yields an increase of the PL intensity of the largest QDs.

Yoon *et al.*[23] showed that an increase of the carrier diffusion in QWs could be obtained by increasing the lattice temperature or the energy of the photon used to excite the sample. Thus, a simple way to confirm that the anomalous temperature behavior of our PL data is due to an enhancement of the carrier motion in the WL is to excite the sample with a laser radiation whose energy is quasiresonant with the WL emission in order to minimize the kinetic energy of the carriers injected in the sample. In such experimental conditions, the thermalization time will be very small and the carriers will not be able to diffuse over long distances before being captured. The results of such an experiment are shown in Fig. 5 where it can be seen that the emissions of the LQDs and SQDs do not become the dominant features in the PL spectra as a function of temperature, unlike in Fig. 4. Since the carriers are excited with a very low kinetic energy, they are almost immobile and no strong enhancement of the diffusion takes place in the WL. In this way, a real spatial transfer of carriers to the vicinity of remote LQDs and SQDs is suppressed. For structures with a low density of quantum dots, Wang *et al.*[24] estimated a very large capture time of 350

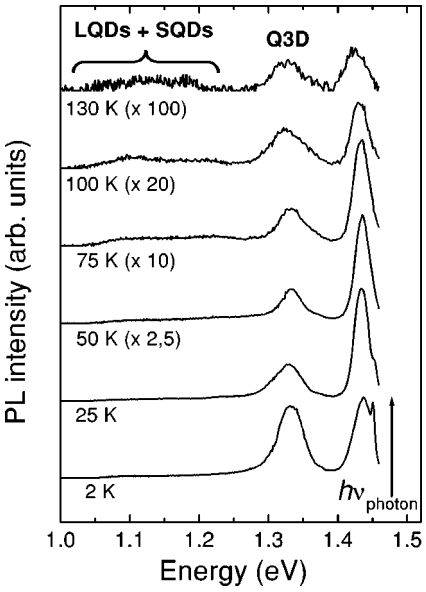

FIG. 5. Temperature-dependent PL spectra of the sample with an InAs coverage of 1.50 ML. In this case, the measurements were carried out with a power density of 10 W/cm^2 and a photon energy of 1.48 eV generated by a titanium–sapphire laser in order to be quasiresonant with the WL emission.

ps due to the dominance of the carrier transport from the barriers toward the QDs. In our case, this limitation is relieved by increasing the temperature.

In the PL spectra of the 1.75 and 2.00 ML samples, we also observed a (weaker) transfer of intensity from a high energy peak to a lower-energy peak with increasing temperature, but this phenomenon was related to the (weaker) carrier transfer from the SQDs to the LQDs, because the density of Q3D islands was very low [Fig. 2(b)] or even absent [Fig. 2(c)] in those samples, as can be seen in the distribution histogram. The 2.50 ML sample did not show this effect because only LQDs were mainly present [Fig. 2(d)]. Since in the 1.75 and 2.00 ML samples the Q3D structures were almost absent, the transfer from the SQDs to the LQDs occurred without any strong enhancement of the integrated PL intensity because both types of structures have comparable areal densities and because their magnitude for the electron–hole wave function overlap integrals are probably similar. Consequently, although the increase of the temperature leads to a larger carrier diffusion in the WL for all the samples, its manifestation was clearer in the 1.50 ML since, due to their very low density, the LQD and SQD families act as very sensible sensors of any enhancement of the effective diffusivity of the carriers in the WL.

V. CONCLUSIONS

In this work we carried out PL and AFM measurements on QD samples grown with different InAs coverages. We showed that all the morphological structures revealed by AFM spectroscopy were also evidenced by the PL measurements. By studying the temperature dependence of the PL emissions we were able to observe a redistribution of carriers among different dots' families that occurred through their transfer inside the WL. We observed that the efficiency of the capture (or emission) process into (out of) the different QD structures determined the behavior of the QD optical emissions: in the $2 \leqslant T \leqslant 136$ K range, carriers confined within the smaller structures are thermally activated to the WL, diffuse laterally, and then are retrapped by the lower-energy states of the larger quantum dots. We also showed that the increase of the total integrated PL intensity for $T > 136$ K was due to the enhanced lateral diffusion of carriers to empty neighboring dots out of the area illuminated by the pumping laser.

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