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Carrier dynamics in stacked InP/GaAs quantum dots

A. B. Veloso, M. K. K. Nakaema, M. P. F. de Godoy, J. M. J. Lopes,[a] F. Iikawa,[b]
M. J. S. P. Brasil, J. R. R. Bortoleto, and M. A. Cotta
Intituto de Física "Gleb Wataghin," Unicamp, Campinas-Sao Paulo, Sao Paulo 13083-970, Brazil

P. F. P. Fichtner and M. Morschbächer
Instituto de Física, UFRGS, Porto Alegre-RS, Brazil

J. R. Madureira
Faculdade de Ciências Integradas do Pontal, UFU, Ituiutaba-MG, Brazil

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We investigated two stacked layers of InP/GaAs type-II quantum dots by transmission electron microscopy and optical spectroscopy. The results reveal that InP quantum dots formed in two quantum dot layers are more uniform than those from a single layer structure. The thermal activation energies as well as the photoluminescence decays are rather independent of the separation between quantum dot layers and the presence of the second layer. The quantum dot optical emission persists for thermal activation energy larger than the calculated exciton binding energy. The photoluminescence decay is relatively fast for type-II alignment. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2789705]

Quantum dots (QDs) are very attractive systems for fundamental studies and optoelectronic applications due to their atomiclike discrete states. It has been shown that piling up QD layers separated by a thin barrier induces QD vertical self-organization along the growth direction[1–3] and improves the dots size uniformity of the second QD layer.[4,5] Structures composed of stacked QDs with type-I interface band alignment have been intensively investigated but few works have been reported for type-II systems, such as Ge/Si,[6,7] GaSb/GaAs,[8–10] and InP/GaAs.[11–13] In these structures, only one type of carrier is confined in the QD whereas the other carrier remains in the barrier layer surrounding the dot. The Coulomb attraction and the overlap of the carriers' wavefunctions are thus significantly reduced for type-II systems. This should also result in longer recombination times for QDs with type-II alignment, which is specially appropriated for some applications such as optical memories. With that in mind, understanding carrier dynamics becomes an essential issue to stablish type-II stacked dots as an interesting material system for such applications.

In this work we investigated the carrier dynamics and the electronic coupling between QDs in a series of samples with two InP/GaAs QD layers using a single QD layer structure as a reference sample. It is believed that the electron in our structures is confined in the InP QD, while the hole remains in the GaAs layer and is only bound to the dot due to the Coulomb attraction to the confined electron. In this case, the weakly bound hole should be easily ionized with increasing temperatures. We obtained, however, rather large thermal activation energies from our measurements as compared to calculated values. Furthermore, the results are rather independent of the separation between the two stacked QD layers. Another unexpected result concerns the measured optical emission decay times, which are quite small for type-II structures and comparable to type-I ones. Our results could actually be more straightforwardly explained if the InP/GaAs system had a type-I interface. This hypothesis is, however, in disagreement with the large blueshift of the photoluminescence peak from a QD structure versus the excitation intensity,[12,13] which is a typical property of type-II systems. We discuss alternative explanations for our results considering a type-II alignment.

Our structures were epitaxially grown by chemical beam epitaxy on GaAs (001) substrates. The thickness of the GaAs spacer between the two InP QD layers was varied from $d = 3$ to 12 nm. All structures were covered with a 50 nm GaAs capping layer. Photoluminescence (PL) measurements were performed using a He–Ne laser and a S-1 photomultiplier, in the cw regime, and a picosecond Ti-sapphire laser and a streak camera for time-resolved (TR) measurements. Cross-sectional transmission electron microscopy (TEM) images were obtained using a Jeol JEM-2010 microscope operating at 200 kV.

Cross-sectional TEM dark-field (002) images from three samples with distinct space layer thicknesses are shown in Fig. 1. The white areas correspond to the InP QDs. We observe a tendency of vertical alignment even when the QDs are separated by a 12 nm GaAs layer. We can also observe that the dots from the second layer tend to be slightly larger than those of the first layer, which is attributed to the faster nucleation of dots induced by the strain field produced by the QDs from the first layer, which is also responsible to the QDs alignment.

Figure 2(a) presents a series of PL spectra at 2 K from our structures. The peak energy shifts to lower energies as the GaAs spacer thicknesses decrease, which is expected considering the coupling of the electron wave functions confined in nearby dots at distinct QD layers.[3,4] Electron tunneling between those nearby dots may also play a role in the observed redshift. Emission from the smaller dot of a pair of nearby dots becomes less likely if the tunneling time between those dots becomes comparable to the recombination time, favoring the occupation of the larger dot and thus the lower energy emission.

[a]Present Address: Institute for Bio- and Nanosystems (IBN1-IT), Research Center Jülich, Germany.
[b]Electronic mail: iikawa@ifi.unicamp.br

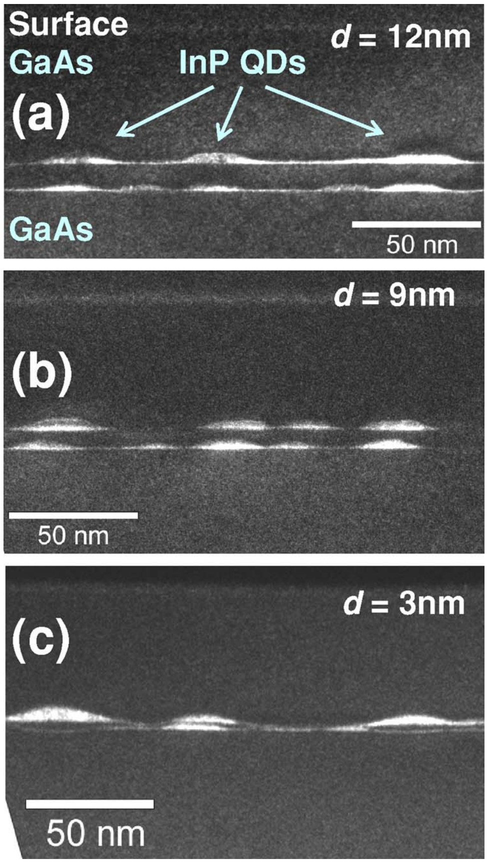

FIG. 1. (Color online) Cross-sectional TEM dark-field (002) images of the stacked InP QDs with different space layer thickness. (a) 12 nm. (b) 9 nm. (c) 3 nm.

We observe that the PL band from the single layer sample is relatively broader and remarkably more asymmetric than the stacked structures. The PL band narrowing can be interpreted as an evidence of the expected increasing of uniformity for stacked QD structures.[3–5,14] The template of strain produced by the QDs of the first layer that induces the

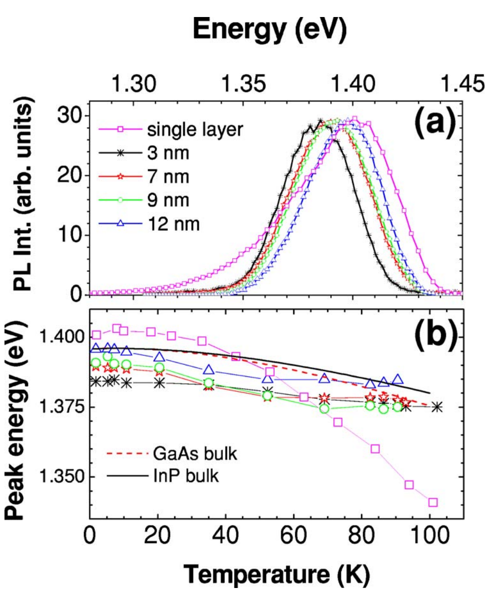

FIG. 2. (Color online) (a) 2 K PL spectra of single and two InP/GaAs QDs layers with d=3, 7, 9, and 12 nm. (b) PL peak position energy vs temperature. The continuous (dotted) lines show the temperature variation of the gap for InP (GaAs).

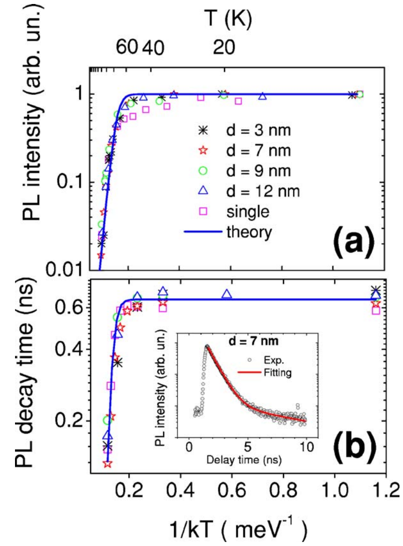

FIG. 3. (Color online) (a) Integrated PL intensity vs inverse of the temperature, where k is Boltzmann's constant. (b) PL decay time vs inverse of the temperature. The inset shows the transient of the integrated PL intensity.

growth of the QDs in the second layer contributes to the simultaneity of the QDs nucleation and, therefore, the size spreading of the QDs.

We also observe very distinct behaviors for the single and stacked structures with increasing temperature. In Fig. 2(b) we present the energy of the PL peak versus temperature. For the stacked structure, the QD emission energy basically follows the thermal dependence of InP and GaAs band gaps (solid and dashed curves), which are rather similar. In contrast, the energy variation for the single layer sample is significantly more pronounced. At 100 K the PL peak energy from all stacked structures is ~1.38 eV, while for the single layer sample, the peak is at ~1.34 eV, which corresponds to the low energy emission tail only observed for this sample at 2 K. The strong redshift of the single layer sample is actually rather typical for QD structures and is usually attributed to a thermal excitation effect.[15] With increasing temperatures, electron localized at the smaller dots (higher energies) are thermally activated to the wetting layer, where they may migrate to larger dots (smaller energies), resulting in a redshift of the emission band. The fact that the stacked QDs do not present a strong redshift indicates that the very large QDs, which are observed in the single layer structure, have a reduced probability to be occupied due to the larger density of the averaged size QDs of the second layer.

The Arrhenius-like plot of the integrated PL intensity is shown in Fig. 3(a). All stacked QDs including the single layer sample present very similar behavior. The resulting thermal activation energy for all samples is E_a $\sim 68 \pm 20$ meV. In contrast, the calculated exciton binding energy for InP/GaAs QDs, which basically corresponds to the hole-binding energy by the Coulombic potential, varies from 6 to 9 meV depending slightly on the dot size. Therefore, E_a must be related to some other thermal excitation process. In fact E_a is close to the energy separation (~60 meV) between the QD and the wetting layer (WL) emission bands.[11] This process should thus correspond to the excitation of electrons from the QDs to the WL. However, as we do not observe an increasing of the WL emission with the temperature, those carriers should have a large probability of being excited to the GaAs barriers. We point out that, considering the WL emission energy, the excitation of electrons from the WL to the GaAs should have a similar activation energy. The fact that the Arrhenius plot is not dominated by the lower activation energy corresponding to the hole-binding energy can be interpreted as an indication that this process is not effective on annihilating the recombination from the QDs. As the QD exciton is broken, the remaining electron must have a large probability of recapturing another hole and still recombine radiatively due to the long range Coulomb interaction. In fact, relatively large activation energies have also been observed for other type-II QD structures such as Ge/Si (~83 meV) (Ref. 7) and GaSb/GaAs (130 meV),[8] which indicates that this must be a general property of type-II systems.

The decay times (τ) from the QD emission obtained by the TR-PL measurements are presented in Fig. 3(b) as function of the inverse of the temperature. All samples present rather similar behavior. At low temperatures the PL decay times are ~650 ps. This is much smaller than the radiative recombination time theoretically estimated for our system, which should be of the order or larger than 10 ns. As the temperature is increased, τ markedly decreases following a typical thermal dependent function given by $\tau(T)=\tau_0/[1+(\tau_0/\tau_{\text{eff}})e^{-(E_a/kT)}]$, where τ_0 is the low temperature decay time, τ_{eff} is the effective scattering time, and E_a is the thermal activation energy. Fitting this equation to our results we obtain $E_a \sim 67 \pm 20$ meV for all samples, which is in agreement with the result obtained by cw-PL measurements. This result suggests that both the carrier lifetime and the cw-PL intensity are limited at high temperatures by the thermal activation of electrons to the GaAs barriers through the WL.

The fact that the recombination time from our structures is remarkably small even at low temperatures is incompatible with a type-II alignment that should imply in a relatively small electron-hole wave function overlap. This unusual result can be related to the capture of carriers by fast trap centers associated to impurities or defects or other effects such as intermixing and strain that may somehow result in a strong localization of the holes close to the interfaces. Large decay times, consistent with type-II interfaces, have been obtained for GaSb/GaAs type-II QDs.[9,10] In those works, however, the QD structure was surrounded by layers that act as potential barriers for the weakly bound carrier, reducing its wave-function spreading. In contrast, in our case, the hole wave function should be largely spread and, therefore, resulting in a large probability of overlap with trap centers.

Additional effects, such as intermixing and strain may also affect the analyzed optical properties. There is not much information concerning those effects in InP/GaAs systems and further investigations are still necessary. The fact that we have not observed any variation of the measured values on our set of samples indicates, however, that the variation of those effects as a function of the separation of the two QD layers is not the major factor that determines their optical properties. This can be due to the large ensemble of QDs probed in our measurements or to a distinct general property shared by all QDs that controls the measured quantities.

In summary, we investigated the carrier dynamics in stacked InP/GaAs quantum dots. The vertical alignment reflects in an increasing of QD size uniformity. We observed that the QD optical emission persists for thermal energies larger than the exciton binding energy and presents a rather short lifetime considering type-II alignment. Both results may be related to the weak Coulombic binding of holes that result in a largely spread wave function for holes in this system.

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