Effective phonon bottleneck in the carrier thermalization of InAs/GaAs quantum dots

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We present a detailed study of the dependence of the spectral characteristics of quasiresonantly excited InAs/GaAs quantum dots on electronic structure, temperature, and hydrogen content. Multiphonon resonances dominate the resonantly excited emission spectra. Such multiphonon resonances are attributed to the competition between two different channels ruling carrier thermalization in an ensemble of quantum dots: decay between polaronic states and efficient nonradiative intradot decay. This supports an effective "phonon bottleneck" in quantum dots, which is only partially relaxed by the presence of a fast polaron thermalization channel.

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I. INTRODUCTION

The nature of the mechanisms underlying carrier relaxation phenomena in quantum dots (QDs) has been largely debated in the last years.[1–9] These phenomena continue to attract much attention since they involve fundamental physical aspects of the zero-dimensional semiconductor systems as well as aspects of carrier dynamics, which are relevant for device applications. However, there is no general consensus on how an electron-hole pair injected in the continuum of a dot relaxes its energy to reach the QD ground state. The use of models well established in bulk and quantum well structures, namely, carrier energy relaxation by irreversible emission of LO phonons, has led to the concept of phonon bottleneck.[1] This shows up whenever energy spacing between levels of order of few meVs inhibits an efficient relaxation channel via LO phonons and carriers relax via the much less efficient channel of acoustical phonons emission. However, general evidence of important phonon bottleneck effects has never been found. As a matter of fact, QDs usually show fast relaxation times (of order of 30–70 ps even in the low injection regimes[10–12]). Only in a few cases of large carrier confinement an inefficient energy relaxation[6] has been accounted for in terms of relaxation through a continuum tail of defect states[4,13] and/or by resonant multiphonon processes between localized states.[3] Verzelen *et al.* in Ref. 14 pointed out that the notion of irreversible phonon emission is not applicable to QDs: A small one-phonon continuum and a strong Fröhlich interaction disable the Fermi golden rule in QDs, where polarons should be the elementary excitations involved in the energy relaxation of photoexcited carriers[8,14–16] as recently claimed in InAs/GaAs QDs.[17] From the experimental point of view, carrier relaxation in a system with an atomiclike density of states (DOS) determines photoluminescence (PL), quasiresonant photoluminescence (RPL), and photoluminescence excitation (PLE) spectra in QDs. When large ensembles of self-organized QDs are resonantly excited, RPL emission spectra are given by relatively narrow resonances, whose widths are smaller than the original QD PL bands,[18,19] while symmetrical excitation resonances dominate PLE spectra above the detection energy.[3,19] An extensive study of the dependence of the resonance energy shift, both in RPL and PLE, on the QD size and shape, led to the attribution[19] of those resonances to QD excited-state transitions. This attribution can be questioned in cases where resonances have been observed that differ from the ground state by multiples of the LO-phonon energy, irrespective of QD size.[3,9,20] These resonances have been explained, instead, in terms of phonon replicas in the framework of the excitonic polaron model by Heitz *et al.* in Ref. 9. The excitonic polaron interpretation,[9,20–22] which was successfully applied to explain the CdSe PL spectra,[23] requires a rather large (for InGaAs materials) electron-phonon interaction and a ground to excited-state energy separation close to the LO-phonon energy.[22] However, multiphonon resonances in PLE have been explained in the literature also in a phonon bottleneck framework where multiphonon relaxation competes with efficient nonradiative recombination channels in the carrier thermalization processes.[3,18] This approach would justify why multiphonon resonances are not always observed in QDs where the content of defects responsible for nonradiative channels may largely vary from sample to sample.[24] In the present work, a detailed study of the dependence of resonantly excited PL spectra of InAs/GaAs QDs on electronic structure and temperature shows that multiphonon resonances dominate the RPL spectra in InAs/GaAs QDs where the defect activity is varied by hydrogen implantation. Atomic hydrogen has been widely used for passivating electronic levels induced by point and extended defects in semiconductors.[25,26] Thus, the hydrogen driven defect activity tuning, due to the effective passivation of strain induced defects in the QDs,[27] gives us a unique tool to settle the

origin of the multiphonon resonances in RPL spectra. In fact, phonon bottleneck and polaronic model predictions strongly differ on the role of the nonradiative exciton recombination probability. We demonstrate that in the QDs under study the multiphonon resonances have to be attributed to competition between carrier thermalization to the ground state and an efficient nonradiative intradot decay channel in the phonon bottleneck framework.[18]

II. SAMPLE GROWTH AND EXPERIMENTAL DETAILS

We have prepared two samples in a molecular beam epitaxy (MBE) Varian Gen-II modular system using a beam equivalent pressure ratio As/Ga≈60. The substrates were radiatively heated and the growth temperature (T_g) was monitored by an optical pyrometer operating in a band where GaAs is opaque. The structures consist of a semi-insulating (100)±0.10° GaAs substrate, a 100 nm thick GaAs buffer grown at 600 °C, and InAs deposits grown at 500 °C with a coverage of 2.0 monolayers (MLs) (QDA) and 2.4 MLs (QDB). The InAs growth rate was 0.011 ML/s. At the end of the buffer growth, a growth interruption of 210 s was performed to change and stabilize the substrate temperature for the growth of the InAs dots. A very sharp (2×4) reflection high-energy electron diffraction (RHEED) pattern was observed after this stage, which indicates a very smooth surface. A second 210 s growth interruption was performed before growing the GaAs cap in order to lower T_g down to 360 °C. At this temperature 20 nm of GaAs were grown by a variant of MBE (atomic layer MBE—ALMBE) where group-III and group-V species impinge on the substrate alternatively in submonolayer amounts per cycle[28] in order to minimize the interactions between InAs and GaAs. This procedure has been shown to give high quality structures.

Hydrogenation of the samples was obtained by ion-beam irradiation from a Kaufman source with the samples held at 300 °C. The H dose was optimized in order to give the highest increase of the PL intensity at $T=11$ K. The optimal H dose was $d_H=2\times10^{17}$ ions/cm^2 for QDA and $d_H=5\times10^{17}$ ions/cm^2 for QDB, giving an increase of the PL intensity of 12 times for QDA and 5 times for QDB, respectively.

CW PL spectra were measured in the 10–270 K T range. The PL was excited with a duplicated Nd doped yttrium aluminum garnet (Nd:YAG) laser ($E_{exc}=2.330$ eV) for excitation in the barrier [nonresonant PL excitation (NPL) conditions] or with Nd:YAG laser ($E_{exc}=1.165$ eV) for excitation resonant with the QD excited states (RPL conditions). Maximum power density was about 100 W/cm^2 for NPL and 400 W/cm^2 for RPL, respectively. PL spectra were measured by a grating monochromator operating with a Peltier-cooled InGaAs photodiode; the spectral resolution was 3.2 nm. A holographic notch filter at 1064 nm (10 nm bandpass) was used in RPL measurements.

III. EXPERIMENTAL RESULTS

The low-temperature PL spectra of the samples are shown in Fig. 1. At low laser excitation power density ($P_{exc}=1$ W/cm^2) the two samples show the usual QD inhomogeneously broadened PL spectra. QDA emission [Fig. 1(a)] peaks at 1.093 eV with a full width at half maximum (FWHM) of 77 meV. QDB PL band [Fig. 1(b)] shows its maximum at 1.058 eV with a FWHM of 55 meV. When increasing P_{exc} by a factor 100, state filling effects take place and the PL spectra show emission also from the QD excited states. At high P_{exc} a single excited state, centered at $E_{emi}=1.174$ eV, is clearly resolvable in QDA PL [Fig. 1(c)] while QDB shows emission from the first ($E_{emi}=1.125$ eV) and the second ($E_{emi}=1.192$ eV) QD excited states [Fig. 1(d)]. Therefore, the two QD samples have different electronic structures. The ground to excited-state energy difference is 81 meV in QDA and 67 meV in QDB. The second excited state observed in QDB is 134 meV away from the ground state.

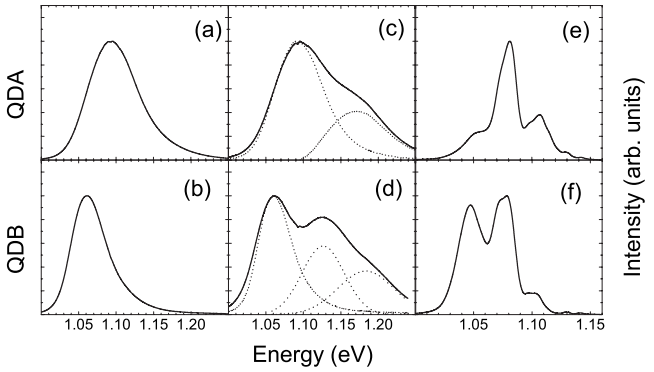

FIG. 1. Upper panels: PL spectra of QDA under NPL conditions at (a) $P_{exc}=1$ W/cm^2 and at (c) $P_{exc}=100$ W/cm^2 and under RPL conditions at (e) $P_{exc}=20$ W/cm^2. Lower panels: PL spectra of QDB under NPL conditions at (b) $P_{exc}=1$ W/cm^2 and at (d) $P_{exc}=100$ W/cm^2 and under RPL conditions at (f) $P_{exc}=20$ W/cm^2. In panels (c) and (d) dotted lines show the decomposition of the PL spectra in terms of Gaussian contributions.

The PL spectra are highly modified by resonant excitation [Figs. 1(e) and 1(f)]. The QD emission in RPL conditions is characterized by a multimodal spectrum whose components are the same in both QDA and QDB, much narrower (FWHM≈20 meV) than the original QD PL band and roughly equispaced in energy. Three modes at 1.047, 1.078, and 1.108 eV can be clearly resolved in the spectra of both samples. However, the relative intensity of the three modes varies among the two QDs: QDA [Fig. 1(e)] is dominated by the resonance at 1.078 eV, with the other two modes reduced to weak sideband at the lower and higher energy sides. On the contrary, in QDB [Fig. 1(f)] the emission is dominated by the two resonances peaked at 1.047 and 1.078 eV.

The temperature dependence of the PL spectrum of QDA is reported in Fig. 2. Under NPL conditions [Fig. 2(a)] the spectra show the T dependence typical of an ensemble of QDs.[29] With increasing temperature, the PL band maximum shifts to lower energy faster than the InAs band gap, the PL peak narrows and reaches a FWHM of 60 meV at $T=220$ K, and the PL yield decreases. The latter T dependence (not reported here) has been fitted using the model reported in Ref. 29. It has been found that the PL quenching mechanism can be accounted for by a thermoionic emission

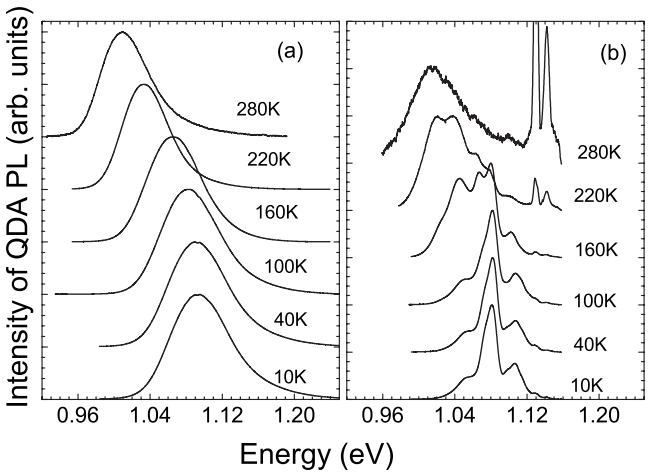

FIG. 2. Temperature evolution of the PL spectrum of QDA under NPL conditions (panel a) and RPL conditions (panel b).

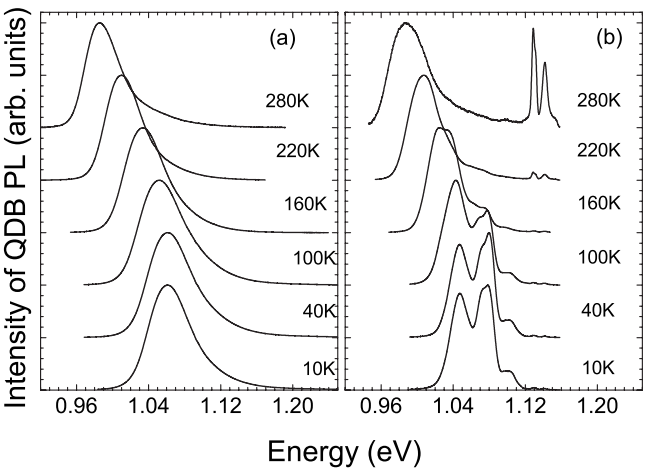

FIG. 3. Temperature evolution of the PL spectrum of QDB under NPL conditions (panel a) and RPL conditions (panel b).

of carriers from QD states toward states located at 1.38 ± 0.01 eV, most likely wetting-layer states from where those carriers recombine nonradiatively.[29,30]

The behavior under RPL conditions [Fig. 2(b)] is quite different. The low-T spectrum does not change roughly up to 100 K, where only minor changes in the relative weight of the low and high energy sidebands respect to the central line are observed and, most important, the emission peak does not shift to lower energy. Above 100 K, the emission spectrum remains multimodal but the relative weight of the low-T components changes and new lines appear on the low-energy side of the spectrum. For further increasing temperatures, the multimodal character of the RPL spectrum looses strength and the emission approaches the form of the NPL one, while it maintains the fingerprint of a multimodal emission in the form of a slight line-shape modulation. At high T the multimodal characteristics of the RPL spectrum are completely lost and the RPL spectrum resembles the NPL one. Please note that, for the fixed excitation wavelength conditions typical of RPL spectra, the absorption coefficient is modified for increasing temperatures because of the energy-gap redshift, thus preventing any consideration upon the PL yield. The temperature dependence of the QDB spectrum, shown in Fig. 3, is quite similar to that of QDA. Under NPL [Fig. 3(a)] conditions, the PL band rapidly redshifts, narrows (FWHM $=44$ meV at $T=250$ K), and quenches with an activation energy compatible with thermoionic carrier emission from QD into wetting-layer states. The RPL spectra [Fig. 3(b)] are independent on temperature up to 100 K. Above such temperature, the relative weight of the PL spectral modes changes and new modes appear on the low-energy side of the emission band. For increasing temperature, the RPL spectrum approaches the NPL one and at high T the two spectra can be hardly distinguished.

Two sharp lines at 1.141 and 1.129 eV, whose intensity is only slightly affected by temperature, are present in all the spectra of QDA and QDB. These two sharp lines with an energy difference, from the excitation laser, of $\Delta E = E_{\mathrm{emi}} - E_{\mathrm{exc}} \approx 25$ meV and $\Delta E \approx 35$ meV, behave differently from all the other resonances in the spectra: (i) their FWHM is resolution limited (≈ 4 meV), while the other resonances share a FWHM of ≈ 20 meV; (ii) the intensity depends only slightly on T, on the contrary of the other resonances which undergo to a substantial quenching (about 3 orders of magnitude) when T reaches room temperature. We attribute the sharp line at $\Delta E \approx 35$ meV to the Raman scattering from GaAs bulk phonons ($E_{\mathrm{LO}} = 36.6$ meV). The sharp line at $\Delta E \approx 25$ meV has been already reported in the literature, but its attribution is still questionable.[19]

IV. DISCUSSION

The observation of a RPL signal requires a two-step process. First, an exciton is generated in an excited QD state by photon absorption and, second, the exciton relaxes to the ground state where it radiatively recombines. Schematizing the process, the dependence on the excitation energy (E_{exc}) and on the emission energy (E) of the RPL spectrum is given by

$$I_{\mathrm{RPL}}(E_{\mathrm{exc}};E) \propto P_{\mathrm{ab}}(E_{\mathrm{exc}};E) P_{\mathrm{rel}}(E_{\mathrm{exc}} - E;E), \qquad (1)$$

where P_{ab} and P_{rel} are the probabilities of absorption and relaxation processes, respectively.[3,19] P_{ab} is proportional to the number of QDs with an excited state at E_{exc} and the ground-state energy at E. P_{rel} is the probability of carrier relaxation from the QD excited state to the QD ground state. The interlevel relaxation processes which lead to exciton thermalization have to compete with alternative nonradiative decay channels.[18] If interlevel relaxation is faster than the nonradiative recombination process, the exciton rapidly thermalize to the QD ground state, meaning a roughly energy independent P_{rel}. The RPL spectrum will be then determined by $P_{\mathrm{ab}}(E_{\mathrm{exc}};E)$,[19] showing inhomogeneously broadened emission bands in correspondence of the ground-state energies of the QDs that have an excited state at E_{exc}.[19] In the opposite limit, when exciton recombination is dominated by nonradiative decay processes, I_{RPL} is mostly determined by the energy dependence of P_{rel}. In this case, if phonon bottleneck plays a fundamental role, RPL spectrum is expected to be characterized by the equally spaced multiplets of the LO-

phonon energies which modulate P_{ab}.[3,18] It's worth noting that we are dealing with QD ensembles, where, due to the intrinsic broad size and shape distributions, $P_{ab}(E_{exc};E)$ is a highly inhomogeneously broadened function of E_{exc} and E.

In addition to the excitation-relaxation related processes, a modulated RPL spectrum is also expected in the case of strong electron-phonon interaction. This interaction leads to a coupled electron-phonon system, the polaron, with eigenstates $\Psi \equiv |\alpha, n_{ph}\rangle$, where $\alpha=0$ labels the system ground state (with no excitons), $\alpha=1$ indicates one exciton in the QD ground state, $\alpha=2$ one exciton in the QD first-excited state, and so on. n_{ph} indicates the number of LO phonons involved in the polaron state.[14–16] Within the polaron theory and in a Frank-Condon framework, RPL spectra of QDs should show multiphonon resonances accounted for in terms of zero-phonon lines and phonon replicas.[23] In this model,[21,23] k-phonon resonances in RPL spectra are due to the absorption of a photon, which causes the transition from the $|0,0\rangle$ state to a $|\beta, m_{ph}\rangle$ excited state, followed by the emission of a photon in the transition from the $|1,0\rangle$ to the $|0,(m+k)_{ph}\rangle$ state (polarons, indeed, are supposed to rapidly thermalize from the $|\beta, m_{ph}\rangle$ to the $|1,0\rangle$ state). The probability of photon absorption and emission is calculated in the framework of a revisited Huang-Rhys (HR) theory, which includes nonadiabatic effects[20,22] and is able to accurately reproduce the resonances in the RPL spectrum of CdSe QDs.[23] The polaronic model description of the RPL spectra modulation, being an intrinsic effect due to the large electron-phonon interaction, is not expected to depend on the relative importance of nonradiative decay channels in the QDs.

The RPL spectra of both samples, reported in Fig. 1, show a clear multimodal emission with equally spaced peaks ($\Delta E=30$ meV). These peaks are located at the same energies in the two samples in spite of the different QD electronic structures. The origin of the modes in the emission of QD ensembles under RPL condition has been widely discussed in the literature. As already stated before, the resonances in RPL spectra could be attributed to phonon-related processes[18,22] as well as to excited-state resonances.[19] LO-phonon energy ranges[18] from 29.0 to 36.6 meV in (InGa)As heterostructures and does not change from sample to sample. Therefore, as pointed out in Ref. 19, phonon related resonances should be observed only in well-defined energy windows, which are the same in all samples and do not depend on excitation energy. On the contrary, if the resonance energies in the RPL spectra sizably vary when the electronic structure of the QD is changed, or when the excitation energy is varied, the observed peaks in the emission must be attributed to excited-state transitions.[19,31] In other words, a quantum size effect should be present when the resonances in the RPL spectra stem from excited-state transitions. As a matter of fact, QDA and QDB do not share the same electronic structure, differing in both ground state and excited-state energies. Nevertheless, RPL resonances appear at the same energies in both samples and their energy spacing is compatible with the LO-phonon energies in (InGa)As heterostructures, thus showing the phonon origin of these resonances. The absence of a quantum size effect in the RPL spectra of QDA and QDB is further confirmed by the lack of correlation between the energy position of the RPL modes and the temperature [see Figs. 2(b) and 3(b)].[19] As a matter of fact, the QD-DOS redshifts for increasing temperature, with a difference between low-T ($T=10$ K) and room-T QD ground-state energies of ≈ 70 meV.[32] Therefore, QDs with different sizes are investigated by changing temperature at fixed E_{exc} (which is equivalent to vary E_{exc} at fixed temperature, as it is more usually done).

As already reported in Sec. I, two different explanations have been provided in the literature as regards the origin of the phonon related resonances observed in RPL spectra: (i) multiphonon relaxation processes[3,18] and (ii) phonon-assisted absorption/emission within the nonadiabatic Huang-Rhys theory.[20–23] In the following, we will verify the ability of each of these two models to describe the observed phenomenology. Multiphonon resonances in QD RPL spectra excited with an Nd:YAG laser at 1064 nm ($E_{exc}=1.165$ eV) were reported by Heitz *et al.* in Ref. 18. In close analogy to the case of hot exciton relaxation in bulk semiconductors or quantum wells, the equally spaced resonances in RPL (and PLE) (Ref. 3) spectra were interpreted in terms of fast, incoherent, multiphonon relaxation between excitonic states in the QDs in competition with a nonradiative intradot channel. On this ground, excitons efficiently thermalize to the QD ground state only when the excited-to-ground-state relaxation is faster than the intradot nonradiative recombination channel. In turns, this happens only when the excited-to-ground-state energy difference matches a multiple of the LO-phonon energy.[3] As previously stated in Sec. I, carrier relaxation between QD electronic states via irreversible, incoherent, phonon emission has been questioned by recent theoretical works where the role of a strong electron-phonon interaction taking place in InAs/GaAs QDs has been stressed.[8,17]

It is worth noticing that in the low-T RPL spectra of both QDA and QDB samples (Fig. 1), the more intense PL mode (the peak at 1.108 eV for excitation energy of 1.165 eV) would correspond, in the HR framework, to the third phonon replica. This points to an exceedingly high value of the HR factor or to a mechanism similar to that invoked for the CdSe QDs.[23] In that case, intense high order resonances are due not to phonon replica but to zero-phonon emission from the ground state of QDs, whose excited states are resonant with the excitation laser and differ from the ground-state energy by a multiple of the LO-phonon energy. However, present samples QDA and QDB do not show excited states with an energy separation from the ground-state multiple of the LO-phonon energy. Moreover, although QDA and QDB do not share the same electronic structure, RPL resonances appear at exactly the same energy in both samples [see Figs. 1(e) and 1(f)]. This raises severe objections to an analysis of present RPL resonances in terms of phonon-assisted optical transitions in a nonadiabatic HR model.[21–23]

On the other side, within the phonon-bottleneck framework, the intensities of the RPL resonances are the result of the product between the probability $P_{ab}(E_{exc};E)$ and of a comblike $P_{rel}(E_{exc}-E;E)$ function peaked at $E_{exc}-E=nE_{LO}$ with $n=1,2,\ldots$. In QD ensembles characterized by broad size and shape distributions and for E_{exc} lying outside the QD ground-state DOS, the E dependence of $P_{ab}(E_{exc};E)$ should

be not so different from the NPL spectrum.[19,29] As a matter of fact, the intensities of three modes at 1.047, 1.078, and 1.108 eV in the RPL spectra of both samples, roughly corresponding to $n=2$, 3, and 4, are modulated according to the intensities of the respective NPL spectra. In sample QDA [Fig. 1(e)] the resonance with the maximum intensity is located, in energy, close to the NPL peak. On the contrary, in QDB, the RPL emission is dominated by two modes with similar intensities whose energies are, roughly, symmetrically located on the high and the low-energy sides of the NPL peak. Also the appearance of new modes on the low-energy side of the PL emission as the temperature increases in the RPL spectra of both samples (Figs. 2 and 3) could be explained in the same framework, due to the redshift, with the temperature, of the QD DOS.

A further test for the relevance of the nonadiabatic model should be provided by the dependence of the RPL resonances on the radiative channel efficiency. On the one hand, indeed, changes in the radiative efficiency of the QD system should not affect the RPL spectrum modulation, if the latter is due to phonon-assisted optical transitions in a Frank-Condon framework. It is quite hard to get QD samples with the same electronic structure and different nonradiative decay channel efficiencies. However, it has been recently demonstrated that hydrogenation of InAs/(In)GaAs QDs sharply increases the overall PL efficiency of the QD system.[27] Gurioli *et al.* in Ref. 27 demonstrated that hydrogen implantation does not modify the QD electronic states whereas it drastically affects the overall emission intensity. The PL yield enhancement observed upon hydrogenation of InAs/GaAs QDs has been attributed to passivation of strain relaxation induced defects, such as dislocation related dangling bonds, spatially located inside or nearby the QDs and energetically positioned below the dot ground state. These states provide efficient nonradiative decay channels for the excitons confined in the QDs.[27]

As a matter of fact, the NPL emission of QDA and QDB samples increases by a factor of 12 and 5, respectively, upon hydrogenation, while PL spectra slightly redshift, possibly due to the activation of larger QDs previously silent due to fast, competitive, nonradiative channels. Moreover, the ratio between the integrated intensities of hydrogenated and nonhydrogenated samples is roughly independent on temperature, thus showing that the nature (wetting-layer states) and the efficiency of the quenching channel do not change.[27]

The RPL spectra of the pristine and hydrogenated (at the optimum dose) QDA and QDB samples taken at different temperatures are shown in Fig. 4. At low T, hydrogenated and pristine samples show similar spectra with resonances at the same energy. However, the peak-to-valley intensity ratio and the relative intensities of successive maxima change on going from hydrogenated to pristine samples. For increasing temperatures, resonances in the RPL spectrum weaken in both pristine and hydrogenated samples due to the concurrence of the temperature activated channel provided by the wetting layer, which efficiently redistributes carriers among different QDs.[29,33] In the hydrogenated samples, resonances are washed out from the RPL spectra at lower temperatures (between 130 and 190 K) than in the pristine samples, where they persist quite clearly up to 190 K, at least in sample QDA. The change in the strength of multiphonon resonances

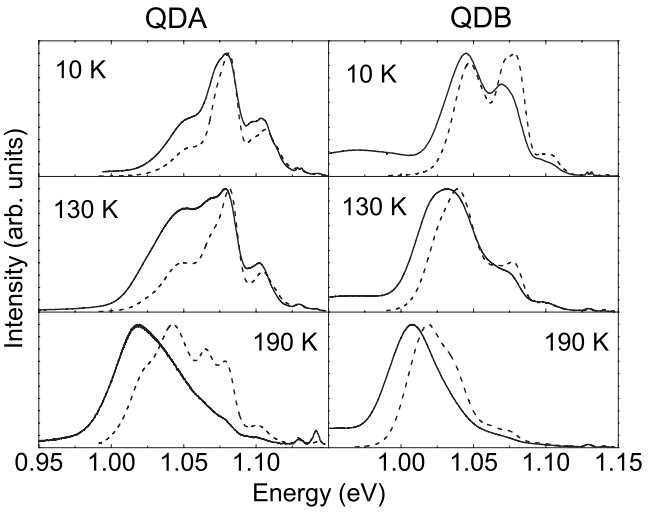

FIG. 4. PL spectra of the pristine (dashed lines) and hydrogenated (continuous lines) QDA (left panels) and QDB (right panels) samples measured at $P_{exc}=20$ W/cm^2 for three different temperatures.

upon hydrogenation, namely, for changing QD radiative efficiency, clearly demonstrates that the observed resonances cannot be due to phonon replica in a nonadiabatic HR model. Indeed, the strength of the phonon replica is determined by the HR parameter, namely, by the electron-phonon interaction, which is independent on QD radiative efficiency. Therefore multiphonon resonances in the RPL spectra of large, low-energy emitting QDs, such as those present in QDA and QDB samples, should be determined by the competition between different relaxation processes. Thus, the competition between nonradiative intradot recombination and multiphonon relaxation leads to an effective phonon bottleneck as suggested by Heitz *et al.* in Refs. 3 and 18: polarons activate efficient, not simultaneous relaxation channels by which thermalization in QDs is achieved. In turn, polaron relaxation is triggered by the instability of the LO-phonons associated with the lattice vibration anharmonicity.[8,14,16] This imposes that differences between polaronic energies should fall in the energy window $\approx 27-45$ meV and lead to finite, although quite large energy windows for a fast polaronic thermalization channel.

V. CONCLUSION

In conclusion, by a detailed study of the dependence of the spectral characteristics of InAs/GaAs QDs on electronic structure, temperature, and hydrogen content we have demonstrated that multiphonon resonances dominate the RPL spectra in the case of large QDs. In particular, the possibility of tuning the defect nonradiative recombination activity in the QDs by hydrogen implantation gives us a unique tool to settle the origin of the multiphonon resonances in RPL spectra. In fact, phonon bottleneck and polaronic model predictions strongly differ on the role of the nonradiative exciton recombination probability. The appearance of such modulated spectrum stems from the presence of an efficient intradot nonradiative decay channel. The ensuing description of

the carrier thermalization in QDs supports the conclusions reached in the old paper[1] of Benisty *et al.*: very fast thermalization takes place only for those QDs whose energy difference between excited and ground state matches a multiple of the LO-phonon energy. The electron-phonon interaction, which gives rise to polarons, only slightly affects this picture of thermalization processes in QDs. Indeed, the main effect of polarons is to permit an efficient carrier thermalization in QDs whose energy differences between the excited and ground states fall in rather large ($\approx$20 meV) energy windows around multiples of the LO-phonon energy (while thermalization rate is quite slow in QDs not satisfying this constraint). The presence of a competing intradot nonradiative recombination channel strongly reduces the PL from these QDs and gives rise to the observed intensity modulation in the RPL spectra.

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