Origin of the inhomogenous broadening and alloy intermixing in InAs/GaAs self-assembled quantum dots

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The photoluminescence (PL) spectra of a single-layer and a multilayer sample of self-assembled InAs/GaAs quantum dots (QD's) intermixed by thermal annealing at various temperatures, have been investigated. Intermixing is found to change both the optical transition energy and the intersublevel spacing of the QD energy levels. The linewidth (which is due to inhomogeneous broadening) of the PL emission peaks of both samples, are very similar, hence showing that the intermixing process is very homogeneous over 25 layers of QD's, whatever the annealing technique used. Moreover it is observed that the width of the PL peaks decreases for increasing interdiffusion down to about 5 meV for the ground-state transition of the multilayer sample. A reduction of the peak width is also observed for higher-energy states within the same ensemble of dots. The present paper shows that this effect can only be explained by some variation of the effective confining potential. Theoretical calculations have shown that the QD height, rather than the diameter, the volume, the composition or the strain, appears to be the key parameter that controls the sharpness of the PL linewidths in the investigated samples. Our model allows the identification of the main mechanisms involved in the inhomogeneous broadening of the optical transitions for the InAs/GaAs QD system.

I. INTRODUCTION

Semiconductor quantum dots structures present quantum effects that are very interesting for both theoretical investigations and optoelectronic applications. It is now possible to control the fabrication of QD's in such a way that they are defect free, and present very good optical properties (excited states are well resolved). The transition energy and the intersublevel spacing can be tuned by optimizing the fabrication parameters.[1,2] However it is difficult to control and limit the full-width-at-half-maximum (FWHM) of the PL emission peaks. The broadening of these peaks is generally attributed to QD size, shape, composition and/or strain inhomogeneities within the ensemble of QD's. Many research groups have studied the QD size distribution as a function of the fabrication parameters, trying to improve the size homogeneity by different means.[3–8] However the origin of the inhomogeneous broadening of the PL peaks is still not clear. Only a few theoretical studies of the size distribution have been made.[9,10] Calculations of electronic energy states show that QD spectra are very much dependent on the QD confining potential and therefore of the QD geometry.[11,12] For QD multilayer stacks, which are of great interest to increase the density of dots and therefore the device efficiency, the PL peak width may increase even further.[13] It is very important to control and be able to reduce the PL linewidths in order to optimize device properties such as detector sensitivity at a particular wavelength and laser gain.[14,15] Intermixing is a way to reduce the PL linewidth. It is a well-known technique used to shift the energy of the optical transitions.[12,16–22] It can affect both the height and shape of the confining potential, hence changing the transition energies and the intersublevel spacing. Moreover it was shown unambiguously that QD's retain their zero-dimensional density-of-states even after a strong interdiffusion of their potential by thermal intermixing.[19] This effect is assumed to occur via a simple atomic diffusion mechanism described by Fick's law. Generally it is observed that intermixing leads to a reduction of the linewidth of the PL peaks in QD's.[2,12,16–21] The effect of intermixing was also found to be dependent on the material system and variations of QD structural parameters such as diameter and height have been observed.[18,22] Several suggestions have been made in order to understand the observations. Since the width of the peaks is generally attributed to the QD nonuniformity, this effect was therefore attributed to a reduction of the QD size[20] and strain[21] inhomogeneities as the degree of intermixing was increased. A simple model of Fickian diffusion and its influence on the confining potential has been used recently to explain the pronounced narrowing of the emission peaks after intermixing.[19,12] The QD PL linewidths were also found to be temperature dependent.[23–25] Thermionic emission out of the narrow dots and recapture in broader QD's has been proposed to explain the narrowing of the PL linewidths as a function of temperature (up to 100 K).[25] Moreover some authors have also pointed out that certain high-energy QD state emissions are narrower than the ground state.[17,12] Different suggestions have been proposed to explain this last observation: (1) the dependence of the

carrier relaxation time on the intersublevel energy may affect the QD emission peak width[17] or (2) the variational change of the transition energy due to QD diameter fluctuations decreases for large diffusion lengths.[12] Considering the carrier wave functions in the QD, the excited states are less confined than the ground state and should therefore be less sensitive to the confinement potential fluctuations. This argument is consistent with narrower excited levels. The degeneracy of the high-energy levels, however, tends to broaden the PL peaks making this argument less valid. The qualitative interpretations and the simple models developed to explain the different factors influencing the inhomogeneous broadening emission of a QD ensemble are all satisfactory to a certain extent. However, considering the importance of controlling the FWHM of the optical transitions for efficient device fabrication, there is a need for a more systematic study of the key parameters that control the inhomogeneous broadening of the QD PL peaks. In the present paper, we investigate both experimentally and theoretically the effects of the confining potential on the QD PL linewidths. This study is an attempt to understand the mechanisms responsible for linewidth narrowing in QD's. The linewidths that we have determined for our as-grown investigated samples are of the same order of magnitude or smaller than most of the published results on a similar system consisting of a single layer or an uncorrelated stack of InAs/GaAs QD's. Indeed in most cases, the FWHM is found to lie in the range 50 meV to 100 meV for large ensembles of quantum dots.[1,13,18,21,24–29] However some research groups showed that it is possible to get even narrower linewidths by optimizing further the fabrication parameters.[2,7] This paper will show how to improve even more the sharpness of the peaks by in-growth and post-growth processing.

II. EXPERIMENTAL PROCEDURE

The two samples investigated in this paper were both grown by molecular-beam epitaxy but with different fabrication parameters. They consist of either one single layer of self-assembled InAs QD's or an uncorrelated stack of 25 layers separated by 30-nm GaAs barriers. The Indium flush technique has been used for both samples.[2,15] In the multilayer sample, we have observed that the Indium flush process of the higher layers produces a slight *in situ* anneal of the QD's already grown. For both samples, the QD's have a truncated lens shape, their average diameter and height are about 20 nm and 2.9 nm, respectively, while the average QD density is about $5\times10^{9}\mathrm{cm}^{-2}$.[15,30] Two different techniques of intermixing have been used. They involve different amounts of defects, depending on the nature of the capping layer deposited onto the surface of the sample prior to annealing. A thick layer of SiO_2 can strongly promote the interdiffusion whereas a thin layer of SrF_2 can prevent it.[31] Indeed a large number of vacancies is created at the interface between the GaAs and the SiO_2 layer. They can enhance the interdiffusion process when they diffuse deep through the heterostructures during the high-temperature annealing, while the Ga atoms evaporate at the surface. The SrF_2 layer is assumed to prevent the Ga atoms outdiffusion hence inhibiting the interdiffusion. In this paper the effect of intermixing on the PL emission linewidths is investigated for both samples and for both annealing techniques (SrF_2 defect-free interdiffusion and SiO_2 defect-induced interdiffusion). The photoluminescence experiment has been performed using an Argon ion laser ($\lambda=514.5$ nm) and a Ti-sapphire laser ($\lambda=810$ nm) as indicated in the legend of the figures. The spectra obtained with the different sources are rather similar (results not shown here). For the characterization of the intermixed and the as-grown samples, a GaAs photomultiplier tube or a cooled Ge detector, respectively, were used for the signal detection. All the experimental data presented in this paper are corrected for the detector response. Unless otherwise stated, the measurements were carried out at a substrate temperature of 14 K and an excitation density of about 200 $\mathrm{W/cm}^2$. The laser spot was defocalized in order to probe a large number of dots ($>10^6$).

III. EXPERIMENTAL RESULTS

The PL spectra obtained when testing intermixed QD's are shown in Fig. 1(a) for the multilayer stack and in Fig. 1(b) for the single-layer QD's. The first low-energy peak corresponds to the QD ground-state transition involving the first electron and hole states while higher energy peaks are associated with QD excited states. The PL peak at the highest energy (about 1.43 eV for the as-grown sample) is related to carrier recombination in the wetting layer (WL). A large blueshift of the PL peaks together with a reduction of the intersublevel spacing are observed for both series of intermixed QD's. Moreover a decreasing FWHM of the peaks is evidenced as intermixing is further carried out. A linewidth of 5.3 meV has been obtained for the ground-state of the multilayer sample intermixed for 30 s at 880 °C. It is the smallest inhomogeneous broadening reported in the literature for this system, to our knowledge. Samples shifted to similar energies (when the ground-state transition energy is similar for both samples, irrespective of the annealing conditions) by different techniques, present rather similar features with respect to the linewidths of the peaks and their relative intensity (spectra not shown here). Therefore the damage due to defects introduced by the intermixing technique is not significant with regard to the changes of the confinement potential induced by thermal atomic diffusion. Comparing the spectra of the intermixed single-layer with the intermixed multilayer QD's, when the ground-state transitions have a similar energy, it is clear that the linewidth of the peaks is comparable (at a given energy). This effect is observed even after strong interdiffusion (at high-annealing temperature) and whatever the technique used. No significant changes are observed in the optical properties of the QD's and thus the intermixing process must be homogeneous over the 25 QD layers (overall thickness of 0.75 μm). This is further confirmed by observing a similar carrier lifetime in the QD's for all the investigated samples (intermixed or not).[32] It should also be noted that similarly to the intermixed samples, the peak widths of the high-energy states of the as-grown samples (for multilayer and single-layer QD's) are narrower than that of the lower-energy states.

Figure 2 shows the QD PL peak width as a function of its corresponding position in energy. These data have been obtained by fitting the PL spectra of Fig. 1 with a sum of Gaussians (one for each resolved peak). In spite of some

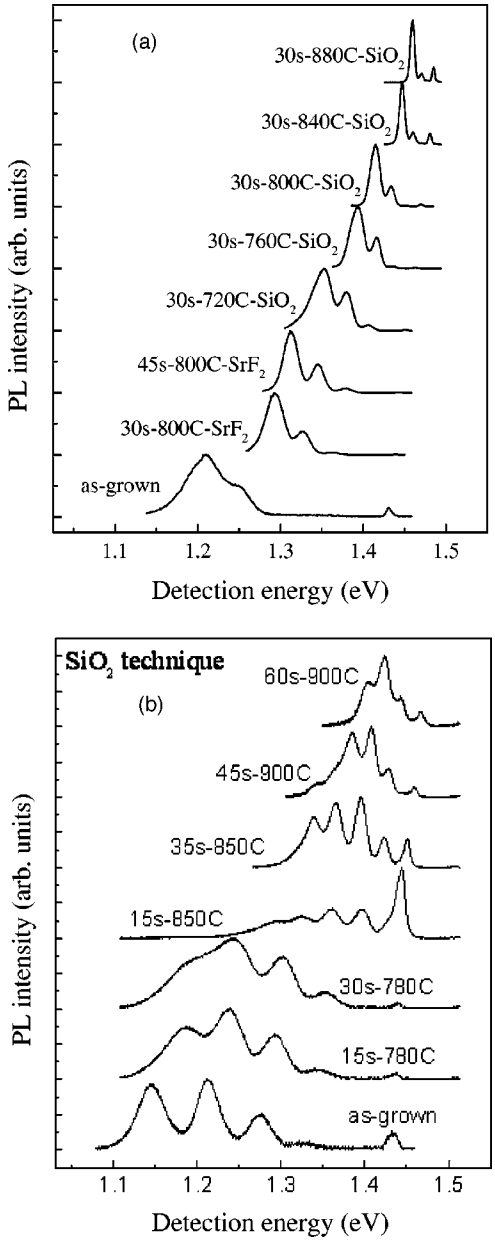

FIG. 1. Photoluminescence spectra obtained at 14 K, at an excitation density of about 200 W/cm^2 with a photon energy of 2.41 eV. The investigated samples correspond to a 25 layer stack of QD's (a) and a single-layer of QD's (b). The intermixing conditions are indicated next to the spectra.

datapoint dispersion, it is observed that the FWHM varies linearly with the optical transition energy, whatever the degree of intermixing and the energy level considered. It is remarkable that two different samples present a very similar behavior: the FWHM of the PL peaks decreases linearly with the transition energy (or energy difference between the barrier and the transition). The PL spectra show that although the confining potential is greatly reduced by the intermixing process, the presence of well-resolved peaks associated with different energy states indicates that the QD system retains its zero-dimensional nature. This result is in agreement with the observation of four distinct QD levels reported after an annealing step at 900 °C.[2,19] The observation of QD level filling as well as the presence of LO-phonon resonances in

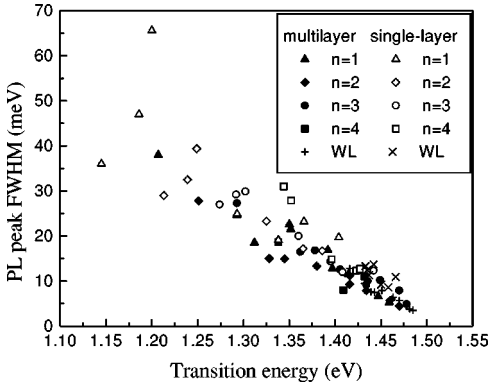

FIG. 2. FWHM of the PL peaks observed in Fig. 1 (from Gaussian fits) as a function of the energy of the transition considered.

the PLE spectra of InGaAs QD's, even after strong intermixing (850–900 °C) further confirms unambiguously the zero-dimensional nature of the QD's.[29] The diffusion length can be deduced from the shift of the WL by using a typical diffusion model of a quantum well. Extrapolating the data presented for a very similar system,[19] the diffusion lengths for the most intermixed samples are estimated to be about 1.7 nm for the single-layer and about 2.7 nm for the multilayer samples. The difference between these two values may be due to the *in situ* anneal that occurred during the growth of the multilayer sample, as discussed in Sec. II. It should be noted that the multilayer QD's do not exhibit larger FWHM of the PL peaks (as one would expect from a larger number of dots over several layers) nor narrower (as observed from correlated QD stacks) compared to the single-layer sample. Hence the main inhomogeneities of the QD ensemble are present in the growth plane and increasing the number of QD layers does not alter the overall uniformity. As observed for InGaAs/GaAs QD's, at low temperatures ($T<100$ K), PL linewidth narrowing could result from thermal emission of carriers out of the smaller dots and recapture by the larger dots (having a deeper confining potential).[25] Thermal emission (see for example Refs. 17 or 23) has been included in our model but it is not sufficient to explain the variation of the linewidth observed experimentally. Moreover it is obvious in Fig. 3(a) that there is no variation of the FWHM for the as-grown sample when the temperature is increased from 15 K up to 195 K. The same behavior has been observed up to 100 K for intermixed samples. In order to understand the results of Fig. 2, we have also checked the potential influence of many-body effects on the QD peak width. The number of carriers per dot and the formation of multiexcitons complexes have been shown to change the single dot optical transition energy, as observed in InGaAs/GaAs QD's.[33] These effects contribute to the PL peak width of the QD ensemble. However these intrinsic effects (few meV) are small compared to the FWHM of the PL features. Moreover, the PL spectra obtained at various excitation densities [Fig. 3(b)] on the as-grown single-layer QD sample, show that higher-energy states are filled when the excitation density is increased while the peak width of the ground-state optical transition does not broaden significantly in the range investigated. Hence we suggest that the linear dependence of the FWHM on the energy position of the QD peak could only result from the influence of the confining potential on

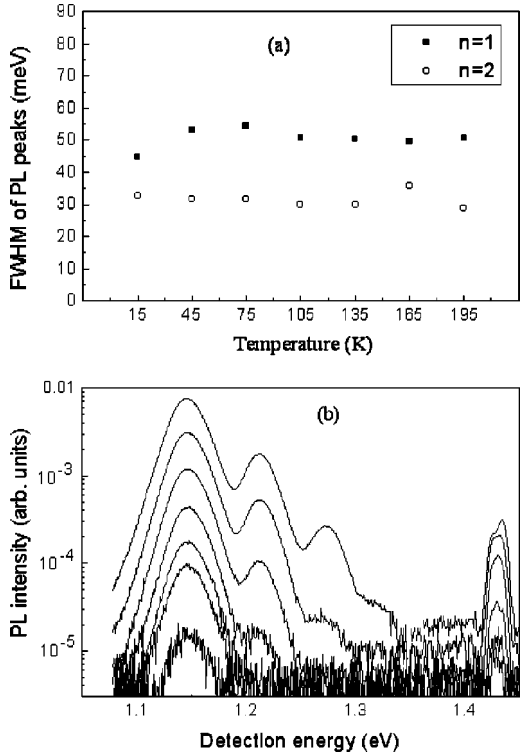

FIG. 3. (a) FWHM of the PL peaks of the as-grown multilayer QD's as a function of the temperature, $E_{photon}=1.53$ eV; (b) PL spectra of the as-grown single-layer QD's measured at an excitation density of 180, 60, 18, 6, 1.8, 0.9 and 0.18 W/cm^2 from top to bottom, $E_{photon}=2.48$ eV.

the energy sensitivity of the QD levels with respect to the size, shape, composition, or strain inhomogeneities.

IV. MODELING

In order to investigate the key parameters that control the inhomogeneous PL peak width of the QD ensemble, we have calculated the effects of small changes of the structural parameters on the energy spectrum. Quantum dots are modeled by considering the top of a sphere laying on a thin quantum well. Effects due to strain, discontinuities in the effective mass, dielectric constant etc., are included. The three-dimensional Schrödinger equation in the effective-mass approximation was solved using the adiabatic approximation. In this approach it is considered that the electron's wave function is strongly confined to the lowest subband of the wetting layer. The effective lateral potential for confined electrons can be calculated considering the motion along the growth axis, for a given thickness of the structure. Details of the calculation method are summarized elsewhere.[11] A simple one-dimensional Fickian diffusion model was used to simulate interdiffusion in the growth direction.[19] Indeed, due to the lens shape of the dots (diameter d much larger than height h), it is the atomic interdiffusion along the growth axis that will cause the stronger effect on the QD optical transition energy. The effective radial potential for lateral confinement was calculated as a function of the distance from the QD center, by modeling QD's as hemispherical caps laying on a WL (a wetting layer thickness of 0.54 nm is assumed in this calculation). It was obtained for various diffusion lengths L_d (characteristic feature of the interdiffusion process), for various sets of parameters (h,d,h/d, In concentration) by using an error function intermixing profile in the growth direction. We assumed that the strain remains directly correlated to the In composition of the QD's, even after intermixing. Moreover the In concentration of the dots is assumed to be homogeneous. Some effective potential profiles are given in Fig. 4 for as-grown and intermixed samples with typical structural parameters. The reference potential is compared to the potential obtained after intermixing and after being truncated. The energy spectrum was then deduced by solving the radial Schrödinger equation for various values of the angular and radial quantum numbers considering a constant effective mass of 0.067 m_o.

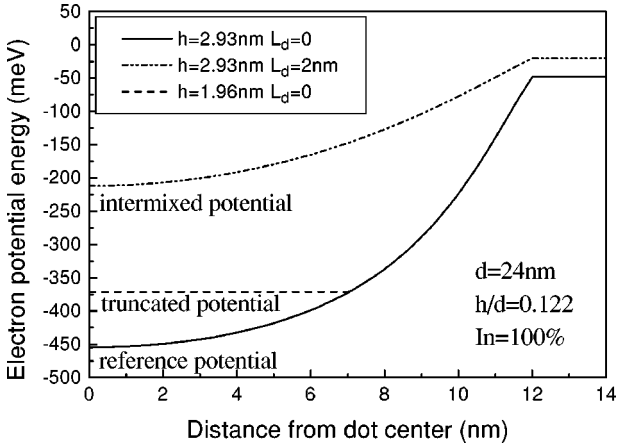

FIG. 4. Effective potential profiles for full-height ($h=2.93$ nm) and truncated ($h=1.96$ nm) as-grown QD's as well as for intermixed QD's ($L_d=2$ nm). Other parameters are as follows: In $=100\%$, $d=24$ nm, $h/d=1.222$.

The square of the electron wave functions in a QD have been plotted in Fig. 5 in order to determine the influence of the interdiffusion and height variations. Two different QD levels are shown: $n=1$ ($m=0$) is the ground state and $n=3$ ($m=2$) is the second excited state. Clearly these wave

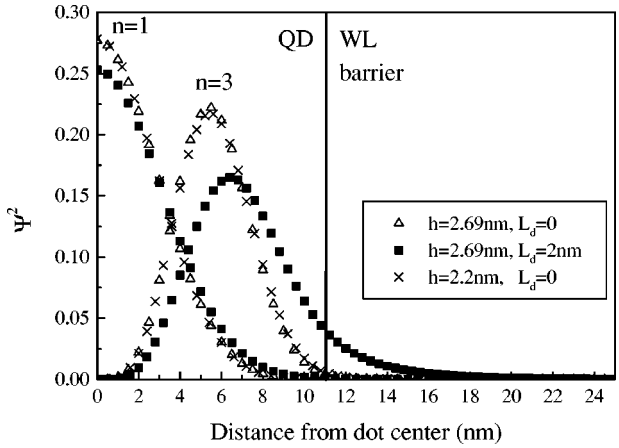

FIG. 5. Calculated squared-wave functions are plotted for the first and third levels in the QD. The vertical line at 11 nm corresponds to the confinement barrier. Wave functions are shown with reference parameters, after interdiffusion ($L_d=2$ nm) and after height variations ($h=2.2$ nm). Reference parameters are: $d=22$ nm, In$=100\%$, $h=2.69$ nm, and $L_d=0$.

functions are well confined inside the QD. The occupancy probability of the electrons inside the QD is greater than 98% for the first three levels of the as-grown QD's ($L_d = 0$) and for the first two levels of the intermixed QD's ($L_d = 2$ nm). In the latter case it is still confined to 90% for the third level. Thus even after a strong interdiffusion, the electron wave functions are well confined inside this hemispheric region of the dot. Moreover a change in the height of the QD's is found to have only a weak influence on the degree of confinement of the first three as-grown QD states and of the first two intermixed QD states. The third level is still confined to over 75% for $L_d = 2$ nm.

For the sake of simplification, we assume that the adiabatic approximation (separation of the z and r wave functions) is still valid in the case of strong intermixing. If interdiffusion occurs in an isotropic way, the whole structure (wetting layer and quantum dot) would be enlarged. The height of the dot would thus remain much smaller than its diameter and the wave function would still be well confined. The zero-dimensional (lateral) confining potential is related to the difference in energy between the ground and first excited state and can be compared to the energy difference between the GaAs and the WL. Experimentally we have found that the energy difference between the first two transitions (about 11 meV for the strongest intermixed multilayer sample) is still much lower than the energetic difference between the GaAs and WL transition energies of about 34 meV. For a comparison, in the as-grown sample, these values are, respectively, 44 and 87 meV. Therefore we believe that the confinement in the z direction (growth axis) is still more effective than in the r direction (growth plane). And the adiabatic approximation would still be valid.

The results presented here take into account the degeneracy of the levels $n = 3$ (radial/angular quantum numbers: 0/2 and 1/0) and $n = 4$ (respectively, 0/3 and 1/1). The results presented in Fig. 6 correspond to variations in the energy of a QD electronic level resulting from small changes of the structural parameters around some nominal values (the reference set of parameters is indicated on each graph). These energy variations can be correlated to the FWHM of PL peaks (assuming that QD hole energy levels would behave in a similar way). For high-energy states we have considered the average energy difference between the degenerate levels but in any case the energy variation due to the degeneracy is negligible compared to the changes produced by the parameter fluctuations. The electronic energy variation is plotted as a function of the diameter [Fig. 6(a)], the volume [Fig. 6(b)], the height [Fig. 6(c)], the In composition [Fig. 6(d)], and the truncated height [Fig. 6(e)] of the QD's. The truncated height corresponds to the actual height of the QD after its top has been truncated, as can be the case for indium-flushed QD's.[15] These energy variations are calculated for the first three or four QD levels for the as-grown sample ($L_d = 0$) and for one strongly intermixed sample ($L_d = 2$ nm). For a nonintermixed sample, we observe similar energy variations resulting from a 10% fluctuation in the structural parameters around the nominal values, for each QD level. These energy variations are about 10 meV for a 10% fluctuation in the QD diameter [Fig. 6(a) or the QD In concentration (Fig. 6(d)]. A 10% fluctuation in the QD height [Fig. 6(c)] or the QD truncated height [Fig. 6(e)] would lead to about 20-meV variations. And finally a 30-meV variation is obtained for a 10% fluctuation of the QD volume at a constant h/d ratio [Fig. 6(b)]. The energy variations for each parameter fluctuations decrease significantly for intermixed samples except for In concentration fluctuations for which the opposite effect is observed. For all cases (but the truncated height variation), the behavior of the QD energy level as a function of one particular structural parameter is approaching a linear dependence. It is observed that the slope of these energy variations is more important for the excited levels, in all cases except for the one corresponding to the height dependency for intermixed samples [Fig. 6(c)]. Since this behavior is the only one compatible with our experimental observations (Fig. 2), these calculations indicate that the QD height is the key parameter that affects the width of the QD PL peaks. Even for the truncated height fluctuations, the high-energy states show smaller variations than the ground state, but this effect becomes less obvious for higher-energy states (except if the dot is very much truncated by half its nominal height) which is not consistent with the experimental results. Moreover the variation of the FWHM with higher energy states is less pronounced after interdiffusion. However, the fact that the FWHM is narrower for the excited levels than the ground-state transitions, when considering the as-grown and weakly intermixed series, is in very good agreement with the experimental data and proves clearly the favorable effect of truncating the QD height. We believe that the optimization of the growth conditions gives rise to an ensemble of dots with reduced standard deviations corresponding to the QD volume and In composition. Note that thickness fluctuations are also responsible for the broadening of the PL peak in the case of a two-dimensional quantum well. As seen for the wetting layer in Fig. 2, linewidth variations are similar to that observed for the QD high-energy states.

Figure 7 shows the energy variations corresponding to a 20% fluctuation in the QD height plotted against the QD electronic energy measured from the bottom of the harmonic potential. The depth of the confining potential is assumed to be 644 meV. The data points include the energy variations calculated for the first three or four levels and for different diffusion lengths ($L_d = 0$, 0.5, 1.0, 1.5, 2.0, and 2.5 nm). For $L_d > 0.5$ nm, the graph shows a linear dependence very similar to the experimental results seen in Fig. 2. Indeed the width of the QD levels gets smaller for increasing diffusion length, or larger interdiffusion, as observed experimentally. This good agreement observed for intermixed QD's confirms our conclusion that the QD height fluctuations are responsible for the inhomogeneous QD PL peak widths. On the other hand, the values calculated for low-diffusion lengths ($L_d \leq 0.5$ nm) show a different trend: the energy variation is almost constant whatever the QD level or the electronic energy considered [Figs. 6(c) and 7]. This observation confirms our statement that the as-grown multilayer sample already has some degree of intermixing originating from the growth process. In general, such intermixing could occur during the growth of the cap layers, especially if it involves higher temperatures and/or long growth times, as can be the case for laser heterostructures.[15] A similar observation has also been reported previously.[34]

We can attempt to give a qualitative description of the interdiffusion mechanisms responsible for the observed PL

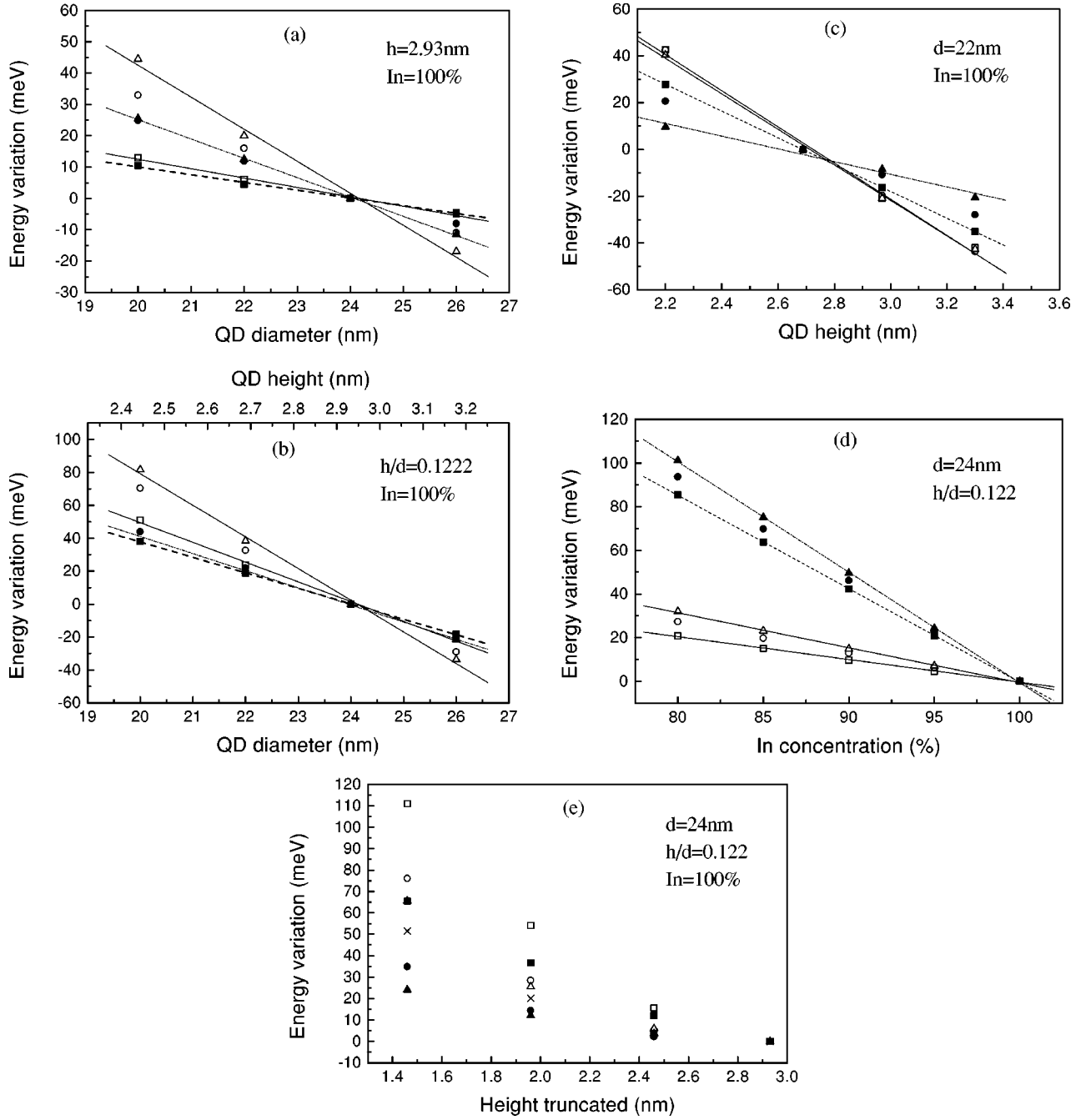

FIG. 6. Energy variation of the QD electronic levels plotted against (a) the QD diameter (with fixed height), (b) the QD diameter (with fixed aspect ratio), (c) the QD height, (d) the QD In composition, and (e) the QD truncated height. The reference parameters used for the calculation of the energy difference are indicated inside each figure. The legend is as follows: squares correspond to $n=1$ (ground state), circles to $n=2$ (first excited state), triangles to $n=3$, and crosses to $n=4$. Open symbols describe nonintermixed QD's ($L_d=0$) and solid symbols, intermixed QD's ($L_d=2$ nm). Straight lines are linear fits for the $n=1$ and $n=3$ QD levels.

spectral changes. Interdiffusion is generally interpreted as the diffusion of species through an interface. Therefore the composition, strains, and QD size can change after intermixing. If the strain fields inside and around the dots are quite uniform, it is reasonable to assume that the diffusion process is mostly isotropic. The validity of this simple assumption is still questionable since contradictory results have been reported (e.g., QD size reduction[22] versus size increase[35]). Due to the very small dot aspect ratio ($h/d=0.122$), this assumption can be considered here since the vertical diffusion will have a stronger influence on the PL spectral shape than the lateral diffusion of species (for a large degree of intermixing this assumption becomes less valid) The transition energy shift and the interlevel reduction are usually attributed to modifications (composition and shape) of the QD confinement potential. Intermixing induces a reduction of the average In content together with a reduction of the strain fields in the dots and in the wetting layer. These variations are responsible for most of the large blueshift observed for the QD and WL emission lines. However they cannot account alone for the reduced interlevel spacing with interdiffusion nor for the reduced linewidths observed for the higher energy PL

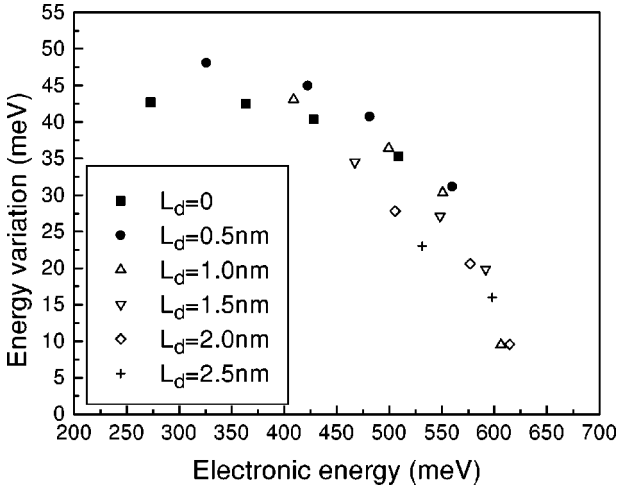

FIG. 7. Effect of QD height fluctuations on the energy of the three or four first QD levels as a function of the electronic energy. The energy difference is calculated from reference values at $h = 2.69$ nm and other values obtained at $h = 2.2$ nm with the constant parameters $d = 22$ nm and In=100%. The energy of each QD level was determined for various diffusion lengths, at $L_d = 0$, 0.5, 1.0, 1.5, 2.0, and 2.5 nm.

transitions [see Fig. 6(d)]. Qualitatively, one can say that intermixing tends to smooth out the compositional and strain dispersion from dot to dot probably because gradients of these parameters act as the driving force for species diffusion. According to our calculations only volume/diameter variations (enlargement) could explain the reduction of the interlevel energy with intermixing [Figs. 6(b) or 6(a)] whereas the decrease of the linewidth for higher-energy levels and increasing interdiffusion can only be due to larger QD height [Fig. 6(c)]. At first sight, increasing uniformly the size of each dot of the ensemble tends to diminish both the interlevel energy and the inhomogeneous broadening of the QD PL peaks. However for a given degree of intermixing characterized by similar diffusion lengths in both r and z directions, the relative variation of the QD diameter is much smaller than the relative variation of its height, which therefore becomes a key parameter for the evolution of the inhomogeneous broadening. It should be noted that our results are in good agreement with other studies on QD (Refs. 4 and 10) or QW structures.[36]

To achieve narrow QD linewidths is very important for improving some characteristic performances of QD photodetectors and lasers. As mentioned before, the intermixing process does not only reduce the linewidth but also the intersublevel energy, while the transition energy increases. However this is not a critical problem since this latter parameter can be adjusted by varying the fabrication parameters in different ways. For example, the transition energies can be adjusted by controlling the amount of InAs deposited onto the substrate. It is found that by changing the size of the dots in this way, it is possible to vary the peak energies while keeping a roughly constant interlevel spacing.[37] Depending on the growth parameters, the height of the dots can change significantly and a change in the thickness of the dots is found to have a large influence on the transition energy.[37] Moreover the interlevel energy spacing can be varied by changing the QD growth temperature.[2,15] Thus by varying the growth parameters, it should be possible to optimize the linewidth at a given transition energy and the interlevel spacing independently.

In supplement to this discussion, we would like to mention that some other research groups have observed that the high-energy PL peaks are broader than the ground-state peak.[12,26] This could be explained by assuming different growth conditions where QD size, volume, composition, or strain inhomogeneities dictated the width of the PL peak and did overwhelm height fluctuations. This is further confirmed by the rather large FWHM of their PL peaks compared to the values presented here.

V. CONCLUSION

To summarize, our results give an insight to the cause of the broad inhomogeneous peaks observed by photoluminescence for large ensembles of InAs/GaAs self-assembled quantum dots. This study shows that it is possible to identify the limiting mechanisms involved in the inhomogeneous broadening in InAs/GaAs self-assembled QD's by comparing the FWHM of the ground and excited peaks. Calculations have shown that although QD diameter, volume, In composition, and/or strain inhomogeneities can be important in some samples, only QD height variations can explain our observation that higher-energy states become narrower than lower-energy states. For the high-quality InAs/GaAs system investigated, the QD height variations can therefore effectively control the sharpness of the PL peak. It should be noted that a very similar behavior was observed for our different InAs/GaAs QD samples and that the results are in agreement with most of the experimental and theoretical work reported for this system. Therefore the model described in this paper can be generalized to the very common and well-studied InAs/GaAs system. More systematic experiments should be performed on different systems (strain fields and diffusion constants can be different), but we can expect that the same behavior can occur in other systems. It could be also very interesting to investigate further the linewidth variations in correlated stacks of QD's, where QD layers are coupled in the growth direction and thus the QD height is also a very important factor.

Intermixing was found to be a powerful tool for adjusting the transition energy and the sharpness of the PL peaks transitions (a FWHM of 5 meV was obtained for the ground state of a 25 layer stack of QD's). Moreover, since the FWHM of the QD PL peaks depends linearly on the transition energy (whatever the QD level considered, the number of QD layers and the amount of intermixing), it should be possible to predict and tune the linewidth by varying the rapid thermal annealing of the QD's. These observations can be very valuable for optimizing devices such as QD lasers and detectors.

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