Barrier-induced carrier localization effects in ordered/disordered/ordered quaternary quantum wells grown on GaAs substrates

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Optical and micro-structural properties of ordered/disordered/ordered InGaAsP quantum wells grown on GaAs substrates were investigated by photoluminescence spectroscopy, high-resolution transmission electron microscopy and selective area diffraction. Strong evidence of carrier localization effects was obtained from low temperature photoluminescence experiments. Photoluminescence spectra of thinner quantum wells were dominated by a broad emission band located at energies below the bandgap of the well material. The energy peak position of this emission varied considerably with the laser excitation power. Carrier localization was attributed to potential fluctuations in the barrier and well layers, as a result of two coexisting effects: Spontaneously atomic ordering and, in a minor degree, alloy inhomogeneities. We show that a reduction of the ordering degree in the bottom barrier layer resulted in a considerable decrease of localization effects in quaternary quantum well heterostructures.

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I. INTRODUCTION

Semiconductor $In_{1-x}Ga_xAs_yP_{1-y}/In_{1-z}Ga_zAs_tP_{1-t}$ heterostructures grown lattice matched to GaAs have received considerable attention due to its potential application to the fabrication of aluminum-free high-power laser diodes emitting in the near infrared wavelength region.[1] However, depending on the growth conditions epitaxial $In_{1-x}Ga_xAs_yP_{1-y}$ layers may exhibit spontaneously CuPt-like ordering and alloy phase separation effects.[2–4] In the case of quaternary quantum well (QQW) structures grown on GaAs substrates both the barrier and the well layer materials may exhibit distinct ordering degrees.[5–7] Anomalous photoluminescence (PL) emission bands from partially ordered ternary and quaternary alloy layers grown on GaAs substrates have been previously reported.[6–9] The emission characteristics of these bands are strongly dependent on the growth conditions and substrate orientation.[8–10] In partially ordered alloys an inhomogeneous distribution of ordered domains with different sizes and ordering degrees gives rise to potential fluctuations in the real space.[9] At low temperatures photo-generated carriers tend to localize in the minima of these potential fluctuation. The effectiveness of the carrier localization process depends on the ratio between the length scale of the potential fluctuations and the excitonic Bohr radius.[9] The low-temperature PL characteristics of partially ordered alloys is frequently described by a broad low-energy emission, with respect to the absorption edge, with long radiative lifetimes, anomalous peak position dependence with temperature and strong blue shift with power excitation levels.[8] The coexistence of ordering and phase alloy separation effects modifies even further the band structure of quaternary heterostructures grown on GaAs substrates. As a consequence, detailed knowledge of ordering and phase separation effects is crucial for further optimization of devices based on quaternary quantum wells.

In this work we present experimental results on the effect of the well width, barrier alloy composition, and barrier growth temperature on the optical emission properties of quaternary quantum wells grown lattice matched to GaAs substrates. A broad emission band located at energies below the bandgap of the well material dominated the low-temperature (LT) photoluminescence spectra of QQWs when the well thickness was varied from 5 to 60 nm. The emission characteristics of this band depend strongly on the PL power excitation, barrier alloy composition and barrier growth temperature. Our results indicate a direct correspondence between spontaneously ordering effects in the barrier and the QQW emission properties. Transmission electron diffraction experiments in the same set of QQW samples confirmed the presence of strong atomic ordering in the barrier layers.

II. EXPERIMENTAL DETAILS

Samples analyzed in this work were grown by low-pressure metal-organic vapor phase epitaxy (LP-MOVPE) on GaAs:Si (100)-oriented substrates. After a 300 nm thick GaAs buffer layer, the quantum well and barrier structure is composed by an $In_{0.13}Ga_{0.87}As_{0.74}P_{0.26}$ quantum well ($E_{gw} \sim 1.55$ eV, at $T=300$ K) of variable thickness (L_z) ($5 < L_z < 270$ nm), sandwiched by 500 nm thick $In_{0.44}Ga_{0.56}As_{0.09}P_{0.91}$ barriers ($E_{gb} \sim 1.85$ eV at $T=300$ K). The growth temperature (T_g) was kept fixed at 670 °C for both the QW material and the top barrier layer, while the bottom barrier was grown at different temperatures. For comparison purposes, two additional samples were prepared: A single 300 nm thick layer of the same $In_{0.13}Ga_{0.87}As_{0.74}P_{0.26}$ well material grown on GaAs, and a SQW structure using 500 nm of ternary $In_{0.485}Ga_{0.515}P$ alloy

TABLE I. Sample identification, quantum well width (L_z), barrier alloy material, barrier (T_{gb}), and well (T_{gw}) growth temperatures of the QQW samples studied in this work.

Sample	L_z (nm)	Barrier material	$T_{gb}/T_{gw}/T_{gb}$ (°C)
Q5	5	Quaternary	670/670/670
Q10	10	Quaternary	670/670/670
Q30	30	Quaternary	670/670/670
Q60	60	Quaternary	670/670/670
Q90	90	Quaternary	670/670/670
Q270	270	Quaternary	670/670/670
T30	30	Ternary	670/670/670
Q30L	30	Quaternary	600/670/670
Q30H	30	Quaternary	730/670/670
B300	300	Single layer, barrier	670
W300	300	Single layer, well	670

as barriers. All layers were grown nominally lattice-matched to the GaAs substrate. This was confirmed by double-crystal x-ray diffraction experiments in all investigated samples. A summary of the samples' structural parameters and corresponding labeling are shown in Table I.

Photoluminescence experiments were performed at 12 K using the 514.5 nm line of an Ar^+ laser as excitation source. The power excitation density was varied from 0.01 to 100 W/cm^2. The PL emission was dispersed by a 0.75 m single spectrometer and detected by a cooled GaAs photomultiplier. High resolution transmission electron microscopy (HRTEM) and selective area diffraction (SAD) patterns were taken using an atomic resolution JEOL JEM-3010 microscope with 300 kV of acceleration voltage. TEM specimens for cross-sectional analysis were prepared by mechanical polishing and dimpling, followed by ion milling using a nitrogen-cooled stage.

III. RESULTS AND DISCUSSION

Normalized low-temperature PL spectra of QQWs grown with identical barrier and well alloy compositions but with different values of L_z (see Table I) are shown in Fig. 1. All the spectra were obtained at similar excitation power conditions. For comparison purposes we also show in Fig. 1 the PL spectra of bulk layer samples with compositions corresponding to the well (W300) and barrier (B300, see inset) materials. The PL spectrum of the quaternary barrier material consists of a single emission band. In contrast, the PL spectrum of the quaternary well material exhibits two peaks. The high-energy (HE) emission is attributed to the direct band-to-band (or excitonic) recombination. The low-energy (LE) emission involves spatially indirect recombination of carriers at the boundaries of ordered domains, yielding the well-known below band gap emission.[7,8,11] The single emission observed in the PL spectrum of the barrier sample is attributed to electron-hole pair transitions in a highly ordered material (see details below) with domains of relatively large lateral extension.[7,8] In the range $5 \leqslant L_z \leqslant 90$ nm (samples Q5 to Q90) the PL spectra are dominated by a relatively broad single emission band with full-width at half-maximum (FWHM) between 13 and 22 meV (see Fig. 1). The PL spectrum of sample Q270 is similar to the bulk QW material layer (W300). Both spectra exhibited two well-defined emission bands. The HE band is relatively narrow (FWHM $\sim$4 meV) while the LE exhibited a FWHM of $\sim$13 meV.

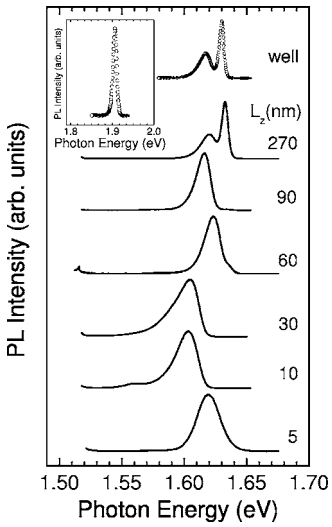

FIG. 1. PL spectra of QQW samples grown with different L_z values. The well and barrier (inset) bulk materials spectra are also plotted for comparison. All data were obtained at 12 K with excitation of 0.6 W/cm^2.

The evolution of the PL peak of QQW samples with well width values shown in Fig. 1 does not correlate quantitatively with the expected results from a single QW within a type-I band alignment in the effective mass approximation. Firstly, energy peak positions of Q5 to Q90 QQW samples were located below the emission of the well material. Then, the QW PL peak shifts to bottom energies as L_z is increased from 5 to 10 nm (see Fig. 1). However, when the well width is further increased to $L_z=30$ and 90 nm, the QW PL peak shifts to an energy position similar to that obtained firstly for the QQW sample with $L_z=5$ nm.

Additional information about the origin of the optical transitions in the QQWs shown in Fig. 1 can be obtained from PL experiments with different laser excitation power levels. LT-PL spectra of QQW samples Q5, Q10, Q30, and Q90 measured with different laser power levels are shown in Fig. 2. The power-excitation dependent PL spectra of the QQW samples with $L_z=5-30$ nm [Figs. 2(a)–2(c)] consisted of a single emission band (LE). As the power is increased the QQW PL-peak position shifts to higher energies. Increasing the excitation laser power by three orders of magnitude resulted in total PL peak blue shifts of 7, 46, and 100 meV for samples Q5, Q10, and Q30, respectively. The PL spectra of sample Q90 [Fig. 2(d)] exhibited a single excitation dependent (LE) emission at low excitation powers. As the laser power is further increased an additional band at higher energies (HE) is observed. The HE band dominates the PL spectra of sample Q90 for higher levels of excitation. Similarly to the well layer and sample Q270, the HE emission band in the PL spectra of sample Q90 is characterized by a narrow line

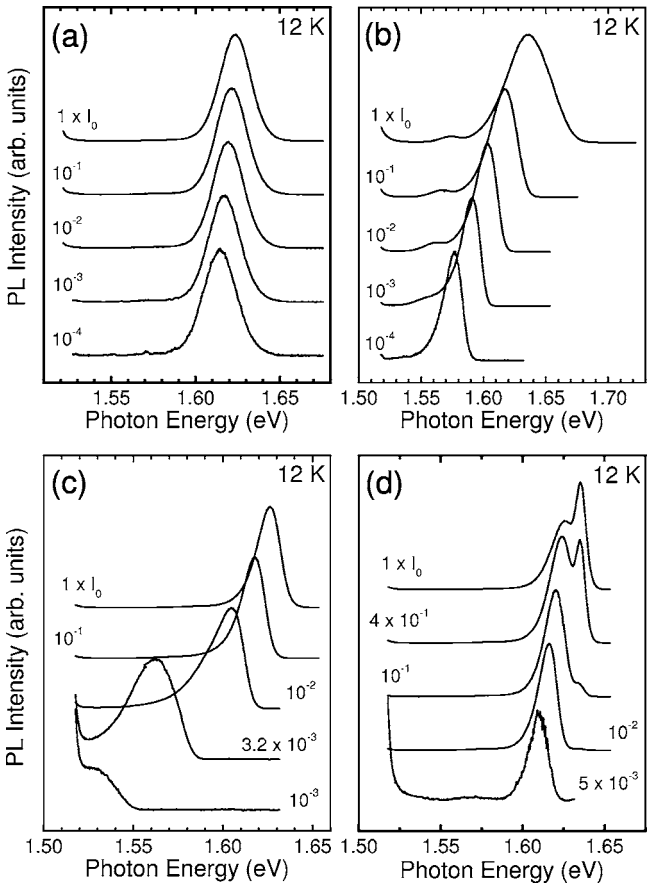

FIG. 2. LT-PL (12 K) spectra as a function of the laser excitation power for QQW samples with: (a) L_z=5 nm, (b) L_z=10 nm, (c) L_z=30 nm, and (d) L_z=90 nm. Maximum excitation was I_0 =100 W/cm^2.

width (FWHM ~4 meV) and by a peak position insensitivity to the laser power. A total blue shift of ~20 meV was obtained for the LE emission of sample of Q90 over three orders of magnitude of laser power change. The PL excitation-power dependent results (not shown) for sample Q60 are similar to those for Q90 [see Fig. 2(d)]. In the case of samples W300[7] and Q270 the PL peak emission corresponding to the LE and HE (see Fig. 1) were observed at all laser excitation levels (not shown). In addition, the nondependent power excitation HE transition dominates the PL spectra even at very low laser powers (see Fig. 1).

A summary of the total LE peak energy blue shift for the QQW samples as a function of L_z is shown in Fig. 3 (solid circles). PL blue shift of QQWs is maximum for L_z=30 nm, decreasing for both narrower and wider QQWs. The results shown in Figs. 2 and 3 indicate that the LE emission characteristics vary significantly with L_z, particularly in the range $5<L_z<60$ nm (at all excitation levels). At larger well widths ($L_z \geqslant 60$ nm) the HE emission is observed at high-excitation power levels and only for widest QQW sample (L_z =270 nm) investigated here a bulk-like behavior was observed. In a previous work[7] both the barrier and well quaternary materials were separately studied by PL and asymmetric x-ray diffraction. Since quantum confinement is very weak for $L_z>30$ nm, one could expect a bulk-like spectrum for the

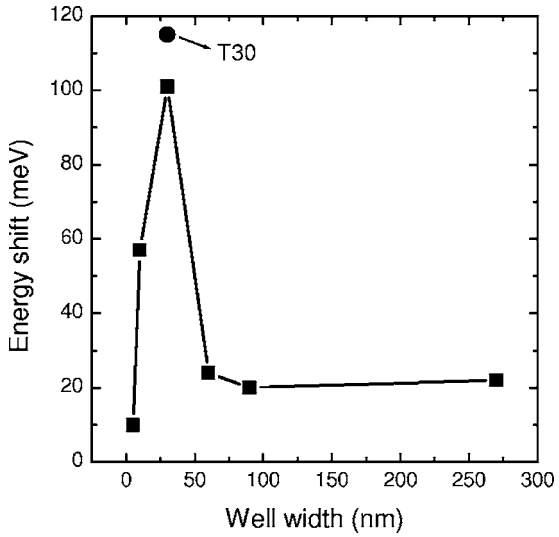

FIG. 3. Total PL peak blue shift for QQWs samples for different values of L_z (squares) and for sample T30 (circle).

emission of QQWs (such as W300 in Fig. 1). The absence of the exciton-related feature (HE emission) in the PL spectra of QQW samples with $L_z<60$ nm (Figs. 1 and 2) and the strong dependence of their PL peak on the excitation power (Fig. 3) suggest that carrier recombination of spatially separated electron-hole pairs, with variable pair-distance (i.e., ordering or phase separation), is the dominant process in the low-temperature PL emission spectra of our QQWs. The blue shift observed in the PL spectra observed in Figs. 2 and 3 could be attributed to the filling of these localized states with different potential energies.As the excitation intensity increases, higher energy states starts to be occupied and a blue shift of the PL peak takes place.

At this point, experimental data may point to ordering influence on the QW emission, since for thicker layers the HE band (thus the bulk spectra) is recovered.[7–9] Before we could identify the physical mechanism of the anomalous PL behavior as a function of excitation power (Fig. 2), other alternatives must be explored.

A possible origin for the observed PL spectra of QQWs might be the presence of low-bandgap interlayer between the bottom barrier and the well material. This effect was observed in GaInP/GaAs heterointerfaces and involves fundamentally the formation of an interlayer as a result of an As/P-exchange and In carry-over processes.[12,13] The suppression of the power dependent LE emission in GaInP/GaAs QW system was obtained by inserting a GaP layer between the bottom GaInP barrier and the GaAs well material.[10] The interfaces at the QQW system, however, are expected to be more stable to the formation of interlayers due to a significant less influence of As/P-exchange as well as In carry-over.[14] Nevertheless, we also fabricated QQWs introducing GaP layers on both bottom and top interfaces between the barriers and the well material (not shown). Our results indicated negligible influence of the GaP layers in the PL emission characteristics of QQWs. This rules out interlayer formation as the main recombination mechanism in the PL spectra of QQWs.

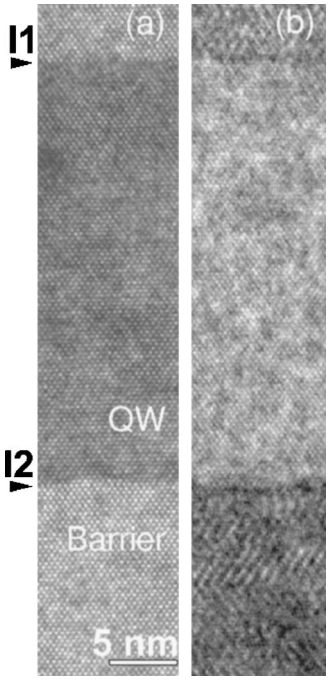

FIG. 4. HRTEM micrograph of sample Q30 taken: (a) Along the $[0\bar{1}1]$ and (b) [011] directions. Interfaces are better defined in (a) where I1 indicates the top interface and I2 arrow the bottom interface, being the well material in the middle. They show a roughness of ±3 ML. In (b) intense ordering is demonstrated in the barrier layers (zigzag pattern) but it might be below the detectable limit in the well material.

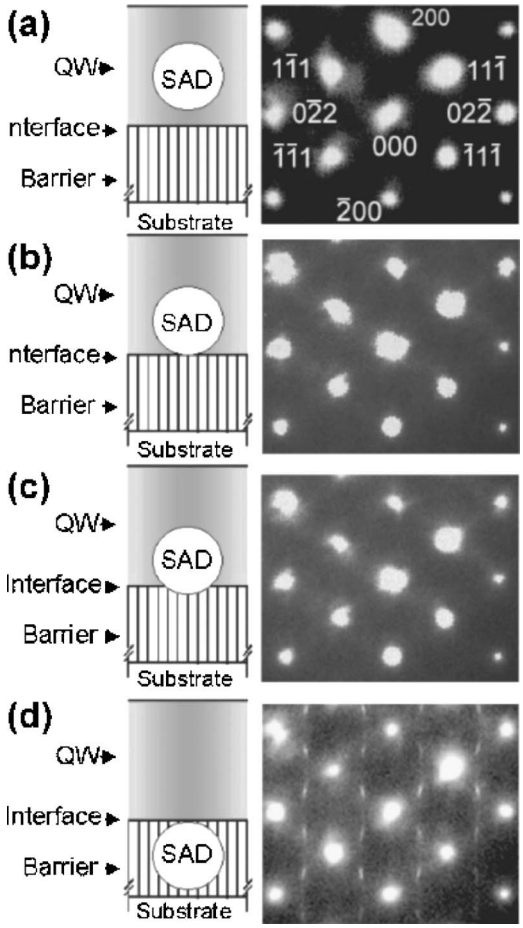

FIG. 5. SAD analysis performed on a quaternary heterostructure sample with L_z=270 nm and no top barrier layer. The selected area (white circle) is schematically drawn on the left side of (a)–(d), with the corresponding diffraction pattern on the right side. Superlattice extra spots of 1/2,1/2,1/2 family, indicating atomic ordering along 111 planes, are exhibited only in (d), where the aperture (150 nm diameter) is over the barrier material.

The origin of carrier (or exciton) localization can be associated to different sources such as potential fluctuations at the heterointerfaces, at the well and/or at the barrier materials. A well-known source of fluctuations in QWs is the interface roughness.[15] The strength of this effect depends on the length scale of these fluctuations, when compared to the extent of the exciton wave function. To better assess the interface conditions in our samples, we performed microstructure analysis on selected samples.

Figures 4(a) and 4(b) show cross-sectional HRTEM images of sample Q30 (which shows the largest energy blue shift in Figs. 2 and 3 taken along the $[0\bar{1}1]$ and [011] zone axis. Well-defined interfaces with roughness of 2 to 3 monolayers (ML) with respect to an average line (I1 and I2 arrows show positions of top and bottom interfaces, respectively) can be seen from the HRTEM image shown in Fig. 4(a). The fluctuations in the well width contribute to carrier localization in the QQWs. However, this effect is expected to be more prominent for QWs with small widths ($L_z < 10$ nm), when the exciton radius is comparable to the QW width, enhancing the contribution of the interfaces to the optical spectrum. The strong blue shift observed even for QQW samples with L_z as large as 60 nm (Fig. 3) indicate that interface roughness is not the main source of potential fluctuation in this system.

Figure 4(b) shows the corresponding image taken along an orthogonal direction, i.e., in the [011] zone axis. At this direction, a clear "zigzag" pattern is observed only in the barrier layers, a characteristic signature of spontaneously atomic ordering due to alternation of atoms of the same group (either III or V) along a same set of {111} planes.[3–5,16] In contrast, no apparent evidence of atomic ordering was found in the well layer.

In order to obtain additional information about possible ordering effects in the well material a specially devised sample (not listed in Table I) was grown. The sample consisted of a 300 nm thick GaAs buffer layer, followed by 500 nm thick quaternary bottom barrier layer and 270 nm thick of the quaternary well material. Intentionally, the top barrier was not deposited in this sample.[17] The whole structure was grown at 670 °C on (100) oriented GaAs substrate. The large thickness of the well material allowed us to probe selected regions of this sample using SAD micro analysis. Results of SAD measurements in four different areas of this sample are shown in Fig. 5. All SAD experiments were carried out with a 150 nm diameter aperture. As the SAD aperture was moved from the top (surface) to the region close to the barrier interface [Figs. 5(a)–5(c)] no clear evidence of atomic ordering was observed in the well material. In con-

trast, superlattice extra spots of $1/2,1/2,1/2$ family were observed only in the barrier material [Fig. 5(d)], consistent with the results shown in Fig. 4(b).

Previous experiments using asymmetric x-ray diffraction technique in similar QW layers revealed the presence of reduced number of weakly ordered domains, when compared to the barrier material.[7] The absence of the $1/2,1/2,1/2$ family spots in the SAD patterns of the QW layers [Figs. 5(a)–5(c)] indicates that the amount of ordering is below the detectable limit for our TEM technique. However, we have been systematically observed better defined and sharper HRTEM images of the QW layer along $[0\bar{1}1]$ zone axis, when compared to those along [011]. Such small reduction of the HRTEM image quality observed for the [011] zone axis can be tentatively interpreted as the result of an undetectable "zigzag" background pattern in the QW layer. This suggests that the total volume of ordered domains remains quite small in the QW layer, even when it is grown over a highly ordered barrier material, consistent with the asymmetric x-ray experiments.[7] As a result, QQWs investigated here correspond to ordered/disordered/ordered structures, where the term disordered is referred to a considerably less ordered material.

The evidence of spontaneously ordering in the barrier material [see Figs. 4(b) and 5(d)] suggests that this effect plays an important role in the optical emission characteristics of the QQWs. As discussed in the introduction, there are two possible physical mechanisms that could account for a LE band in the PL spectra of quaternary alloys. Besides the atomic ordering discussed above, phase separation (the formation of GaP-rich and InAs-rich domains within the alloy layer) also gives rise to a similar low-energy emission band.[2] This effect is more pronounced in the barrier alloy material which compositions lies deeply inside the spinodal decomposition isotherm diagram.[2,6,7] In the case of the well material, phase separation effects are expected to be very small. This was confirmed by a previous study[7] and by the results obtained from the well material PL spectrum (Fig. 1) that shows the presence of the relative narrow HE emission band. Phase separation decreases drastically with the increase of the growth temperature, since this favors random alloy formation. On the other hand, ordering can be very pronounced only at specific growth conditions.[8,18] Previous reports on GaInP/GaAs heterostructures, with ordered GaInP materials, revealed the presence of power excitation dependent PL emission lines that are similar to those observed in Fig. 4.[8,10,19] It is well known that GaInP ternary alloys exhibit negligible phase separation effects within the usual growth temperatures (600–700 °C).[2] In order to verify possible phase separation effects in the barrier layer on the emission characteristics of the QQWs, an additional sample was grown (T30) with exactly the same characteristics as sample Q30 but now using a lattice matched GaInP alloy as the barrier material. SAD experiments (not shown) in this sample confirmed the presence of ordering in the ternary barriers. Figure 6 shows the excitation dependent LT-PL spectra of sample T30. Similarly to Q30 [Fig. 2(c)], the PL spectra of sample T30 consisted of single excitation dependent LE emission band with total peak blue shift of

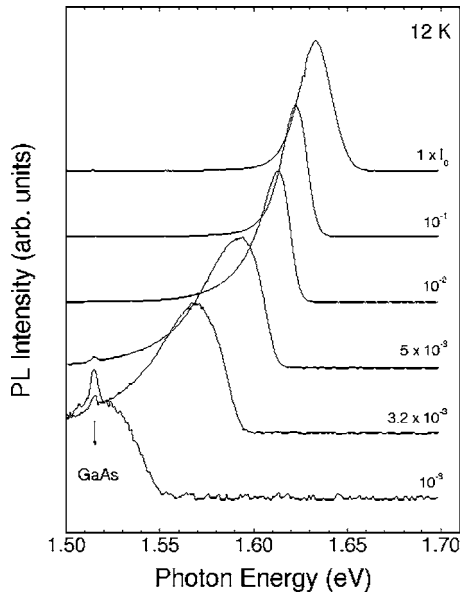

FIG. 6. LT-PL (12 K) spectra as a function of the excitation power for sample T30. Maximum excitation was $I_0=100$ W/cm^2.

~117 meV, roughly the same value obtained for sample Q30 (see also Fig. 3, solid circle). Thus, these results ruled out the hypothesis of phase separation as the driving mechanism for influencing the QW emission. Also it was not verified by HRTEM any indication of phase separation either in the barrier or in the well layers. The similarity between the PL spectra shown in Figs. 2(c) and 6 indicates that ordered quaternary (or ternary) barriers do play an important role in the PL emission characteristics of the QQWs investigated in this work.

It is well known that the degree and the lateral extension of the ordered domains depend on the growth temperature.[5,8] In the QQW system bandgap fluctuations are expected to be enhanced by an inhomogeneous distribution of domains with different size and ordering degrees, specially in the barrier material. These fluctuations change locally the material bandgap giving rise to strong localization effects. In partially ordered InGaP alloys a maximum ordering degree occurs in the growth temperature range of 660–690 °C.[8] Growth temperature values below or above this critical range resulted in layers with reduced ordering degrees, mainly for higher T_g values. We experimented with quaternary samples grown at different temperatures. Similar to the results reported for GaInP ternary alloys,[8] maximum ordering effects were observed for quaternary samples grown close to 670 °C.[7] With the purpose to investigate the effect of the barrier ordering degree on the QQWs' optical properties two samples were grown with barrier temperatures of 600 °C (Q30L) and 730 °C (Q30H). In this case, only the growth temperature of the bottom barrier was modified.[20] The well and the top barrier layer characteristics were kept identical to the sample Q30 (see Table I). The excitation-power dependent PL spectra of the samples grown at different barrier temperatures are shown in Fig. 7. Two distinct characteristics can be directly observed from this figure. First, the LE PL peak shift with increasing laser power for both samples is considerably

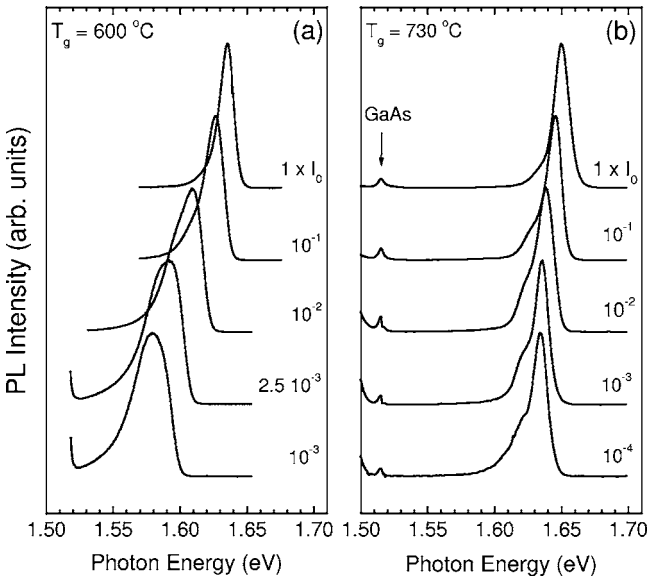

FIG. 7. LT-PL (12 K) spectra as a function of the excitation power for the samples (a) Q30L and (b) Q30H. Maximum excitation was $I_0=100$ W/cm^2.

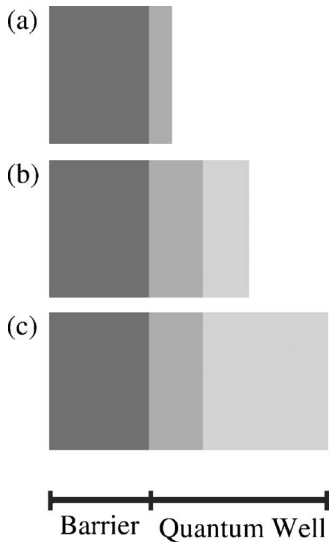

FIG. 8. Sketch of the suggested physical mechanism behind the anomalous PL behavior. (a) The bottom barrier ordering degree acts as a template for the growth of the QW material, inducing a higher ordering degree in the first QW layers. (b) after a "critical thickness," the QW material progressively decreases its ordering degree until it reaches its thermodynamic equilibrium condition. (c) After reaching an equilibrium (bulk) situation, QW material grows without further changes in its ordering degree.

smaller than that obtained for sample Q30 [Fig. 2(c)]. Total blue shifts of 55 and 16 meV were observed for samples Q30L and Q30H, respectively. Second, in the case of the sample grown with the bottom barrier at $T_g=730$ °C [Fig. 7(b)], the PL spectra were located above the well material bandgap emission for all laser excitation powers. These observations are consistent with what one would expect from the ordering behavior in semiconductor layers. Since the alloy composition of the quaternary barrier layer has low arsenic-content, one expects a change in ordering degree similar to that of the ternary InGaP alloy as a function of the growth temperature. Consequently, the ordering degree in the barriers of QQW samples grown at $T_g=670$ °C is expected to be higher than those samples grown at $T_g=600$ °C (Q30L) or $T_g=730$ °C (Q30H). Also, the quaternary barrier layer grown at $T_g=730$ °C is expected to exhibit the lowest ordering degree among all the samples investigated here.[8] The decrease in the ordering degree in the bottom barrier of samples Q30L and Q30H (Fig. 7) resulted in pronounced reduction of the PL peak total blue shift with increasing laser power levels. Moreover, a larger decrease in the barrier-ordering degree values resulted in PL recombination peaks above the well material bandgap [Fig. 7(b)]. However, it should be pointed out that localization effects were not completely suppressed by decreasing the barrier ordering degree, as clearly shown in Fig. 7(b). Alloy fluctuations (in the barrier and/or well) are probably the source of the "residual" localization effects observed in the QQW sample with the lowest ordered (bottom) barrier material. Thus, the results shown in Figs. 6 and 7 strongly suggest that the presence of ordering in the barrier material is the major source of the additional potential fluctuations in the QQWs investigated here.

Having identified ordering in the bottom barrier as the main source of potential fluctuations in the QQWs, both the microscopic physical mechanism and the effect of L_z on the magnitude of the total PL peak blue shift (Fig. 3) can be tentatively explained. We suggest a picture where the ordering in the barrier may work as a template for the growth of the less ordered well layer. As a result, the initial QW monolayers would start to grow with a slightly higher ordering degree, as compared to further QW material, but still with no significant increase on the volume of domains to be detectable under HRTEM and SAD experiments, as indicated in Fig. 8(a). After a given thickness, the quaternary well material would tend to progressively grow toward its usual (and lower) degree of ordering [Fig. 8(b)], until it reaches its own thermodynamic equilibrium. After a particular thickness, the QW layer would grow homogeneously as if it were a "bulk" material [Fig. 8(c)]. This picture coherently describes the individual and the overall PL behavior of our QQW samples. For low L_z values (5–10 nm) the exciton diameter is larger than or comparable to the well width. In this case the potential fluctuations are averaged, decreasing the mean potential barrier difference among available localized states. For the thinner QW (5 nm), the altered degree of ordering in the well is homogeneous, leading to negligible energy shift with excitation power and a slightly broadening for the PL line. At large values of L_z ($10<L_z<60$ nm) the well material would be in the process of recovering its bulk degree of ordering, which gives rise to domains with variable ordering degree along the growth direction. This leads to the observed progressive increase on the PL peak blue shifts were observed up to $L_z=30$ nm [Figs. 2(a) and 2(b)], while it starts decreasing for sample Q60 (not shown). Further increase of L_z results in layer thickness comparable to the hole diffusion length and the PL spectra of these very large QQWs exhibit bulk-like properties (Fig. 1), meaning that the well material

thickness is effectively larger than the initial ordering fluctuations. The existence of a maximum PL peak blue shift observed in Fig. 3 suggests a lateral extension of potential fluctuations (increased ordering) of ~30 nm, when highly ordered barriers are used. A direct comparison between Figs. 2(c) and 7(b) demonstrate that these fluctuations can be significantly reduced by decreasing the ordering degree of the bottom barrier material, in accordance to the suggested microscopic interpretation for this effect, as discussed above.

IV. CONCLUSIONS

Quaternary quantum wells grown on GaAs substrates with different well width, barrier composition, and bottom barrier growth temperature were investigated by low-temperature photoluminescence spectroscopy. LT-PL spectra of QQWs were dominated by emissions below the fundamental gap of the well material. The dramatic dependence of the PL peak position with the laser excitation power indicates strong localization effects in the studied samples. This anomalous localization was attributed to an increase on the ordering degree of the well material at the beginning layers of its growth, induced by the presence of well-defined atomic ordering in the bottom barrier material. HRTEM and SAD experiments confirmed the presence of strong ordering in the barriers but did not show clear evidence of this effect in the well material. Reduction of ordering degree in the barrier material was achieved by changing its growth temperature. QQWs grown with bottom barriers at 600 °C and 730 °C revealed considerable less PL emission blue shift. The significant suppression of the localization mechanisms in these samples indicates that the ordering conditions of the barrier does affect the initial growth of the quaternary quantum well. We expect that these results would stimulate first-principle calculations on similar systems in order to confirm and/or clarify the physical mechanisms that lead to our experimental observations.

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[17] During the development of this work, we have grown many additional samples for assessing different possibilities for the anomalous PL behavior. First, we inserted GaP layers at the interfaces (both, top interface only and bottom interface only), as already discussed. Second, barrier growth temperature was varied (both barriers, top layer only, and bottom layer only); PL experiments on these samples lead us to conclude that the top barrier conditions have no contribution for the PL anomalous behavior. We have even grown large "wells" (270 nm thick) without the top barrier and their emission is similar to W300 and Q270 (Fig. 1). From this point our attention was focused on the bottom interface/barrier of the QQW's.

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[20] As discussed previously, we have grown samples in which the growth temperature was varied in both barriers, in the top layer only, and in the bottom layer only. Since no influence of the top barrier temperature was observed, we focused our attention on the bottom interface/barrier influence on the optical properties of the QQw's.