Strain and electronic interactions in InAs/GaAs quantum dot multilayers for 1300 nm emission

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We have investigated the emission properties of low-growth-rate InAs/GaAs self-assembled quantum-dot multilayer samples with spacer layers of different thicknesses. For two layers with the same InAs coverage and a spacer of 40 nm, emission from the two layers is shown to be at different wavelengths. This is discussed in terms of local strain and surface undulation caused by the first layer influencing the growth of the second layer. We show that by annealing the surface before the growth of each subsequent layer this effect can be avoided for spacers of 40 nm and above. Furthermore, it is shown by collecting photoluminescence over a limited area of an *N*-layer sample, grown with surface annealing, that this sample exhibits a maximum modal gain *N* times that of a single layer. For spacers below 10 nm, only one emission peak is observed. However, we show that the observation of a single peak is due, in this case, to tunneling between adjacent layers, and that the maximum modal gain at 1300 nm of such electronically coupled layers is comparable to that of a single layer. © *2002 American Institute of Physics.* [DOI: 10.1063/1.1429797]

I. INTRODUCTION

Self-assembled quantum dots (QDs) are predicted to produce lasers with low, temperature independent thresholds. It is generally assumed that an increase in the relatively low modal gain of a single layer laser can be obtained through the use of multiple layers of QDs. Several groups have investigated the structural changes that occur when QD layers are deposited sequentially. Imaging techniques have demonstrated vertical ordering (stacking)[1–3] of the islands for thin spacer layers. The consensus is that stacking results from a strain interaction during the growth.[2,4] Stacking can also have a significant effect on the electronic properties; for small spacer layers, a redshift of the emission (compared with that of a single layer) has been reported and attributed to electronic coupling or tunneling between the QD layers.[5–10] However, blueshifts have also been reported[11–14] and attributed to enhanced intermixing of In and Ga atoms in the upper layers. Ideally, a multiple layer QD laser would contain layers with coincident emission, narrow inhomogeneous broadening and a high dot density. In addition, we would like to place as many of these layers close to an antinode in the optical electric field to maximize the confinement factor of the lasing mode. Realizing such a system is not a trivial problem. It is therefore desirable to optimize the design of the active region prior to the time consuming and expensive processing and packaging stages required to create a laser. Since the main goal of using multiple layers is to increase the maximum achievable ground state (GS) gain (which is necessary to obtain lasing from the GS with smaller cavity length), it is important to be able to assess easily this property. The maximum GS gain is obtained when all the dots have their GS fully occupied (i.e., with two electrons and two holes), and this saturated gain is therefore directly proportional to the number of dots emitting at the target wavelength per unit surface. Once the design of the active region is optimized, it is also important that its properties are not affected by the growth of the upper structure needed in a real laser diode. For example, the temperature of growth of the AlGaAs layers have to be limited to $\approx 600\,^{\circ}\mathrm{C}$ in order to avoid any annealing effect on the QDs properties.

In this work, we describe measurements made on a series of samples containing multiple layers of low-growth rate QDs designed for 1300 nm applications with a range of spacer thicknesses. It is shown that the emission from these structures depends not only upon the spacer thickness *d* but also on its surface morphology, which was imaged using atomic force microscopy (AFM). Even for relatively large spacers (at least up to 60 nm), a strong blueshift of the emission from the second layer is observed. For spacers equal to or larger than 30 nm, this blueshift can be avoided by flattening the surface prior to growth of the second layer through annealing. The results are interpreted in terms of strain interactions with the underlying dot layer during the growth of the subsequent layer. A variable pump wavelength (VPW) photoluminescence (PL) technique is used to identify the emission from different layers. Limited-area PL (LAPL) is then used to compare the number of QDs emitting at the target wavelength (and therefore the maximum modal gain at that wavelength) for samples exhibiting a single emission peak. The observation of one peak in samples with small spacers (10 nm) is shown to be the result of carriers tunneling between layers. The maximum GS modal gain is therefore not always increased in stacked layers, which demonstrates that care has to be taken in designing the active region of a QD multilayer laser.

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TABLE I. Samples investigated in this work.

Sample	Layers	Spacer d (nm)	Annealed
A	2	40	No
B	2	40	Yes
C	2	20	Yes
D	2	10	No
E	1	N/A	N/A
F	3	40	Yes

II. EXPERIMENTAL DETAILS

All samples were grown by solid-source molecular beam epitaxy (MBE) on epiready n^+ (Si-doped) (001) GaAs substrates in a Vacuum Generators V80 MBE machine. After oxide removal, a 150 nm GaAs buffer layer was deposited and annealed for 10 min at a substrate temperature of $T_S = 580\,^{\circ}\mathrm{C}$. Each QD layer was grown by deposition of 2.3 monolayer (ML) of InAs at $T_S = 495\,^{\circ}\mathrm{C}$, a background As pressure of $(2.2 \pm 0.2) \times 10^{-7}$ mbar and a growth rate of 0.01 ML s^{-1}. These growth conditions result in the formation of relatively large dots emitting at the technologically important wavelength of 1300 nm (at room temperature) with a small full width at half maximum (FWHM) of ≈ 25 meV.[15,16] However, the QD density is relatively low ($\leqslant 10^{10}$ cm^{-2}) making the use of multilayers necessary. The islanding transition was monitored with reflection high energy electron diffraction (RHEED) along the $(1\bar{1}0)$ azimuth. The dots are then capped with 10 nm of GaAs at $T_S = 495\,^{\circ}\mathrm{C}$. For samples A and D, the entire spacer layer was grown at $T_S = 495\,^{\circ}\mathrm{C}$ and this was immediately followed by the deposition of the second QD layer. For samples B, C and F, only the first 10 nm of the spacer layer is grown at 495 °C and the temperature is then increased at $\approx 1\,^{\circ}$ C s^{-1} to 580 °C for the remainder. The surface is annealed at this temperature for 10 min before deposition of the second layer. Annealing the surface was seen to change the ''streaky'' RHEED pattern to a well-defined Laue circle of diffraction spots, suggesting a more ordered surface had been obtained. The second dot layer was grown and capped with 10 nm of GaAs at 495 °C followed by a further 90 nm at 580 °C. The two dimensional to three dimensional transition time in the second layer was indistinguishable from that of the first layer, except for sample D where a small reduction of $\approx 10\%$ was measured. Table I summarizes the structure of the samples studied in this work. PL measurements were made at a low temperature (10 K) using an Ar^+ or HeNe laser, dispersing the light with a SPEX 0.5 m monochromator and detecting with a cooled Ge diode. AFM images were taken using a Burleigh AFM in constant-force mode.

III. RESULTS AND DISCUSSION

A. Strain interactions

Ideally, a laser containing N layers of dots would have N times as many dots emitting at the wavelength of interest, and N times the maximum modal gain at that wavelength.[17] However, this is true only if the QDs in each layer have the same density and emission wavelength. Figure 1 shows low temperature PL spectra obtained from sample A under low excitation. This sample consists of two QD layers separated by a relatively large GaAs spacer ($d = 40$ nm). The peak seen at 1.05 eV is also observed in the spectrum of a single QD layer grown under identical conditions. However, another well-resolved peak is observed at higher energy (1.2 eV). To identify the origin of these peaks, we measured PL spectra with either a HeNe laser (penetration depth ≈ 260 nm) or an Ar^+ laser (penetration depth ≈ 100 nm). The use of two lasers of different wavelengths (VPWPL technique) alters the ratio of photoexcited carriers captured by each layer, and hence the relative emission intensities of the layers.[14] As the Ar^+ laser creates a higher concentration of carriers near the surface, it will favor emission from the upper QD layer and we can unambiguously assign the higher energy peak to the top layer. The upper layer, although grown under the same conditions as the first one, is strongly blueshifted (by ≈ 150 meV) and this must be a result of growing above an existing QD layer. Such a strong effect is surprising considering the relatively wide spacer (40 nm). Lipinski *et al.* have reported similar blueshifts[13] but only for smaller spacers (20 nm) and we attribute this to differences in QD size. In our case, the islands in the first layer are twice as large as those studied in Ref. 13.

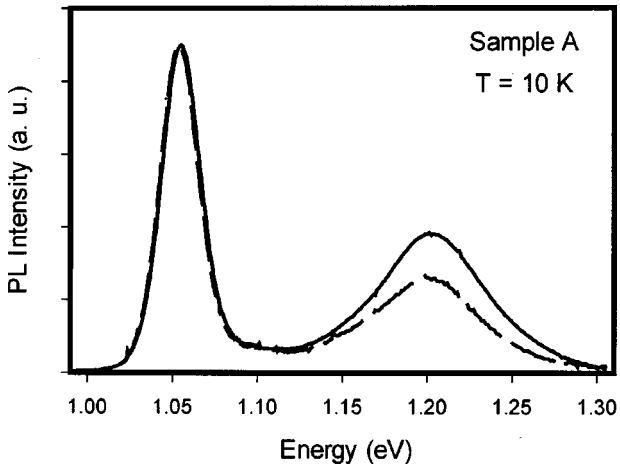

FIG. 1. Low temperature PL spectra excited with an Ar^+ laser (solid line) or HeNe laser (dashed line) from sample A consisting of two layers of dots separated by an unannealed spacer of 40 nm. The excitation density is very low (≈ 0.1 W cm^{-2}) to avoid any emission from the excited states.

It is well known that the strain field from an underlying dot layer can give rise to a variation in the surface energy across a sample[18–21] and some groups have also reported a strain-induced undulation in the surface of the spacer layer.[22–24] To investigate these effects and the resulting blueshift, we obtained AFM images of the surface prior to growth of the second layer. Figure 2(a) shows the uncapped QDs in the first layer; the islands are ≈ 7 nm high with a base diameter of ≈ 40 nm. Figure 2(b) shows an AFM image of the surface after growth of a 40 nm spacer layer at 495 °C above the first QD layer. We observe ''ridges'' measuring ≈ 150 by 50 nm in the plane of the surface, oriented along the $(1\bar{1}0)$ direction and 2–3 ML high. The density of ridges is of the same order as the density of dots measured in the first layer.

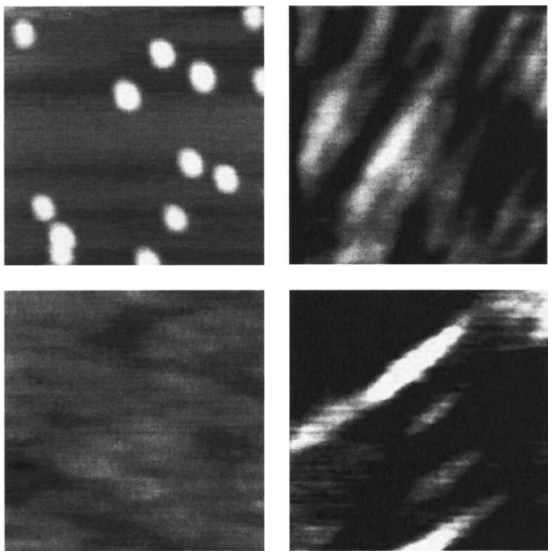

FIG. 2. 500×500 nm AFM images of (a) an uncapped single layer of QDs. The dots are 40 nm across and 7 nm high. (b) The GaAs surface on which the second QD layer is grown in the case of an unannealed spacer of 40 nm (sample A). We observe elongated ridges, 2–3 ML high, with a density similar to that of dots in the first layer. (c) Same as (b) after annealing the surface at 580 °C for 10 min. The surface is much flatter and the ridges have disappeared. (d) Same as (b) for a 10 nm annealed spacer. Ridges are still observed.

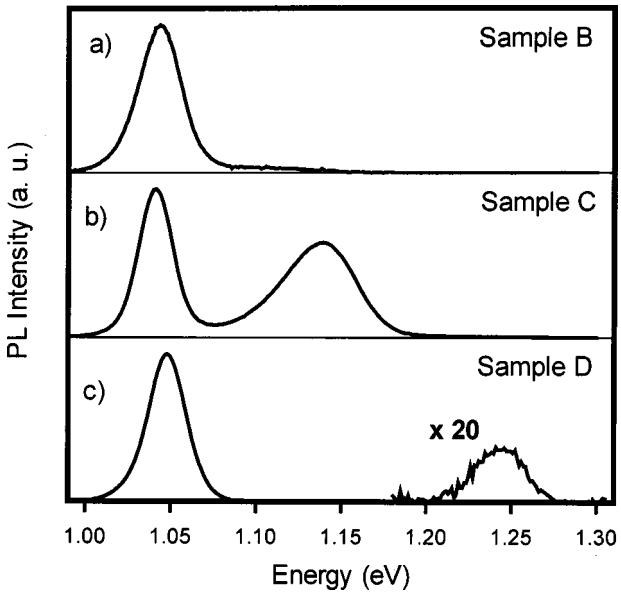

FIG. 3. Low temperature (10 K) and low excitation (0.1 W cm^{-2}) PL spectra excited with the Ar^+ laser for samples B, C, and D.

They must therefore be the result of nonuniform growth of the spacer layer induced directly by the strain field of the buried dots, or clusters of dots, in the layer beneath. Similar features were observed by Joyce *et al.*[24] for QDs of similar dimensions and spacers (40 nm). The anisotropy can be attributed to enhanced migration of Ga adatom along $(1\bar{1}0)$ during capping.[24] The strain modulation and surface undulations prior to the growth of the second layer are the main cause of the strong blueshift. Schmidt and Eberl[19] have argued that the strain modulation enhances Si/Ge intermixing during the growth of a second QD layer and such effects are also likely to occur in the InAs/GaAs system. Stronger In/Ga intermixing during the growth (and possibly the capping) of QDs in the second layer, due to strain/surface modulation, could then explain the observed blueshift.

To avoid this interaction, we can increase the spacer thickness *d*, but a strong blueshift is still observed for $d = 60$ nm. Such large spacers are unsuitable for many device applications where it would limit the active region to a single QD layer. Another possibility is to attempt to alter the state of the surface on which the dots grow through annealing. Figure 3(a) shows the PL spectrum of sample B, identical to sample A except that the surface was annealed for 10 min at 580 °C before the growth of the second QD layer. A single emission peak at 1.04 eV with a narrow FWHM of 28 meV is observed, similar to what is seen for a single layer. We conclude that the second layer was not affected during growth by the presence of the lower QD layer and that the two layers in sample B are identical (this is confirmed later using LAPL). Wasilenski *et al.*[25] have also used an annealing stage to modify the growth surface for multiple QD layer samples, although these authors argued that the purpose of this procedure was to evaporate the indium atoms which had segregated onto the surface from the layers beneath. In our case, the main effects of surface annealing are related to strain and surface morphology. Indeed, the AFM image of the annealed spacer of sample B before growth of the second layer [Fig. 2(c)] shows that the ridges have disappeared, and the surface appears flat. This confirms that the annealing procedure has the effect of removing any surface undulation and possibly the strain modulation. Combined with the PL results, it also confirms that the blueshift arises from a strain/surface modulation.

For some applications, it is desirable to have spacer layers less than 40 nm. However, using the annealing procedure described above, for a spacer of $d = 20$ nm (sample C), the PL spectrum still exhibits two peaks [see Fig. 3(b)]. Although the peak from the second layer is less blueshifted than without the annealing procedure, it is not enough to obtain coincident emission. This means that when the spacer layer is thin, the annealing procedure we apply does not yield a sufficiently flat surface. This is confirmed by the AFM image of a 10 nm annealed spacer [Fig. 2(d)], which presents a surface similar to that seen for a 40 nm unannealed spacer [(Fig. 2(a)]. This also demonstrates that the main effect of the annealing procedure is not to evaporate segregated indium atoms, since it should also work for small spacers. It is possible that increasing the annealing temperature of the surface increases the mobility of the Ga atoms sufficiently to obtain a flatter surface, but we then risk blueshifting the emission from the first layer.[26,27] Alternative procedures are currently under investigation.

The results presented here suggest that there are metastable surfaces for layers grown on QD layers. Although there have been several reports of surface modulation,[24,28,23] this study shows that this can have a significant effect on the emission energy of QD layers. Blueshifts of the emission in bilayer samples have previously been attributed to enhanced In/Ga intermixing in the second QD layer induced by tensile

strain immediately above each island.[13,19] We cannot exclude such an explanation for sample A in the present study, but this then raises the interesting question of how annealing can alter the strain state of the surface. The AFM images show that the surface is undulating before growth of the second layer but is flatter following the annealing. The undulations are a consequence of the strain field extending from the dots in the first layer. For large spacers ($d \geqslant 40$ nm), it is unlikely that this strain field extends all the way across the spacer. The undulations observed are therefore probably due to kinetic effects during growth of the spacer and do not represent the most energetically favorable state of the surface. The annealing step enables the surface to reach its preferred state which is a flat unstrained surface. For smaller spacers, the strain field of the underlying layer extends throughout the spacer and the annealing step is not sufficient to flatten the surface. We conclude that, due to the strain field of the dots beneath, the energetically most favorable state of the surface is in this case an undulating surface. This limits the narrowest spacer that can be used and this value is determined by the dot size in the first layer. These results suggest that the blueshift in the PL emission for spacers is intimately linked to the presence of surface undulation.

Finally, when decreasing the spacer further to $d = 10$ nm without the annealing procedure (sample D), we see in Fig. 3(c) that one narrow dominant peak (FWHM of 26 meV) is observed in the PL spectrum. Strong strain interactions between the layers mean it is unlikely that the two layers are identical and we attribute the observation of a single peak to carriers tunneling between the layers and recombining in the layer which contains dots of the lowest energy.

B. Gain and electronic coupling

To investigate this further, we used LAPL. This technique involves probing a fixed, microscopic area on the sample. Previously, this technique has been used to investigate the radiative lifetimes of excited states[29] and many-particle effects under high excitation[30] but we use it here to compare the number of QD ground states emitting at 1300 nm in different samples. To reduce the probed area, it is possible to etch small mesas[30] or to define a metallic mask on the sample surface.[29] In our case, we coated a quartz wafer with a photolithographically defined Ti/Au layer containing an array of widely spaced holes of diameter 60, 110 and 210 μm, which could be placed metal layer down on any sample. Pumping occurs through one of these holes. Carriers may diffuse away from the probed area but any resulting emission will not be detected. When studying the excitation density dependence of the PL, it is expected that the GS emission will saturate when the dots are completely filled (2 $e-h$ pairs per dot). The inset to Fig. 4 shows the excitation dependence of the PL intensity measured at the GS peak for normal PL and LAPL with a 110 μm hole. Although we observe a sublinear dependence for the case of normal PL, a complete saturation is not observed. As the power increases, more and more carriers diffuse outside the central region. We then see a spectrum which includes excited state emission from those dots in the center of the beam which capture a high concentration of photoexcited carriers but also a large contribution to the GS peak from dots at the periphery of the pump beam, which are not saturated. Using LAPL, a complete saturation of the GS signal and then of the higher excited states is observed. Figure 4 shows the PL spectra obtained under the same excitation density for conventional PL and LAPL collected through holes of different diameters for a single layer (sample E). As expected, much more level filling is obtained from the LAPL spectra, reflecting the nonuniformity of the excitation and the lateral diffusion effects.

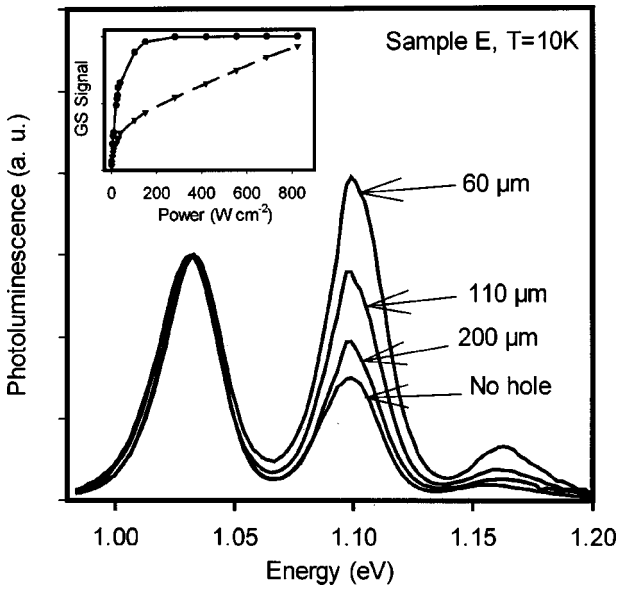

FIG. 4. Low temperature PL spectra of a single QD layer (sample E), at an excitation of 150 W cm^{-2}, collected and excited either conventionally or through circular holes of different diameter (LAPL). The intensities are normalized to the GS peak at 1.04 eV. Increasing level filling is clearly observed as the probed area is reduced. The inset shows the GS intensity from PL (solid line) and LAPL through a 110 μm hole (dashed line) as a function of the pump intensity.

In a LAPL experiment, the GS saturated intensity I_{sat} is proportional to $2N/\tau$, where the factor of 2 accounts for the degeneracy, τ is the radiative lifetime, and N is the number of dots contributing to the signal.[29] The maximum achievable gain at the target wavelength (GS saturated gain) is also proportional to N and, by comparing I_{sat}, it is therefore possible to compare the relative GS saturated gain that would be obtained from these multilayer structures were they placed in a laser diode. Figure 5 shows the LAPL spectra obtained at increasing excitation from sample E (single layer), F and D. Looking first at E and F, we observe very similar spectral shape, but the PL intensity is different. The saturation of the GS signal is clearly seen in both cases at high excitation and we can easily extract the relative values of I_{sat}. We measure a factor of ≈ 3 enhancement in I_{sat} for sample F compared to E. This confirms that the three layers have nearly identical optical properties, and that the maximum GS gain is increased by a factor of 3. We conclude that the surface annealing prior to growth of subsequent layers is an effective technique to obtain identical layers for 1300 nm emission with spacers of 40 nm.

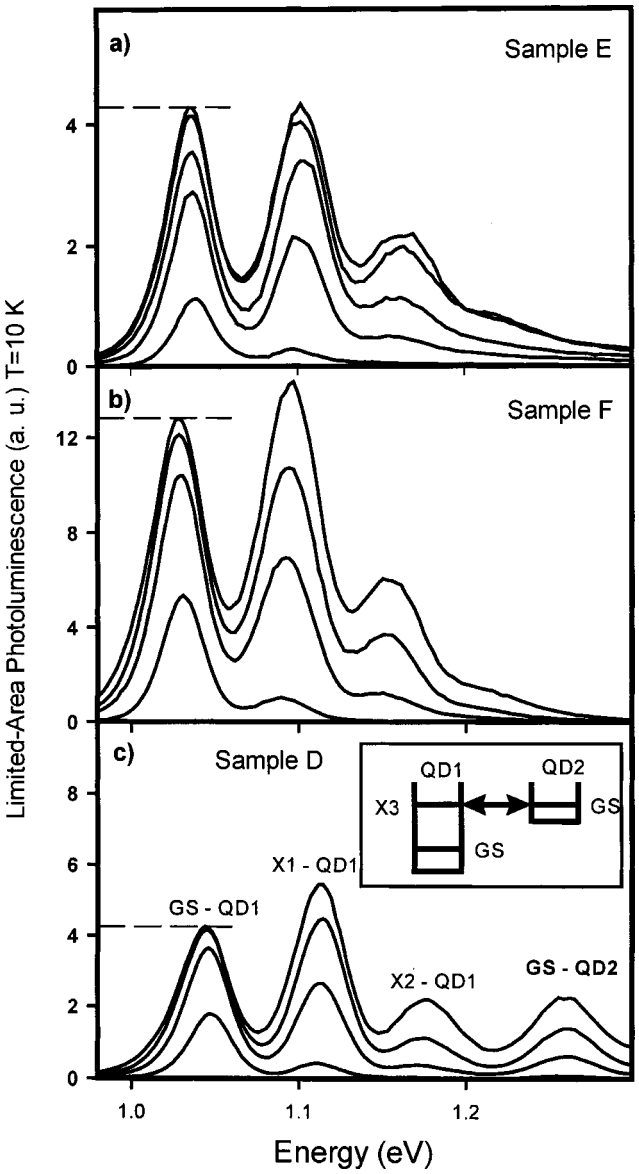

FIG. 5. Low temperature LAPL spectra collected through 110 μm holes at increasing excitation densities for: (a) sample E, (b) sample F, and (c) sample D. The absolute PL intensity scale is arbitrary but reflects the relative emission intensities for each sample. The saturated intensity of the ground states, marked with a dotted line, is the same for samples E and D, and is three times larger for sample F.

We now focus on sample D. We saw in Fig. 3(c) that this bilayer sample exhibits a single peak under low excitation. However, Fig. 5 shows that the value of I_{sat} is comparable with that of the single layer (sample E), implying that the number of dots contributing to GS emission at 1300 nm is the same in these two samples. Moreover, under high excitation, the spectral shapes are quite different. When the GS saturates, we observe in addition to the first and second excited state signals, another peak at higher energy ($\approx$1.25 eV) for sample D. This feature is absent in the high excitation spectra of sample E. Interestingly, the weak feature seen in the low excitation PL spectrum of sample D [Fig. 3(c)] appears at the same energy. These results can be interpreted as follows: we have seen that for intermediate spacers, the GS emission of the upper layer is strongly blueshifted. There is no reason why this effect should vanish when the spacer is further decreased. However, because the vertically aligned dots are very close to each other, there is a possibility that carriers can tunnel between them. Under low excitation, carriers are captured in both layers, but can tunnel very rapidly between the GS of a dot in the second layer and the third or fourth excited states of a dot below in the first layer (all these states having a similar energy around 1.25 eV) as illustrated in the schematic of Fig. 5(c). When a carrier is present in the GS of a dot, its radiative lifetime is typically between 500 ps and 1 ns. However, carriers in the third excited state of a dot from the first layer can undergo fast relaxation to lower states (on the order of 1 ps). Therefore, providing that tunneling times are fast compared to radiative lifetimes, most carriers are expected to relax to the lower states of the first layer, eventually radiatively recombining in the GS. This explains why a dominant peak is observed at low excitation, with only a weak signal from the GS of the dots of the upper layer. When the level of excitation increases, the GS emission (at 1.04 eV) from sample D saturates at the same level as a single layer, because it only originates from recombination in the QDs of the first layer. At higher excitations, the lower states of the dots in the first layer are filled, blocking the fast relaxation channel from the third excited state. The carriers then have more time to tunnel back to the GS of the upper layer where they can eventually recombine radiatively, explaining the signal observed around 1.25 eV for large excitations (which is not present in sample E). We conclude that, although the PL spectrum of sample D shows a single, narrow peak, the number of states emitting near 1.04 eV (1300 nm at room temperature) for such electronically coupled layers is not increased compared with a single layer. The same study on a similar sample with five QD layers and $d=10$ nm exhibited exactly the same behavior (not shown). Only the first layer contributed to the GS emission, and a very large signal was observed under high excitation from the four other layers. Such samples with small spacers, where the QDs in upper layers are different from the QDs in the first, are therefore inadequate for applications where a large gain is required. We have also modeled the dynamics of carriers in two coupled layers with simple rate equations. The predicted ratio between the two peaks under low excitation (much less than one electron–hole pair per dot) is equal to

$$R=\frac{I_2}{I_1}=\frac{\tau_r}{\tau_2}+\frac{\tau_t}{\tau_2}\frac{g_2}{g_1+g_2}, \tag{1}$$

where τ_2 is the radiative lifetime in the GS of the QDs in the upper layer (which is on the order of 500 ps), τ_r is the relaxation time from the (electronically coupled) third excited state to lower states in QDs of the lower layer, and τ_t is the tunneling time between the two coupled states. g_1 and g_2 are the generation rates in lower and upper layers, which in a first approximation we assume to be equal. From Fig. 3(c), we measure a ratio between the integrated intensities of the two peaks of $R\approx 1/64$. This ratio is independent of excitation under such low powers. From Eq. (1), if $\tau_t \ll \tau_r$, then $R \approx \tau_r/\tau_2$, leading to $\tau_r \approx 8$ ps, which is a reasonable value. If $\tau_r \ll \tau_t$, then $R\approx \tau_t/(2\tau_2)$ leading to $\tau_t \approx 16$ ps, equivalent to a coupling energy on the order of $\pi\hbar/(2\tau_t)\approx 0.12$ meV

(splitting of 0.24 meV). These figures are again reasonable considering the relatively large spacer of 10 nm. Although it is difficult to distinguish between the two possibilities in this case, the calculation demonstrates that it is in principle possible to extract information on the coupling and the dynamics of carriers from simple PL experiments. Further experiments and theory are also needed to know which type of carriers (electrons or holes) are predominantly tunneling.

Finally, comparing the spectra obtained from the three samples in Fig. 5 under the lowest excitation, when the GS is unsaturated, we can note that the PL intensity is larger when more QD layers are present (one for E, two for D and three for F). This is due to the fact that more layers can capture more photoexcited carriers. This aspect can be interesting for the case of electronically coupled layers (like sample D). Although the GS maximum gain is not increased, the presence of the upper layers can improve the collection and capture of carriers, which can then tunnel and relax to the first layer GS. Inserting such an active region in a laser structure, the transparency current density would not be larger than for a single layer and the improved capture of carriers should then lead to a reduction of the threshold current.

IV. CONCLUSION

We have studied the emission properties of multilayer QD samples grown at a low growth rate for 1300 nm emission at room temperature. The strain modulation and surface undulation caused by the dots in the first layer results in a large blueshift of the second layer emission, even for relatively large spacers (up to $\approx$60 nm). We have demonstrated a simple annealing procedure to reduce the spacer layer between QD layers while still obtaining coincident emission from all layers at the desired wavelength. LAPL was used to show that a sample containing N layers of dots separated by an annealed 40 nm spacer has N times as many dots emitting at 1300 nm as a single layer, and that a laser based on such an active region should therefore have N times the maximum modal gain. The same technique was used to demonstrate that the GS gain of multiple closely spaced (10 nm) QD layers is equal to that of a single layer. The properties of such samples can be explained in terms of tunneling between excited states of the dots in the first layer and the ground states of dots in the upper layers.

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