Tuning of polarization sensitivity in closely stacked trilayer InAs/GaAs quantum dots induced by overgrowth dynamics

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Abstract
Tailoring of electronic and optical properties of self-assembled InAs quantum dots (QDs) is a critical limit for the design of several QD-based optoelectronic devices operating in the telecom frequency range. We describe how fine control of the strain-induced surface kinetics during the growth of vertically stacked multiple layers of QDs allows for the engineering of their self-organization process. Most noticeably, this study shows that the underlying strain field induced along a QD stack can be modulated and controlled by time-dependent intermixing and segregation effects occurring after capping with a GaAs spacer. This leads to a drastic increase of the TM/TE polarization ratio of emitted light, not accessible from conventional growth parameters. Our detailed experimental measurements, supported by comprehensive multi-million atom simulations of strain, electronic and optical properties, provide in-depth analysis of the grown QD samples allowing us to give a clear picture of the atomic scale phenomena affecting the proposed growth dynamics and consequent QD polarization response.

Keywords: quantum dot stacks, optoelectronics, polarization response, strain, electronic properties, TM/TE ratio

1. Introduction

Since their theoretical proposal in 1981 in the seminal paper of Arakawa and Sakaki [1], semiconductor self-assembled quantum dots (QDs) have inspired an extremely large quantity of experimental studies aimed at the employment of such nanostructures in a variety of optoelectronics devices, due to their intriguing atomic-like electronic properties. The flexibility in controlling their geometry by means of growth conditions and, in certain cases, substrate patterning [2] has stimulated several studies on more complex structures able to add further potentialities to lens-shaped or pyramidal nanostructures commonly obtained by the Stranski–Krastanov process. In this context, one relevant physical example is closely stacked quantum dots, either consisting of QD layers separated by a thin GaAs spacer [3–5] or without any GaAs spacer (also known as columnar QDs [6] or quantum posts [7]). In these kinds of nanostructures, the strong compressive biaxial strain component at the centre of a typical flat shaped dot can be reduced to zero or towards tensile values by increasing the stack height (adding QD layers), thus providing the optical polarization insensitivity desirable for relevant technological applications such as semiconductor optical amplifiers for high speed communication networks [8–10]. Such a requirement cannot be fulfilled by standard flat dome-shaped QDs, where the
baxial compressive strain induces valence band splitting into heavy hole and light hole states, thus providing a strongly TE polarized optical transition [11]. This connection between quantum dots’ electronic structure and their morphology highlights the importance of complex and interplaying phenomena occurring during growth and overgrowth of this kind of nanostructure.

Normal QD growth and capping procedures are lattice mismatched heteroepitaxial processes where strain release, segregation, faceting, intermixing and strain-enhanced diffusion may occur at the island/cap interface [12, 13]. We recently demonstrated how the actual single layer QD composition profile is relevant in determining the real optical behaviour of these InAs-based nanostructures [14]. This, in turn, is determined by complex nanoscale phenomena such as In–Ga intermixing and atomic segregation occurring during overgrowth with GaAs. In the case of multi-layer stacked QD devices, these phenomena become even more complex, due to the underlying strain field related to buried QDs, and the consequent QD shape/composition is more difficult to assess, even with the most reliable and sensitive experimental investigation techniques. All these effects generate a complex scenario involving kinetics and thermodynamics but, at the same time, represent strategic tools for engineering of the structural and optical properties which these nanostructures can exhibit.

So far, several QD engineering solutions have been explored [6, 15–18] for both InAs/GaAs and InAs/InP material systems, in order to change their optical properties by varying their aspect ratio and/or by controlling the biaxial strain in QDs thanks to strained barriers. The shape anisotropy has been engineered either by embedding a single layer of QDs into InGaAs strain reducing layers [16, 17] or by closely stacked QDs (CSQDs) [15] and columnar QDs (CQDs) [10] formed by several repetitions of ML-thin InAs/GaAs layers. In particular, the last two QD geometries have shown a significant enhancement of TM-mode photoluminescence [3, 5, 8, 19] due to a reduction of the biaxial strain in the centre of the CQDs and CSQDs.

In this work, we show the engineering of shape anisotropy and related polarization sensitivity of Stran ski–Krastanov closely stacked InAs/GaAs QD layers by overgrowth phenomena and surface kinetics. The analysis reported is based on photoluminescence (PL) measurements at low temperature (10 K) as a function of excitation power, and at room temperature as a function of polarization state, and atomic force microscope (AFM) investigations and is theoretically supported by multi-million atom calculations of strain, electronic structure and optical spectra. Our results suggest that different QD reorganization processes related to longer post-growth interruption times and consequent different surface strain conditions can be an alternative tool to tune the actual polarization behaviour in InAs nanostructures and to obtain TM/TE polarization ratios as high as 0.8.

2. Experimental details

The self-assembled QD structures used in this study were grown on semi-insulating GaAs substrates by a COMPACT 21 Riber molecular beam epitaxy (MBE) system equipped with a reflection high energy electron diffraction (RHEED) gun to monitor in situ the surface evolution during growth. After the growth of a GaAs buffer layer at 600 °C, the substrate temperature was lowered down to 500 °C and QDs were formed by covering this buffer with 2.8 MLs of InAs. The 2D–3D transition after deposition of ~1.7 MLs was demonstrated by the RHEED pattern evolution from streaky-like to spotty-like. Afterwards, dots were immediately capped by a GaAs spacer layer grown at the same low temperature. A single layer QD sample, labelled sample 1, was grown as reference, with a 20 nm GaAs cap terminating the structure. The stacked samples for polarization investigation were realized by growing three QD layers closely spaced by 5 nm of GaAs. For all samples the final GaAs capping layer was 20 nm thick, whereas for the formation of QDs in the second and third layers only 2 MLs of InAs were deposited, because the high strain field coming from the underlying QDs reduces the critical layer thickness of the stacked layers [12]. In our experiments, we focused on the evolution of QD shape and composition with respect to the growth interruption time (GIT) used before the deposition of the second and third QD layers on the GaAs thin spacer. Such a step was always performed in As-rich conditions, with a duration of 20 s (labelled sample 2) or 120 s (labelled sample 3). The resulting polarization behaviour of the multi-stacked structures was found to be heavily affected and tunable by this feature.

In order to experimentally study this effect, the stacked samples were lithographically processed to define apertures of 400 μm diameter inside a metal thin film (40 nm Ti/120 nm Au), thermally evaporated on the sample surface. The apertures were designed to optically pump a limited area of the samples, while the metal cover blocked the emission from the top of the sample, thus providing a consistent and reliable measurement of the polarization state of the edge emitted signal [20]. The PL signal excited from the top with an Ar+ laser (λ = 514 nm) collected from the [110] cleaved edge surface of the samples was filtered by a linear polarizer and detected by an NIR CCD positioned at the end of a 320 mm spectrometer.

3. Theoretical models and simulations

In order to provide an understanding of the experimental measurements, we performed multi-million atom simulations of the electronic and optical properties of the grown QD samples and compared the calculations with the experimental results. The simulations were performed using the well-known NanoElectronic MOdeling (NEMO 3D) simulator [21, 22], in which the strain is computed from the atomistic valence force field (VFF) model [23–25] and the electronic structure is computed by solving a twenty-band sp3d5s∗ tight-binding Hamiltonian [26]. The polarization-dependent interband optical transition strengths (TE and TM modes) are calculated using Fermi’s golden rule [3, 27]. Furthermore, the calculated polarization-dependent TE and TM modes are cumulative sums of the interband optical transition strengths between the lowest conduction band state and the highest five
for a stacked QD layer configuration, where the buried QD ratio), as shown in figure 1.

The present analysis is based on our recent study [14], where we described the dependence of the polarization-dependent optical emission on the actual chemical composition of QDs for a single QD layer, as modulated by atomic scale phenomena occurring during overgrowth, i.e. In–Ga intermixing, strain-enhanced diffusion and In segregation. The effect of these phenomena has been accurately described from small variations of QD dimensions, etc. It should be noted here that theoretical modelling of QD stacks possesses a two-fold challenge. Firstly, it requires modelling techniques with atomistic resolution that can calculate electronic structure and optical properties including the correct symmetry and interface roughness. Secondly, the relatively large size of QD stacks requires calculations to be performed over several millions of atoms to properly include the long-range effects of strain. Our atomistic simulations performed over about 18 million atoms in the device take both factors into account and therefore are capable of explaining experimental data with a high degree of accuracy, allowing us to identify the main atomic scale mechanism behind the observed polarization response of the investigated samples.

4. Experimental results and discussion

The present analysis is based on our recent study [14], where we described the dependence of the polarization-dependent optical emission on the actual chemical composition of QDs for a single QD layer, as modulated by atomic scale phenomena occurring during overgrowth, i.e. In–Ga intermixing, strain-enhanced diffusion and In segregation. The effect of these phenomena has been accurately described only with a complex QD compositional model (shown in the left inset of figure 1) having an In-rich core region surrounded by a lower In content external shell, which actually allows one to fit the experimental data in terms of both optical emission (PL peak wavelength) and polarization response (TM/TE ratio), as shown in figure 1.

A more complex atomic scale dynamics is expected for a stacked QD layer configuration, where the buried QD layers strongly modify subsequent QD growth with a highly localized strain field originating at each QD apex [12, 13]. In this scenario all the involved growth parameters, and among them the GIT, can play a crucial role in the assessment of actual QD structure, stoichiometry and resulting optical polarization behaviour.

4.1. Impact of GIT on QD stack morphology and optical properties

InAs QDs undergo several atomic scale phenomena when overgrown with GaAs cap layers [28, 29]. The evolution of these phenomena can be correlated to the morphology of the grown QD samples, as shown in [28]. Therefore, in order to obtain a realistic picture of the morphology of the grown QD samples, we performed AFM analysis on the top GaAs surfaces of samples 1, 2 and 3 respectively, (shown in figure 2). As expected, the underlying QDs lead to a mound-like morphology in the top surfaces of all the samples. In the reference single QD layer (sample 1), a mound density of $4 \times 10^{10}$ cm$^{-2}$ is measured, with average height and diameter of 1.5 nm and 50 nm, respectively (figure 2(a)). Such morphology directly reflects the dot size and distribution observed in the uncapped single QD layer sample (figure 2(d)).

A significant change is observed as the interruption time (GIT) is increased from 20 s (sample 2) to 120 s (sample 3) in the two stacked samples. In sample 2, the mounds are rather well organized, forming oriented and elongated chains, with an average length of 100 nm and an average lateral size of 30–40 nm (figure 2(b)). Elongation along the [110] direction is usually observed during initial QD overgrowth by GaAs or other compounds [28, 30, 31] as an effect of anisotropic surface diffusion, since Ga adatoms tend to migrate far away from the high stress regions. After a few monolayers, such an elongation is usually no longer evident, rather leading to an undulating GaAs surface morphology prevailing up to a capping thickness of the order of a few tens of nanometres [28]. The persistence of the elongated morphology after 20 nm of GaAs capping in sample 2 suggests that buried stacked QDs do not have an ideal cylindrical symmetry. Conversely, in sample 3, the overgrown GaAs morphology (figure 2(c)) is very similar to that of the single layer sample (figure 2(a)) apart from an increase of mound lateral size up to 60 nm. Moreover, for this long GIT an uncapped three QD layer sample was also grown for comparison. The AFM analysis on this sample (figure 2(e) and related diameter histogram) clearly shows that the QD size distribution and density reproduce those of a single layer (figure 2(d) and related diameter histogram).

PL spectra collected at 10 K for samples 1, 2 and 3 show that the QD ground state (GS) emission blue shifts in both the stacked samples with respect to the single layer sample. A line-width shrinking of the GS spectrum with the stacking is also observed: from 31 meV in the single layer to 24 meV in sample 2 and to 19 meV in sample 3, suggesting a wide size distribution in the reference sample, whereas a size filtering effect occurs among the vertically coupled QD layers [32].
The blue shift of the GS emission with the stacking at thin spacers (in the strong quantum coupling regime) with respect to single QD layers was found in previous studies and was generally attributed either to strain-driven In–Ga intermixing [15, 33, 34] or to a reduced wave function overlapping along the stacked layers [35]. Actually, the variation of the GIT parameter in our growth can promote the time-dependent In out diffusion phenomenon from the buried QDs. This, in turn, modifies the nucleation dynamics of subsequently grown QD layers, since the exposed surface conditions are consequently altered. Therefore, different localization effects of electron or electron/hole wave functions can be expected in the final QD stack, finally modifying the resulting QD polarization response and enhancing the TM/TE ratio.

By the analysis of low temperature PL spectra as a function of optical pumping power density (figure 3), performed by multi-Gaussian curve fitting, it can be noted that the sample 1 (single QD layer) PL spectrum exhibits (figure 3(a)) the clear presence of the ground state (GS) and the first excited state (ES) as a function of excitation power, separated by a large energy spacing (~80 meV), typical for flat single QD layers [36]. Similarly, the PL spectrum of sample 2 (figure 3(b)) presents a second peak at the high energy side, spaced by 58 meV from the GS, and exhibiting the band filling behaviour with increasing power density expected from a first excited state. On the other hand, in sample 3 (figure 3(c)) a shoulder is evident at the high energy side, at about 21 meV from the GS emission. The relative intensities of the two lowest energy peaks were found to not be dependent on the excitation density. Therefore, we associated this additional emission with the anti-bonding state emission due to the QD molecule formation [33, 37, 38], demonstrating stronger wave function hybridization in this sample with respect to sample 2.

The most striking effect of employing a different GIT is highlighted in the polarization responses of the two stacked samples. Figure 4 shows the measured polarized RT PL spectra of samples 2 and 3, taken under the same experimental conditions. While the single QD layer sample is strongly TE polarized (TM/TE = 0.26) as shown in figure 1, the polarization ratio is progressively increased in sample 2 ($\rho = 0.62$) and in sample 3 ($\rho = 0.8$) where longer growth interruption times are employed. This demonstrates the possibility of tuning the polarization response towards the isotropic value (TM/TE ~ 1.0) in QD nanostructures by controlling the overgrowth process, without growing a large number of stacked QD layers which can increase the overall strain of the structure.

4.2. Discussion

The QD formation and their GaAs overgrowth, at such a high substrate temperature (510°C), are well known [13, 28–31] to result from the complex interplay between In–Ga intermixing at the dot/barrier interface, In vertical segregation through the dot height and In desorption out of the dot and back to the vapour phase. Considering the observed AFM morphology (figure 2(d)) and previously performed TEM investigations [33], buried QDs in the first layer are probably left partially uncapped by the overgrowth
with a 5 nm GaAs spacer. At this stage, different growth interruption times after the spacer growth and before further InAs coverage can cause a modification of the strain-induced surface conditions, thus altering subsequent QD formation.

When the growth interruption is delayed after capping, it is likely that both In–Ga intermixing inside the buried nanostructures is enhanced and In atoms from the partially uncapped apex of buried dots tend to out-desorb in a relatively large, time-dependent amount. Therefore, the stress induced on the reconstructed surface by the lattice mismatch underneath is lowered and during the overgrowth with the second QD layer the migration length of impinging In adatoms is consequently increased. In general, strain-driven migration leads In adatoms to diffuse towards the buried QD positions which represent favourable nucleation centres for the second and third QD layers, in both GIT cases. However, the time-dependent In–Ga intermixing in the buried dots slightly reduces the underlying strain field, and a short surface migration length of In adatoms is expected. In this case stacked QDs are enlarged in size with a low degree of cylindrical symmetry. Indeed, the AFM image of the GaAs capping surface over the 3 QD layers shows an elongation enhancement during the overgrowth. Moreover, in sample 2 the increase in lateral size along the stack is also reflected in the weaker overlapping of electron wave functions and the absence of bonding and anti-bonding transitions in the luminescence spectra.

For a longer interruption time, as for sample 3, increased In out diffusion in the buried dots produces nucleation sites with a lower binding energy for the In adatoms provided for the second and third layer growth. This leads to a QD formation dynamics more similar to that of the first QD layer with respect to the lower GIT case, resulting in stacked QDs with morphology closer to the buried ones (dome shaped). In this case, stacked QDs should be characterized by a more pronounced cylindrical symmetry, as confirmed by the AFM results for the third uncapped layer which also exhibits the same size distribution and density as the first QD layer (figures 2(d) and (e)). Such shape and size preservation, in turn, leads to the reduced line-width broadening of sample 3 (with respect to sample 2) and reduced low temperature blue shift PL emission (with respect to a single layer). Moreover, the ‘columnar’ shape of the stacked QDs in sample 3 gives the observed presence of bonding and anti-bonding states in sample 3 (GS_b and GS_a), as evident from figure 3(c).

5. Numerical analysis based on atomistic simulations
A better understanding of the proposed dynamics can be obtained by exploiting multi-million atom simulations that
allow us to correlate dot shape/morphology and consequent optical and electronic properties, with a particular focus on strain field and polarization sensitivity.

5.1. Theoretical modelling of QD stack confined wave functions as a function of GIT

Firstly, we performed our atomistic simulations to evaluate the impact on the wave function confinement of the different QD evolutions along the stack related to the GIT employed. For this purpose, we simulated the following two QD systems, based on the analysis of AFM investigation.

MS1. A stack composed of three vertically stacked layers of dome-shaped QDs, with increasing diameter and decreasing height: the lowest QD has diameter of 15 nm and height of 5 nm (as in [14]), the middle QD has diameter of 20 nm and height of 4 nm, and the top QD has diameter of 25 nm and height of 3 nm.

MS2. A stack comprised of three identical vertically stacked dome-shaped QDs (as observed in sample 3 with GIT 120 s), with equal diameters of 15 nm and heights of 5 nm.

In both cases, MS1 and MS2, all the QDs are placed on top of 0.5 nm thick InAs wetting layers.

Figure 5 gives an insight into the evaluated electron confined states (E1, E2, and E3) for the two proposed QD systems (MS1 and MS2). In this figure the electron wave function spatial distribution and symmetry can be visualized by side and top view maps. Energy difference values between electron states (E2–E1 and E3–E2) are also indicated for the investigated configurations.

For the trilayer QD stack with GIT = 120 s (MS2) shown in figure 5(a), the QD layers are strongly coupled and, therefore, the lowest two states E1 and E2 show the formation of bonding and anti-bonding states, respectively, separated by a relatively smaller energy difference of 22 meV. In this system, the first excited (p) state is the third state E3 which is further separated by 40 meV from E2. This is
In order to quantitatively explain the observed increase in the TM/TE ratio for sample 3 (figure 3(c)) where bonding and anti-bonding type transitions are observed separated by only 21 meV, validating the assumption of shape preservation and strong wave function localization along the QD vertical stack.

5.2 Theoretical modelling of electronic and optical properties of long GIT stacked QDs

The hybridization of wave functions becomes significantly weak when the QD base diameter is assumed to increase along the vertical direction (MS1) as in the case of sample 2 (figure 5(b)). For this stack, our calculations indicate that E1 (s-state) is weakly hybridized and predominantly in the upper QD region. The next state E2 is the first excited state of p-type symmetry and is separated by 38 meV from E1, while the E3 state is the second s-state separated by only 4 meV from E2. This finding is in agreement with both excitation dependent, low temperature PL measurements (figure 3(c)) and the elongated morphology evidenced by AFM analysis shown in figure 2(b). The two proposed stacking models (MS1 and MS2) are characterized by different strain profiles, as shown in figure 6 where we plot line scans of hydrostatic ($\varepsilon_{xx} + \varepsilon_{yy} + \varepsilon_{zz}$) and biaxial ($\varepsilon_{xx} + \varepsilon_{yy} - 2\varepsilon_{zz}$) strain components through the centres of both trilayer stacks. Figure 6(a) compares the hydrostatic components, clearly showing a decrease in the magnitude of the hydrostatic strain for the larger upper QDs (QD2 and QD3) in case of MS1 (sample 2). This will result in deeper conduction band edges for these QDs [39] and therefore the electron wave function will largely reside inside the larger QDs towards the top of the stack, in consistency with the wave function plots of figure 5(b). The strong electronic coupling of QDs for the MS2 case accompanied by roughly equal magnitudes of the hydrostatic strain in the three QDs (due to their equal size) leads to strong hybridization of the electron wave functions forming bonding and anti-bonding states.

Figure 6(b) compares the biaxial strain components for the two trilayer stacks MS1 and MS2, indicating an opposite trend: the larger size of QDs in MS1 leads to an increase in the magnitude of the biaxial strain component (as shown by arrows in figure 6(b)). This will result in larger splitting between the heavy hole (HH) and the light-hole (LH) band edges for the larger QDs [39], thereby reducing the TM-mode component. The equal size of QD layers for MS2 favours the hole wave functions residing inside the bottommost QD layer. Finally, a much reduced biaxial strain for MS2 (GIT = 120 s case) as compared to MS1 (GIT = 20 s case) is expected to significantly impact on the polarization-dependent optical properties of this stack.

In agreement with what we found from excitation-dependent PL measurements for sample 3 (figure 3(c)) where bonding and anti-bonding type transitions are observed separated by only 21 meV, validating the assumption of shape preservation and strong wave function localization along the QD vertical stack.

The preliminary assumption was that each QD layer had identical composition of both the outer shell and inner core. The considered compositions are listed in table 1 (double compositions 1–6). For an outer shell In composition of 0.4 and an inner core In composition of 1.0, as found for the single layer sample [14], the values of GS and the TM/TE ratio are 0.98 eV and 1.047, respectively, thus not fitting the measured values (1.057 eV and 0.8, respectively). The agreement between theory and experimental results can be improved by lowering the inner core composition to 0.9 and 0.8, which blue shifts the GS to 1.07 eV and decreases the TM/TE ratio to 0.88. Additional modifications of this assumption to get the TM/TE ratio closer to its experimental value, such as unrealistically small values of In composition in the core region, obviously increase the GS energy. On the other hand, any decrease in the outer shell composition (double compositions 4–6 in table 1) to blue shift the GS expected In/Ga intermixing effect was mimicked by using the two-composition model with a high In composition QD core surrounded by a lower In composition shell. We assigned different compositions to each region of the QDs as shown by the schematic of figure 7. The dimensions of the high In core region were also kept fixed to 11 nm height and 4 nm diameter in accordance with [14].

Our analysis of the single QD layer predicted the inner core of QDs with In content close to 1.0 and the outer shell with lower In content ($\leq 0.4$) [14]. Therefore, we performed a set of systematic simulations as a function of QD compositions while keeping $x_1, x_3, x_5 \geq 0.8$ and $x_2, x_4, x_6 \leq 0.4$, and compared our computed values of the GS and TM/TE ratio with the corresponding experimentally measured ones.

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Table 1. The calculated values of the ground state peak energy (GS) and the TM/TE ratio are provided for various double composition models (as shown by the schematic diagram of figure 7) mimicking In/Ga intermixing effects in sample 3. The GS and TM/TE values that are closest to the experimental values of GS ≈ 1.057 eV and TM/TE ≈ 0.8 are highlighted by using bold font.

| In composition configuration no | Outer shell compositions | Inner core compositions | GS (eV) | TM/TE ratio |
|--------------------------------|-------------------------|-------------------------|---------|-------------|
| Double composition 1           | 0.4                     | 1.0                     | 0.98    | 1.047       |
| Double composition 2           | 0.4                     | 0.9                     | 0.914   | 0.96        |
| Double composition 3           | 0.4                     | 0.8                     | 1.07    | 0.88        |
| Double composition 4           | 0.3                     | 1.0                     | 0.97    | 1.329       |
| Double composition 5           | 0.3                     | 0.9                     | 1.02    | 1.218       |
| Double composition 6           | 0.2                     | 0.8                     | 1.08    | 1.046       |
| Double composition 7           | 0.4                     | 0.3                     | 1.0     | 0.98        |
| Double composition 8           | 0.4                     | 0.3                     | 0.9     | 1.03        |
| Double composition 9           | **0.4**, **0.3**, **0.2**| **0.85**               | **1.05**| **0.82**    |
| Double composition 10          | 0.4                     | 0.3                     | 0.8     | 1.07        |
| Double composition 11          | 0.4                     | 0.3                     | 0.9     | 1.02        |
| Double composition 12          | 0.4                     | 0.3                     | 0.9     | 1.04        |
| Double composition 13          | **0.4**, **0.3**, **0.2**| **0.8**                | **1.06**| **0.88**    |
| Double composition 14          | **0.4**, **0.3**, **0.2**| **0.75**               | **1.08**| **0.83**    |
| Double composition 15          | 0.4                     | 0.3                     | 0.8     | 1.06        |

Figure 7. Schematic diagram of the trilayer quantum dot stack. Each quantum dot layer is comprised of a dome-shaped QD placed on top of a 0.5 nm thick InAs wetting layer. The composition profile of the QDs is chosen according to the two-composition model to mimic the In–Ga interdiffusion effect, where the inner core (red region) is of higher In composition (>0.8) and the outer shell (green region) is of lower In composition (<0.4). The size of the inner core region (red region) is 11 nm is diameter and 4 nm in height and is the same for all three QDs. The QD stack is embedded inside a very large GaAs buffer consisting of roughly 18 million atoms.

As a second step, we introduced a composition gradient also in the inner core of the QDs, while keeping the outer shell composition gradient of 0.1. The long GIT employed in sample 3 is likely to enhance In out diffusion and vertical segregation in buried QDs and, consequently, a vertically increasing gradient could be reasonably expected for the inner core composition. The corresponding simulated composition profiles are listed in table 1 (double compositions 11–14). A systematic increase of the GS energy along with a lowering of the TM/TE ratio was calculated as the average inner core composition was lowered from 0.95 to 0.85, with the best fit with experimental measurements found for the composition models 13 and 14.

The three double composition models that provided the best match with the experimental data (lines 9, 13 and 14 in table 1) were also used to recalculate the electron wave function spatial distributions and symmetries (as shown in figure 8). In all these three cases, the presence of bonding and anti-bonding states, consistently with experimental PL (figure 3(c)), is confirmed, in agreement with what was calculated earlier using a pure InAs composition profile (figure 4(a)). The presence of bonding and anti-bonding states in sample 3 can then be inferred to the strong coupling between identically sized QD layers inside the stack.

value is accompanied by very large values of the TM/TE ratio, as expected from the resulting reduced strain field in the nanostructures.

Therefore, also based on the fact that the localized strain induced by the lower QD layers should lead to reduced In out diffusion in the subsequent QD layers, we have introduced a vertical gradient in the composition profiles of the QD stack.
Figure 8. Plots of the lowest three conduction band states (E1, E2 and E3) calculated for the double composition models 9, 13 and 14 (table 1). In each case, we show side views of the electron wave functions as well as the top views in the bottom left corners. The presence of strong bonding and anti-bonding states is clearly evident in all three cases, which conforms with the PL measurements performed on sample 3 (figure 3(c)).

Furthermore, we also calculate and plot the highest three hole wave functions (H1, H2, and H3) in figure 9 for the three double composition models under study. All the hole wave functions are found to be confined in the bottommost QD layer. We also note from the comparison of the top views of the hole wave functions that the variations in the In/Ga intermixing only result in very small changes in the orientations of the hole wave functions.

Overall, we conclude that the In/Ga intermixing effect, regulated by the surface kinetics induced by underlying strain field, only slightly affects the actual confinement and symmetry of the electron and hole wave functions in such a QD stack, whereas it is the crucial parameter impacting the related polarization response, giving us the possibility to obtain a high TM/TE ratio even with only a triple QD layer stack.
Figure 9. Plots of the highest three valence band states (H1, H2 and H3) calculated for the double composition models 9, 13 and 14 (see table 1). In each case, we show side views of the hole wave functions as well as the top views in the top right corners.

5.3. Theoretical modelling of electronic and optical properties of short GIT stacked QDs

Based on our analysis of the AFM images (figure 2) and the electronic state simulations (figure 5), we have shown that the short GIT employed in sample 2 is likely to induce a lateral size increase in the stacked QDs, as commonly observed for closely spaced QD stacks. The model MS1 has already shown a reduction of the hydrostatic strain and a reinforcement of the biaxial strain towards the stack top (figure 6). The TM-mode intensity is therefore expected to decrease for the GIT = 20 s sample, as it is directly related to the intermixing of HH and LH character in the valence band states which reduces due to the larger magnitude of the biaxial strain thereby increasing the separation between the HH and LH bands [36]. In order
to estimate only the impact of the QD lateral size along the stack (thus ignoring the In/Ga intermixing effect), we applied atomistic calculations to calculate TM/TE ratios for the two model systems presented above (MS1 and MS2). We found a TM/TE ratio of only 0.48 for MS1 and of 0.84 for MS2. This difference is in agreement with the trend of polarization-dependent PL measurements shown in figure 4, where the TM/TE ratio for sample 2 is found to be much smaller (0.62) as compared to the value of 0.8 for sample 3. Therefore, we conclude that the lateral size of the QD layers inside the stack, controlled by the strain-induced surface dynamics, has a very strong impact on the polarization ratio.

Furthermore, since from our AFM analysis we would expect a much weaker In/Ga intermixing for sample 2, we again applied the two-composition model shown by the schematic diagram of figure 7. Due to the increasing lateral size of the QD layers in sample 2, we proportionally increased the size of the inner core. Based on our understanding of the much reduced intermixing for sample 2, we assumed 5% In composition in the outer shell region, i.e. \( x_2 = x_4 = x_6 = 0.05 \), while the inner core compositions \( (x_1 = x_3 = x_5) \) were varied from 1.0 to 0.9. A corresponding systematic increase in the peak GS energy from 0.95 to 1.01 eV with a TM/TE ratio ranging from 0.55 to 0.62 was found, in a good quantitative agreement with the experimental findings. This behaviour indicates that the vertical composition profile along the stack has a weaker effect in sample 2, whereas the QD shape plays the dominant role in controlling the spatial distribution of wave functions and their overlaps.

6. Conclusions

In this work we describe how the growth of vertically stacked QDs is influenced by the atomic scale phenomena of intermixing, segregation and out diffusion, affecting the surface kinetics of further provided In adatoms. The proposed dynamics was investigated by combining experimental investigation of QD morphological and optical properties with atomistic numerical calculations giving a clear picture of wave function localization and strain field distribution along the stacked structure. The QD structural and optical properties were found to be affected by the interruption time after buried QD capping with GaAs, which plays a relevant role in determining the strain energy conditions of the surface exposed to stacked QD layers. As a result, the QD self-organization process allows a strong increase of the TM/TE polarization ratio of the emitted light to be achieved by only three closely stacked QD layers, which is promising for technological applications based on QD optoelectronic devices.

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