The polarization response in InAs quantum dots: theoretical correlation between composition and electronic properties

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Abstract

III–V growth and surface conditions strongly influence the physical structure and resulting optical properties of self-assembled quantum dots (QDs). Beyond the design of a desired active optical wavelength, the polarization response of QDs is of particular interest for optical communications and quantum information science. Previous theoretical studies based on a pure InAs QD model failed to reproduce experimentally observed polarization properties. In this work, multi-million atom simulations are performed in an effort to understand the correlation between chemical composition and polarization properties of QDs. A systematic analysis of QD structural parameters leads us to propose a two-layer composition model, mimicking In segregation and In–Ga intermixing effects. This model, consistent with mostly accepted compositional findings, allows us to accurately fit the experimental PL spectra. The detailed study of QD morphology parameters presented here serves as a tool for using growth dynamics to engineer the strain field inside and around the QD structures, allowing tuning of the polarization response.

(Some figures may appear in colour only in the online journal)

1. Introduction

Semiconductor nanostructures are being more and more applied in several optoelectronics technologies ranging from lasers [1] to optical amplifiers [2] or single-photon sources [3] where they have successfully overcome critical challenges such as extremely low threshold, high speed response, or entangled photon emission, respectively. In most of these applications a crucial parameter is the polarization response, typically measured in terms of the degree of polarization [DOP = (TE−TM)/(TE+TM)] [4, 5] or the TM/TE ratio [6]. Understanding how this feature is related to the quantum dot (QD) structural symmetry and composition could be very helpful in its tuning. For example, semiconductor optical amplifiers (SOAs) for telecommunications require engineered QDs for isotropic polarization behavior [2, 4, 5]. Polarization control is also crucial for other applications, such as polarization-entangled photons emitted by single QDs [3] and polarization sensitive applications of vertical cavity surface emitting lasers [7, 8].
The structural and electronic properties of III–V QDs epitaxially formed by the self-ordering Stranski–Kruskalov (SK) process are strongly affected by surface and growth conditions. Several techniques such as surface structuring [9], nanostructure engineering via strain coupling [4–6, 10, 11], and tuning of growth conditions [12] are currently being studied for controlling the QD distribution and geometric symmetry. As a common trend, epitaxial InAs nanostructures exhibit asymmetry not only in the growth plane (with respect to the [110] and [110] directions) [4, 5] but also in the vertical plane (along [001]) [13] due to their preferential flat, lens shape. The vertical confinement leads to a strong compressive biaxial strain suppressing the light hole (LH) component in the valence band states, leaving mostly the heavy hole (HH) component and resulting in predominantly in-plane (TE) polarization emission from the interband transitions [14].

InAs QDs obtained via the SK process have been found to be significantly influenced by In–Ga intermixing and In segregation effects during the capping and post-growth annealing processes [15–18]. Several composition profiles have been proposed and different investigation techniques employing high resolution transmission electron microscopy, x-ray diffraction, photoelectron microscopy or scanning probe microscopy have been used to exactly map InAs nanostructures [19–21]. As recently reviewed by Biasiol and Heun [22], results in the literature do not lead to a unified model, where the actual composition profile of the QDs is strongly related to the growth conditions. Their review highlights a common tendency that the chemical composition of a typical SK QD has gradients along both the growth and the in-plane directions: the In composition increasing from base to top due to In segregation effects and decreasing from the center towards the edges in the lateral directions due to In–Ga intermixing effects. However, such a complex structure has not been considered so far in efforts to theoretically understand the polarization response of InAs QDs.

Previous theoretical [4, 10, 23] studies of QD polarization response are based on a pure InAs type QD composition profile, thus significantly limiting their accuracy and leading to discrepancies between theory and experiment. A clear example of such a discrepancy is the failure to reproduce the large values of the experimentally measured DOP by using both \( \mathbf{k} \cdot \mathbf{p} \) [10, 23] and atomistic tight binding methods [4]. Thus a better understanding of the correlation between the QD structure and the degree of polarization remains an outstanding challenge.

In this paper, the polarization response of InAs QDs is theoretically studied by means of atomistic simulations, introducing a compositional model capable of fitting experimental measurements both of the electronic transitions and of the TM/TE ratio. The actual complex composition and geometry of SK InAs QDs is mimicked by using a two-composition QD model, reproducing the experimentally measured polarization behavior of a single QD layer and highlighting the relevance of atomic scale processes like segregation and intermixing.

2. The experimental procedure

The QD samples 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 for monitoring the surface evolution in situ during growth. After growth of a GaAs buffer layer at 600 °C, the substrate temperature was lowered to 500 °C and QDs were formed by covering the buffer with 2.8 MLs of InAs. The 2D–3D growth mode transition is demonstrated by the RHEED pattern evolving from streaky-like to spot-like after deposition of 1.7 MLs of InAs. Afterwards, dots were immediately capped by a GaAs spacer layer grown at the same low temperature. The single-layer QD sample was then covered with a 20 nm GaAs cap terminating the structure. An uncapped single-QD-layer sample was also grown under the same conditions and its morphology analyzed by atomic force microscopy (AFM), providing a dot density of \( 3.5 \times 10^{10} \) dots cm\(^{-2}\) with average height of 5 nm, as shown in figure 1(a). The GaAs capped QDs were estimated by transmission electron microscopy (TEM) to have a dome-like shape with base diameter \( \approx \)15 nm and height \( \approx 5 \) nm (see the inset above figure 1(a)).

For investigating the polarization behavior, samples were excited from the top with a cw Ar\(^+\) laser (\( \lambda = 514 \) nm). The room temperature photoluminescence (PL) signal from the cleaved edge of the samples was first collected by a long focal length lens (200 mm) and it was then filtered by a linear polarizer and focused by a second lens into the monochromator.

3. The theoretical model

The theoretical modeling is performed using NEMO 3D [24, 25]. NEMO 3D is an atomistic simulator based on the valence force field (VFF) method [26] for strain calculations and the twenty-band sp\(^d\)\(^3\)s\(^*\) tight binding model [27] for the electronic structure. NEMO 3D is a multiscale simulator capable of performing multi-million atom simulations for realistic QD dimensions surrounded by large GaAs buffers to model the long range impact of the strain and piezoelectric potential. This tool has already been used to model QD structures, providing results in good agreement with experimental data [4, 5, 28–30]. Piezoelectric potentials, both linear and quadratic, are calculated by solving Poisson’s equation according to a published recipe [30] and are included in the calculations of the electronic spectra. The interband optical transition strengths are calculated using Fermi’s golden rule, with the squared magnitude of the optical matrix elements found by summing over the spin degenerate states. The polarization dependent TE and TM spectra are calculated along the [100] and [001] directions respectively, as a cumulative sum of optical transitions between the lowest conduction band energy level (E1) and the highest four valence band energy levels (H1, H2, H3, and H4), where each transition strength is artificially broadened by multiplication with a Gaussian distribution centered at the wavelength of the transition [4]. The highest four hole energy levels are
chosen here instead of just the top most level (H1), because the valence band states are closely spaced on the energy scale (H1–H4 ≤ kT ≈ 26 meV at T = 300 K) and multiple hole states contribute to the ground state optical emission peak at the room temperature [5].

4. Model 1: uniform compositions

We start our analysis by first assuming a uniform material profile throughout the QD geometry as in previous theoretical studies of the polarization response [4, 5, 10, 23, 31, 32]. Quantum dots are assumed to be lens-shaped with the base diameter and height set at 15 nm and 5 nm, respectively, in accordance with the experimental TEM analysis [33]. Figure 1(b) plots the experimental normalized PL spectra taken under the two polarization conditions (TE and TM) and compares them with the theoretically calculated spectra for both the ideal case of a pure InAs QD and an In0.7Ga0.3As random alloy quantum dot. The InGaAs alloy configuration has been suggested by some previous theoretical studies [23, 31, 32] to mimic the In–Ga intermixing effect.

The pure InAs QD model clearly fails to reproduce the experimentally measured spectra. The TM/TE ratio is calculated to be 0.097, which is significantly lower than the experimentally measured value of 0.265. The peak wavelength in the PL spectra, hereafter referred to as ground state wavelength (GSW), is also overestimated by 123 nm. This is consistent with the earlier theoretical studies [4, 10, 23] where a pure InAs QD type model predicted TE-dominant optical emissions and failed to match the experimentally measured PL spectra. Lowering the average In composition inside the QD to 70% to mimic the effect of In–Ga intermixing during the capping process increases the TM/TE ratio to 0.115 due to the reduced biaxial strain, but at the same time blue shifts the GSW by 72 nm with respect to the experimental value of 1226 nm. To further investigate the impact of lowering the In composition of the QD, we gradually reduce its value from 100% (pure InAs QD) to 55% (an alloyed In0.55Ga0.45As QD), the GSW decreases drastically whereas the TM/TE ratio only slowly increases. The experimental values of the GSW and TM/TE ratio are marked as dashed lines. Clearly a single In composition in the uniform composition model cannot be used to simultaneously reproduce experimental values for the GSW and TM/TE ratio.

Several experimental [21, 22, 34–38] and theoretical [15, 16, 39, 41, 42] studies have shown that In–Ga intermixing and In segregation effects significantly influence the chemical composition of QDs during the growth of the GaAs capping layer. Thus we hypothesize that a quantitative agreement between theory and experiment for both the GSW and TM/TE ratio. Therefore, a more complex compositional model is required in order to understand the experimental results.

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5. Model 2: non-uniform compositions

In order to get the most reliable configuration, as close as possible to the experimental one [22], a simplified double-region scheme is adopted, consisting of an In-rich inner core of $\text{In}_{0.2}\text{Ga}_{0.8}\text{As}$ (x2 typically $\geq 80\%$) material, surrounded by an In-poor thin region of $\text{In}_{0.3}\text{Ga}_{0.7}\text{As}$ (x1 typically $\leq 40\%$), as schematically shown in figure 2(a). These ranges for the compositions x1 and x2 have been chosen from previous experimental results [21, 34]. The actual size of the In-rich core, defined by the inner region base width ($B$) and height ($H$) as shown in figure 2(a), strongly depends on the growth dynamics of the capping layer. The overall sizes of QD obtained from TEM [33] places constraints on the values of $B$ and $H$: $H \leq 5 \text{ nm}$ and $B \leq 15 \text{ nm}$. On the basis of this QD model, we perform a systematic investigation by changing the parameters $B$, $H$, x1, and x2 and comparing the corresponding GSW, TE, and TM modes with the experimental measurements. Finally, it is worth noting that the two cases studied earlier in the uniform composition model, an InAs QD and an In$_{0.7}$Ga$_{0.3}$As QD, can be included by this two-composition model by assuming x1 = x2 = 100% and x1 = x2 = 70%, respectively.

6. Variations of $B$ and $H$

First, in order to investigate the impact of the parameters $H$ and $B$ on the polarization response, we choose x2 = 100% and x1 = 40%. Such an assumption is motivated by considering that the central core of InAs nanoislands is always reported as being In-rich and that the investigations reported so far indicate that the maximum value for the In composition can reach 100% [22].

Figures 2(b) and (c) plot the TM/TE ratio and the GSW, respectively, as a function of the aspect ratio ($AR = H/B$) of the inner core. The target experimental values are shown as horizontal dotted lines. The TM/TE ratio in figure 2(b) linearly increases with $AR$ when $H$ is increased for a fixed value of $B$. This is because an increase in $H$ will result in a reduction of the biaxial strain close to the center of the QD which will reduce the splitting between the HH and LH bands. As a result, the magnitude of the TM mode will increase, thus increasing the TM/TE ratio. However, our calculations show that the slope of the TM/TE ratio as a function of $AR = H/B$ is different for different values of $B$. In general, it decreases as $B$ increases, indicating a decreasing impact of $H$ on the TM/TE ratio. This is consistent with a previous theoretical study [43] where hole energy levels in smaller QDs were found to be more sensitive to changes in QD AR as compared to those in larger QDs. We want to highlight here that our theoretical calculations show that even changes of 1–2 nm in $B$ or $H$ values, which can actually be induced by highly controlled growth dynamics, result in a drastic change in the polarization properties of the QD sample.

The plots of GSW as a function of $AR$ in figure 2(c) exhibit a linear dependence of GSW on $H$ for a fixed value of $B$ (shown by the solid lines), as well as a linear dependence of the GSW on the value of $B$ for a fixed value of $H$ (shown by the dotted lines). The analysis of figures 2(b) and (c) indicates that two sets of values for ($B$, $H$), namely (10 nm, 4 nm) and (11 nm, 4 nm), give transition wavelengths and a TM/TE ratio close to the experimental values. A slight change in the sets of values of $B$ and $H$ ((10 nm, 5 nm) and (9 nm, 5 nm)) leads to a GSW value close to the experimental one, but introduces a discrepancy in the corresponding TM/TE ratios. We therefore choose ($B$, $H$) to be (10 nm, 4 nm) and (11 nm, 4 nm) to analyze in the following sections the impact that varying $x_1$ and $x_2$ has on the calculated values of the GSW and TM/TE ratio.

7. Variation in $x_1$

The outer shell composition $x_1$ was assumed above to be 40% in the study of the effect of the $B$ and $H$ parameters on the
Figure 3. (a) Plots of the TM/TE ratio as a function of the compositions \(x_1\) and \(x_2\) as described in the QD schematic of figure 2(a). The plots are for two dimensions of the core region: \((B, H) = (10 \text{ nm}, 4 \text{ nm})\) and \((11 \text{ nm}, 4 \text{ nm})\). The arrows indicate an increase/decrease of the TM/TE ratio with respect to a decrease in the compositions \(x_1\) and \(x_2\). (b) Plots of the GSW as a function of the compositions \(x_1\) and \(x_2\). The arrows indicate increase/decrease of the GSW with respect to a decrease in the compositions \(x_1\) and \(x_2\).

8. Variation in \(x_2\)

Next, we analyze the impact of the last unknown parameter \(x_2\) in our proposed two-composition model (see figure 2(a)), keeping \(x_1 = 40\%\). Figures 3(a) and (b) plot variations in the TM/TE ratio and GSW, respectively, when \(x_2\) is decreased from 100% to 90% and 80%. Contrary to the case for \(x_1\), a decrease in \(x_2\) has similar impacts on the TM/TE ratio and GSW: both decrease as the composition \(x_2\) decreases. From figures 2(b), (c)), these trends imply that the values of both the TM/TE ratio and GSW will become closer to the experimental values only for the two dimensions (11 nm, 5 nm) and (10 nm, 5 nm). However, the GSW for the dimensions (10 nm, 5 nm) is very close to the experimental GSW whereas the TM/TE ratio for this dimension is significantly different from the experimental value. Hence a combined agreement of both the GSW and TM/TE ratio with the experimental values is not possible for a decrease in \(x_2\). For example, at \(x_2 = 90\%,\) the GSW decreases to 1187 nm (≈34 nm below the experiment value) whereas the TM/TE ratio is still 0.31 (≈0.045 above the experimental value). For the \((B, H) = (11 \text{ nm}, 5 \text{ nm})\) case, if \(x_2\) decreases to 90%, the GSW and TM/TE ratio decrease to 1208 nm and 0.27, respectively, which are in reasonable agreement with the corresponding experimental values of 1221 nm and 0.265.

We complete our analysis of QD compositions by considering simultaneous decrease in \(x_1\) and \(x_2\) and finding that their cumulative impact on the GSW and TM/TE values is roughly equal to the sum of their individual effects. We find that the TM/TE ratio is more sensitive to \(x_2\) as compared to \(x_1\), and therefore any simultaneous decrease in the compositions \(x_1\) and \(x_2\) will reduce the values of both parameters (GSW and the TM/TE ratio).

9. QD structural parameters

From the preceding discussions on the effect of \(B, H, x_1\), and \(x_2\) in the QD structure of figure 2(a), we conclude that for our experimentally measured QD sample, the most effective model is represented by the following parameters: \(B \approx 11 \text{ nm}, H \approx 4–5 \text{ nm}, x_1 \approx 40\%\), and \(x_2 \approx 90–100\%\). The calculated PL spectra based on these sets of parameters are compared with the experimental PL spectra in figure 4. It is worth noting that the proposed structures roughly reproduce the complex composition profile experimentally found by many authors [22], including the anisotropic In–Ga intermixing behavior with respect to the crystallographic direction, with a larger in-plane inter-diffusion region (nearly 2 nm around the base of the dot) and a negligible inter-diffusion at the top (0–1 nm) where segregation predominated.

10. Relaxed strain energies

The QD systems have been relaxed using the VFF model to reach a minimum strain energy configuration [24, 26].
Figure 4. We show the normalized PL spectra from the experimental measurement (black lines) and the theoretical calculations (green and red lines). Both TE (dotted lines) and TM (solid lines) components are plotted. The slight difference between the GSW of the calculated TM and TE modes is due to the fact that the TE mode is dominantly from the E1–H1 transition and the TM mode is dominantly from E1–H3 and E1–H4 transitions. Such a difference will be difficult to observe in the experiment, because of the broadening of the PL spectra which are typically collected from multiple QDs.

Figure 5. Plots of the total strain energy of the relaxed QD systems as a function of the total number of In atoms in the system. Both the uniform and non-uniform composition models have been considered. The relaxed strain energies for the non-uniform composition configurations are smaller than the relaxed strain energies of the uniform composition configurations with similar number of In atoms.

The comparison of the relaxed strain energies for the various QD systems under study may provide additional insight into how to find a more likely experimental geometry. Figure 5 plots the relaxed strain energies as a function of the total number of In atoms for the various configurations after the VFF minimization is achieved. We consider two cases of the uniform composition model: an In\textsubscript{0.4}Ga\textsubscript{0.6}As QD with decreasing In composition (see figure 1(c)) and a pure InAs QD with decreasing base diameter. We also consider two cases of the non-uniform composition model: (B, H, x\textsubscript{1} = 40\%, x\textsubscript{2} = 100\%) and (B, H, x\textsubscript{1} = 40\%, x\textsubscript{2} = 90\%). Clearly, the relaxed strain energies belonging to the two-layer composition configurations are lower than the strain energies of the uniform composition configurations for a similar number of In atoms. This suggests that a two-composition model is more favorable as a proposed experimental geometry, compared to a uniform composition model, in accordance with our earlier findings.

11. Strain profile analysis

Our model 2 agrees with the experimental TE and TM PL spectra. However a comparison of strain profiles is required to further understand the difference between the calculated PL spectra for models 1 and 2. In figures 6((a)–(c)), we compare three QD configurations: (i) a pure InAs QD with base = 15 nm and height = 5 nm, (ii) an In\textsubscript{0.7}Ga\textsubscript{0.3}As alloy QD of the same size, and (iii) a two-layer composition as in figure 2(a) with B = 11 nm, H = 4 nm, x\textsubscript{1} = 40\%, and x\textsubscript{2} = 100\% which gives the best agreement with experiment. For reference, the previously calculated values of the GSW
and TM/TE ratio are also listed underneath. To understand the shifts in the values of the TM/TE ratio and GSW in (b) and (c) with respect to (a), we compare the hydrostatic and biaxial strain plots along the [001] direction through the center of the QDs in figures 6(d) and (e), respectively. For the In0.7Ga0.3As alloy QD, the strain values inside the QD region are randomly distributed due to the disordered composition of the alloy. We mark the average values of the hydrostatic and biaxial strains with dotted horizontal lines.

The hydrostatic strain shifts the conduction and valence band edges, therefore increasing the band gap, whereas the biaxial strain is mainly responsible for controlling the splitting between the HH and LH valence band edges [29]. The strain driven band edge shifts in table 1 are calculated from simple analytical expressions involving InAs/GaAs deformation potentials [29].

### 12. The blue shift of the GSW

The blue shifts in the GSW for the models (b) and (c) with respect to (a) indicate an increase of the optical gaps (E_g), which can be understood as a cumulative effect of the relaxed hydrostatic strains and the increase of the band gaps due to the reduced average In compositions of the QDs [32]. By comparing the hydrostatic strain profiles in figure 6(d), we deduce that the presence of InGaAs alloy, in general, reduces the strain, which can be attributed to reduced QD lattice mismatch with the GaAs buffer. As a result, the band gaps for (b) and (c) will be smaller than for (a) due to smaller strain driven shifts in the conduction and heavy hole band edges. For example, from table 1, the strain driven shifts lead to a prediction that the band gap for the In0.7Ga0.3As QD is 373.7 − 231.6 = 142.1 meV smaller than the InAs QD. However if the relative increase (∼310 meV) in the band gap of the In0.7Ga0.3As QD due to the reduced average In composition is also added, the cumulative change is an 310 − 142 = 168 meV increase with respect to the InAs QD, consistent with the increase of the optical gap. To summarize, the relaxation of the hydrostatic strain due to the presence of the InGaAs alloy in (b) and (c) introduces reductions in the band gaps which are smaller than the corresponding band gap increase due to the reduced average In compositions, and hence the cumulative impact of these two effects results in an overall increase of the optical/band gaps (blue shifts of the GSWs) for (b) and (c) with respect to (a).

### 13. Biaxial strain relaxation increases the TM/TE ratio

Figure 6(e) compares the biaxial strain components and table 1 provides the values of the corresponding changes in the HH and LH band edges for the three QD configurations. When compared to the pure InAs QD, the biaxial strain component is reduced for both the InGaAs alloy QD and the two-composition QD model. This reduces the splitting between the HH and LH bands, thus increasing the LH component in the topmost valence band states. A past theoretical investigation [40] has also reported an increased HH/LH intermixing for an InGaAs QD when compared to a pure InAs QD. The increased LH character in turn enhances the TM component [4], as we compute for configurations (b) and (c). The smallest HH/LH splitting responsible for the enhanced TM component occurs for the two-layer composition model.

### 14. Conclusions

In conclusion, multimillion-atom simulations are performed to understand the correlation between the chemical composition of self-assembled QDs and their electronic properties by reproducing the experimentally measured polarization properties. A single-composition QD model, being either a pure InAs QD or an alloyed InGaAs QD, failed to reproduce the experiment. To fully understand the experimental polarization behavior, the model must take In–Ga intermixing effects into account, by representing the QDs with variable compositions. On the basis of a systematic analysis of the dependence of the TM/TE ratio on the various QD morphology parameters, we propose a two-composition model that accurately reproduces the measured PL response. The model gives results consistent with the experimental compositional findings and highlights the strong anisotropy of atomic scale phenomena like intermixing and segregation affecting the polarization behavior of these nanostructures. These results could indicate a way for using growth dynamics to engineer the strain field inside and around the QD structures, allowing tuning of the polarization properties, a critical parameter for several challenging applications.

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