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Citation for published version (APA):

Grossi, D., Smereka, P., Keizer, J. G., Ulloa, J. M., & Koenraad, P. M. (2014). Height control of self-assembled quantum dots by strain engineering during capping. *Applied Physics Letters*, 105, 143104-1/4. [143104]. <https://doi.org/10.1063/1.4897345>

DOI:

[10.1063/1.4897345](https://doi.org/10.1063/1.4897345)

Document status and date:

Published: 01/01/2014

Document Version:

Publisher's PDF, also known as Version of Record (includes final page, issue and volume numbers)

Please check the document version of this publication:

- A submitted manuscript is the version of the article upon submission and before peer-review. There can be important differences between the submitted version and the official published version of record. People interested in the research are advised to contact the author for the final version of the publication, or visit the DOI to the publisher's website.
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Height control of self-assembled quantum dots by strain engineering during capping

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(Received 28 August 2014; accepted 25 September 2014; published online 6 October 2014)

Strain engineering during the capping of III-V quantum dots has been explored as a means to control the height of strained self-assembled quantum dots. Results of Kinetic Monte Carlo simulations are confronted with cross-sectional Scanning Tunnel Microscopy (STM) measurements performed on InAs quantum dots grown by molecular beam epitaxy. We studied InAs quantum dots that are capped by $\text{In}_x\text{Ga}_{(1-x)}\text{As}$ layers of different indium compositions. Both from our realistic 3D kinetic Monte Carlo simulations and the X-STM measurements on real samples, a trend in the height of the capped quantum dot is found as a function of the lattice mismatch between the quantum dot material and the capping layer. Results obtained on additional material combinations show a generic role of the elastic energy in the control of the quantum dot morphology by strain engineering during capping. © 2014 AIP Publishing LLC. [<http://dx.doi.org/10.1063/1.4897345>]

The epitaxial growth of Stranski-Krastanov Quantum Dots (QDs) has been extensively studied in the last decades.^{1,2} In this growth process, the strain, due to the lattice mismatch between the deposited material and the substrate, controls to a large extent the formation of self-assembled QDs. The three dimensional confinement of carriers in these nanostructures makes them interesting for applications like QD lasers,³ single photon sources,⁴ and single electron transistors.⁵ The optoelectronic properties of QDs are determined by the confinement of the carriers⁶ and are therefore directly connected to their morphology, which is the result of a delicate interplay between the elastic relaxation and the surface energy.⁷

In many studies, only the shape obtained after the QD growth process is investigated.^{1,8} In order to be used in an actual device, however, the QDs need to be overgrown with a capping layer, and, in this process, the morphology and the properties of the QDs can dramatically change.^{9,10} The choice of the capping material can thus have an important influence on the final properties of these nanostructures. For instance, the tuning of the composition of the capping layer makes it possible to control the strain field inside the QDs¹¹ and the QDs erosion process.¹² This means that in order to control the structural properties of the nanostructures, strain engineering during capping is just as important as during the formation of the QDs. In this letter, we use a Kinetic Monte Carlo (KMC) model¹³ to investigate the role of the strain in the control of the morphology of the QDs during capping. The system considered consists of InAs QDs grown on GaAs that are capped with $\text{In}_x\text{Ga}_{(1-x)}\text{As}$, where the lattice mismatch between the QDs and the capping layer is engineered by tuning the In concentration in the capping layer.¹⁴ The effects on the QDs

morphology are studied, and the results obtained from the simulation are compared with cross-sectional Scanning Tunnel Microscopy (X-STM) measurement on QD samples grown by Molecular Beam Epitaxy (MBE).¹⁵

In our KMC model for hetero-epitaxial growth every pair of III-V atoms in the system is modeled with a cube, as shown in Fig. 1. The short range interaction between each cube and its neighbors is described using a bond-counting model. A ball and spring model is used for the description of long range elastic interaction. In the model, only the surface atoms are considered to be mobile, since their mobility is much higher than the bulk mobility,¹⁶ but a certain degree of intermixing is allowed by the thermal roughening of the surface. Through this process, atoms deposited on the surface can be incorporated in subsurface layers. In the simulation, each cube at the surface can jump to one of his neighboring sites with a hopping rate given by

$$R = R_0 \exp \left[-\frac{B - \Delta W}{k_B T} \right], \quad (1)$$

where R_0 is the hopping attempt frequency, $k_B T$ is the thermal energy, ΔW is the elastic energy, and B is the bonding energy.

FIG. 1. Representation in the KMC model of an InAs “adatom” on a GaAs surface. The interaction of the “adatom” with the adjacent cubes, which correspond to the first, second, and third nearest neighbors (marked in progressively darker blue), is described with a bond counting model.

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There are different possible bonds in the system, depending on the type of “atoms” involved. In this letter, the system studied is composed by InAs QDs grown on GaAs and capped with $\text{In}_x\text{Ga}_{(1-x)}\text{As}$. Therefore, the possible bonds in the system are GaAs–GaAs, GaAs–InAs, and InAs–InAs bonds. The bonding energy for a generic cube is given by

$$B = B_{\text{Ga-Ga}} + B_{\text{Ga-In}} + B_{\text{In-In}}. \quad (2)$$

The three terms count the number of bond that an “atom” has of each type and are given by

$$B_\alpha = (aN_\alpha^I + bN_\alpha^{II} + cN_\alpha^{III})\gamma_\alpha, \quad (3)$$

where α identifies the type of bond, N_α^I , N_α^{II} , N_α^{III} are the number of α -type bonds with the first, the second and the third neighbors, and γ_α is the energy of a single bond. The three parameter a , b , and c are chosen to reproduce the anisotropy of the interaction and the different surface energies. In our simulation, we choose $a = 0.3$, $b = 0.5$, and $c = 1.0$, which give rise to pyramidal QDs. For the other parameters, we take $\gamma_{\text{Ga-Ga}} = 0.2506$, $\gamma_{\text{Ga-In}} = 0.2217$, and $\gamma_{\text{In-In}} = 0.2169$ all in eV.¹⁵

The elastic energy ΔW is the change in the total elastic energy given by the removal of the atom considered. The elastic interactions in the system are calculated with a ball and spring model, using two spring constants, k_L and k_D , and the misfit μ between InAs and GaAs. We take $k_L = 2.89 \text{ eV}/l_1^2$ and $k_D = 9.64 \text{ eV}/l_1^2$, where l_1 is the lattice constant of GaAs, and a misfit $\mu = 0.075$. This set of parameters gives a reasonable approximation for the bulk and surface properties of GaAs.¹⁷ The key approximation of the model, which has been thoroughly tested,¹⁸ is that the movement of an adatom does not depend on the elastic field. Thanks to this approximation the code is 10–15 times faster. This allows us to simulate systems on a realistic time scale that are as big as $90 \times 90 \text{ nm}^2$ which can be directly compared to experimental observations.

The QDs for the X-STM analysis were grown by MBE on a n-doped (100) GaAs substrate. The growth process is

initiated with the deposition of 350 nm GaAs at 580°C as a buffer layer. The sample contains 4 layers of capped QDs, grown by deposition of 2.7 ML of InAs at 450°C and with a growth rate of 0.04 ML/s. Every QD layer is overgrown with 5 nm of $\text{In}_x\text{Ga}_{(1-x)}\text{As}$, with $x = 0.00$, $x = 0.05$, $x = 0.10$, and $x = 0.15$, respectively, for the first, second, third, and fourth layer of QDs. A growth rate of 0.75 ML/s and a growth temperature of 450°C were used for the $\text{In}_x\text{Ga}_{(1-x)}\text{As}$ layer. The QDs layers are separated by GaAs spacer layers of 50 nm, grown at 580°C and with a growth rate of 0.7 ML/s. Finally, a QD layer is grown on top of the sample for the analysis of uncapped QDs by Atomic Force Microscopy (AFM). The growth parameters used in the simulation match the ones used for the MBE grown sample, except for the growth rate for the QD layer, which is taken 0.15 ML/s in the simulations, roughly four times bigger than the experimental value. X-STM analysis was performed at 77 K under Ultra High Vacuum (UHV) conditions on a (110) surface obtained by cleaving *in-situ*. The measurements were all done at high negative bias voltages and low tunneling currents ($V = -3 \text{ V}$, $I = 20 \text{ pA}$).

In Fig. 2, we show the result of the simulations for an ensemble of InAs QDs after being capped by 5 nm of $\text{In}_x\text{Ga}_{(1-x)}\text{As}$ with different In concentrations. In the figure, the capped QDs are represented using the isosurfaces corresponding to an In concentration of 66%. From the comparison between the images shown in Fig. 2, a number of trends can be observed when the indium concentration in the capping layer is increased: (1) The average height of the QDs increases; (2) the number of QDs preserved after capping increases; and (3) the number of ring-like features decreases. The observed trends are all related to the strain induced destabilization of the QDs. If the lattice mismatch between the dot materials and the capping layer is large, the dot erosion will be strong, reducing the average height of the QDs and desolving more QDs. The evolution of QDs in ring-like shape after capping is a phenomena already experimentally observed.¹⁰ This is attributed to the lateral compression exerted by the capping on the QD, which cause the In to flow

FIG. 2. Top view of simulated capped QDs, represented as the isosurfaces corresponding to an In concentration of 66%. The indium concentration in the $\text{In}_x\text{Ga}_{(1-x)}\text{As}$ capping layer is $x = 0, 0.05, 0.10, 0.15, 0.20$ in (a), (b), (c), (d) and (e), respectively. The dimensions of the simulated system are $90 \times 90 \text{ nm}^2$.

FIG. 3. Height distribution for capped QDs. Increasing the In concentration in the $\text{In}_x\text{Ga}_{(1-x)}\text{As}$ capping layer increases the average height of the QDs.

out of the center of the QD. A capping layer with a higher In concentration has a smaller lattice mismatch with the QD, which means that the compressive strain applied to the QD will be lower, and this explains the lower number of rings that are formed. Our observation that the number of ring structures decreases when the capping layer has a higher In concentration supports the proposed mechanism for the ring formation.

In order to show the effect of the composition of the capping layer on the height of the QDs more quantitatively, the height distribution of the capped QDs obtained from the simulation is shown in Fig. 3. The average height for QDs capped with a higher In concentration is shifted to higher values, confirming that the strain engineering during capping plays a central role in determining the final height of QDs in our KMC model.

In order to confirm this trend observed in the simulation, the results are compared with the X-STM analysis on MBE grown QDs. In the first three capped layers of QDs, overgrown with $\text{In}_x\text{Ga}_{(1-x)}\text{As}$ with an indium concentration of $x = 0.00$, $x = 0.05$, and $x = 0.10$, many QDs are analyzed and the average height of the QDs is determined to be 3.9 ± 0.4 nm, 4.6 ± 0.4 nm, 5.1 ± 0.4 nm, respectively. With

the AFM, the average height of uncapped QDs is also measured, giving $h = 5.6 \pm 0.7$ nm.

The cross-section of a capped QD obtained from the simulation is shown in Fig. 4, where we can see a good qualitative agreement between the morphological features of a real QD and one obtained in the simulations. The average height results obtained from the simulation are compared with the results obtained by X-STM measurement in Fig. 5.

There is a qualitative agreement between the trend observed from the experimental data (red open symbols) and the results of the simulation (black filled symbols) which confirms that a smaller mismatch between QD and capping layer will reduce the dot erosion process¹⁵ and will result in QDs with a larger height. In Fig. 5, additional experimental points are shown (blue open symbols), obtained by X-STM measurements on other InAs QD samples that are grown under different conditions and capped with GaAsSb instead of InGaAs.^{12,19}

FIG. 4. (a) Cross-sectional view of a QD capped with GaAs obtained with X-STM. (b) X-STM image after the application of a high-pass filter to enhance the contrast of In atoms (bright spots) over Ga atoms. (c) Color map of the indium density obtained for a QD capped with pure GaAs in the simulation, where the In concentration varies from 0% (black) to 100% (white).

FIG. 5. Average height of capped QDs as a function of the lattice mismatch between the QD and the capping layer. The average height of the QDs is increased when the lattice mismatch between the QD material and the capping layer is reduced. Both the average QD heights obtained in KMC simulations and the X-STM measurements are presented in this graph (respectively, the black filled and red open symbols). Furthermore, experimental results from X-STM measurements performed on samples where InAs QDs grown under different conditions were capped with GaAsSb (blue open symbols) are shown in this graph.

The qualitative agreement with the trend found for the systems studied in this letter and the points obtained for different InAs QDs systems is striking, in spite of the differences between the systems considered (i.e., different capping and different morphology of the uncapped QDs). This suggests that the morphological properties of uncapped QDs are of secondary importance with respect to the misfit between the QD and the capping layer material. Therefore, the offset in the trend obtained for the simulation and the experimental result is not attributed to the difference in height between the uncapped QD studied (5.6 ± 0.7 nm for the real system and 5.1 ± 0.5 nm for the simulations), but can be explained considering the different level of intermixing in the real and simulated QDs. In the KMC model, the intermixing is taken into account only at the surface during the growth, whereas bulk intermixing is not considered. This is an underestimation with respect to a real system where both types of intermixing occur. The intermixing increases the Ga content in the InAs QD, reducing the misfit with the $\text{In}_x\text{Ga}_{(1-x)}\text{As}$ capping layer. Therefore, a real QD will have an higher Ga content and a smaller mismatch with the capping layer than a QD obtained in the simulations under the same conditions. This argument is supported by the result shown in Fig. 4, where a filtered STM image and a map of the In density for a simulated QD are shown. In the STM image the In atoms appear as bright spots, and a fluctuation in In content inside a real QD can be seen. The map of the In density for a QD obtained from the simulations, instead, shows an internal region of high and uniform In concentration, confirming that in these QDs a smaller degree of intermixing has occurred. A 10% higher Ga concentration in the real QDs as compared to the simulated QDs nicely explains the shift between the curves describing the QD height vs. mismatch dependence for the simulated and measured QDs, shown in Fig. 5. The observation that all the experimental points qualitatively follow the same trend found in the simulations suggests that the relation between the mismatch of the QDs with the capping and their final height can be general for many strained QDs systems.

In conclusion, strain engineering during capping by tuning the mismatch between capping layer and QDs makes it possible to effectively control the height of QDs.

The good qualitative agreement between the simulated QDs obtained by KMC model and those observed by X-STM in MBE grown samples confirm that the KMC model can be successfully used, in combination with experimental techniques like X-STM, to gain a deeper understanding in growth processes occurring during hetero-epitaxial growth.

The authors acknowledge the support from the Dutch Foundation for Fundamental Research on Matter (FOM) and from the National Science Foundation, Grant No. DMS 1115252. We also acknowledge the Spanish MINECO for funding through project MAT2013-47102-C2.

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