Size-Dependent Strain Relaxation in InAs Quantum Dots on Top of GaAs(111)A Nanopillars

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The size-dependent strain relaxation in InAs quantum dots on the top face of GaAs(111)A nanopillars is studied experimentally by scanning transmission electron microscopy (STEM) and theoretically using molecular static simulations. In the experiment, a dislocation-free, coherent state is observed for InAs dimensions below 10 nm in width and 7 nm in height, while 60° misfit dislocations occur for larger sizes. Experimental strain maps reveal the presence of a narrow-strained region at the heterointerface with a 20–80% width of 1.2 nm in the coherent case and 0.4 nm in the dislocated state, in agreement with simulations. Moreover, an analysis of the amount of misfit relaxed by the observed dislocations shows that the transition between the purely elastic and plastic relaxation regimes appears to be gradual. Intensity profiles of STEM images reveal that the misfit dislocations are located directly at the GaAs/InAs heterointerface.

1. Introduction

InAs quantum dots (QDs) are promising for optoelectronic applications such as single-photon emitters for quantum optics and cryptography,[2] lasers,[3,4] photodetectors,[5,6] and photovoltaics.[7] Typically, a uniform QD size, low or controllable strain and absence of defects are required or at least beneficial for high-performance devices. However, a low QD height-to-width aspect ratio like in the case of pyramid-shaped InAs QDs on GaAs(001) results in a large compressively strained zone in the QD.[7-9] Moreover, a considerable Ga diffusion from the GaAs(001) substrate into the InAs islands is observed, depending on the growth conditions.[10]

In order to reduce the heavily strained InAs volume and interdiffusion phenomena, InAs growth on GaAs(111)A nanopillar tops appears to be an alternative worth considering.[11] This has been accomplished by low-temperature molecular beam epitaxial growth on nanopillar-patterned GaAs, where the restricted adatom migration together with an enhanced nucleation rate of the second atomic layer on monolayer islands leads to QD formation on the pillar tops.[12,13] Since these QDs have steeper sidewalls than conventional InAs QDs grown at higher temperatures on planar GaAs(001),[14] a more efficient elastic strain relaxation and possibly an increased critical volume for misfit defect formation are expected. Another promising feature of InAs heteroepitaxy on GaAs(111)A is the absence of Ga/In alloying.[15,16] In general, heterostructures consisting of mismatched semiconductor islands on nanopillar tops have aroused significant interest because of the enhanced capabilities to position the islands, to control their size, and to relieve misfit strains elastically.[17-20]

The present study analyzes the size dependence of the elastic and plastic strain relaxation mechanisms including strain distributions in InAs QDs on GaAs nanopillar tops by means of atomic-resolution high-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM) imaging and molecular static simulations. Section 2 presents and discusses the results, and Section 3 draws conclusions. Finally, details on the performed experiments as well as theoretical calculations are given in Section 4.

2. Results and Discussion

In the following, the size-dependent occurrence of misfit dislocations and the spatial distribution of lattice strain in nanoscale InAs islands grown on top of GaAs nanopillars are examined. For a nominal InAs deposit height of 15 nm dislocated InAs islands are observed. The example shown in Figure 1 has a width of 24 nm and a height of 16 nm, and four misfit dislocations at the GaAs–InAs interface, which are of 60° type according to Burgers circuit analysis with Burgers vector \( \mathbf{b} = a/2[101] \) (using the right-hand start-to-finish convention with dislocation sense vector [0Ti]).[21] The portion of the misfit geometrically accommodated by these dislocations is given by Equation (1)

\[
\epsilon_{pl} = \frac{N\beta N_{dis}}{w_{ins}}
\]

where \( N \) denotes the number of dislocations, \( b_{eff} = \sqrt{6}/4a \) the magnitude of the effective Burgers vector, i.e., the edge
component of $b$, and $w_{\text{InAs}}$, the width of the InAs island at the heterointerface.\cite{22} As with misfit dislocations at axial heterointerfaces in GaAs/GaSb (111)B nanowires having a similar lattice mismatch,\cite{23} the Burgers vector lies in the heterointerface plane. This results in a larger effective Burgers vector as compared to dislocations on glide planes tilted against (111),\cite{24} which were detected in planar In$_x$Ga$_{1-x}$As layers on GaAs(111)B substrate.\cite{25,26} Moreover, dislocations along $\{11\bar{2}\}$ directions have not been found in our samples. For the island depicted in Figure 1, the dislocations accommodate \approx 90\% of the misfit $f = 2(a_{\text{InAs}} - a_{\text{GaAs}})/(a_{\text{InAs}} + a_{\text{GaAs}})$ with lattice parameters $a_{\text{GaAs}}$ and $a_{\text{InAs}}$ (along one direction; since 60° dislocations can run parallel to each of the directions [111], [10\overline{1}], [01\overline{1}], the lattice will also experience plastic relaxation along the normals to these symmetry-equivalent directions). As visible in the map of the relative $(\overline{2}11)$ plane distance variations $\Delta d/d_{\text{GaAs}} = (d - d_{\text{GaAs}})/d_{\text{GaAs}}$ (with $d_{\text{GaAs}}$ the lattice distance of unstrained GaAs) obtained by geometrical phase analysis (GPA) in Figure 1c, both InAs and GaAs outside the heterointerface are approximately completely relaxed, indicating that \approx 10\% of the misfit is relaxed elastically. A line profile along the growth direction shows a 0.4 nm narrow transition zone at the heterointerface in which the strain concentrates. In addition to the misfit dislocations, a number of rotational twins and stacking faults is observed on the InAs (111) planes.

In the case of a nominal InAs deposition of 5 nm on 30-nm diameter GaAs nanopillars, the InAs island shown in Figure 2 has a width of 11 nm and a height of 7 nm (for the crystalline part disregarding amorphous surface regions), and reveals a single dislocation accommodating \approx 56\% of the misfit, according to Equation (1). In order to determine the exact position of this misfit dislocation relative to the heterointerface, the intensities of the (Ga,In) and As atomic columns along the [0\overline{1}1] direction have been evaluated and the (Ga,In)/As intensity ratio plotted as a function of distance to the heterointerface (Figure 2b). The normalization to the As column intensity, a measure of the specimen thickness, is necessary, because the specimen thickness varies considerably with position across the interface. The observed transition of this atomic number sensitive intensity ratio between GaAs and InAs indicates that the dislocation core is situated directly at the GaAs/InAs heterointerface. Apart from that, a minimum of the intensity ratio occurs in a narrow GaAs zone near the heterointerface, which could be attributed to an As enrichment in near-surface regions owing to decomposition of AsH$_3$ formed during HF acid and atomic H treatments before the growth.\cite{25,26} Figure 2c presents maps of the relative $(\overline{2}11)$ plane distance variations obtained from the experiment by GPA (Figure 2c) and a molecular static simulation (Figure 2d). These maps evince that the dilatation zone on the InAs side of the dislocation core contributes to the reduction of misfit-induced compression of InAs (orange to red areas above dislocation cores in Figures 1c and 2c,d). The residual strain in the InAs regions >1 nm away from the heterointerface is close to zero, i.e., lower than the value expected from the partial misfit accommodation by the dislocation. This indicates that part of the misfit strain relaxes elastically at the nearby free surfaces.

For a nominal InAs deposit height of 2 nm an even smaller InAs island size is observed. The island shown in Figure 3, sitting on a 25-nm-diameter GaAs nanopillar, has a width of 6 nm and a height of 6 nm (for the crystalline part), and is dislocation-free. Figure 3a depicts the maps of the relative $(\overline{2}11)$ plane distance variations resulting from the experimental HAADF-STEM image and from molecular static simulation,

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**Figure 1.** a) HAADF-STEM image of a dislocated InAs island on a GaAs(111)A nanopillar (diameter 40 nm) in [0\overline{1}1] zone axis orientation. b) The same island (rectangular box in a) imaged at atomic resolution with the misfit dislocations marked in red. The inset shows an area with one of the dislocations at higher magnification. Also drawn is a Burgers circuit around the dislocation core, starting at point S and finishing at point F. The vector $\vec{SF}$ transferred into the perfect lattice represents the edge component of the Burgers vector. c) Corresponding GPA map with the profile of the relative $(\overline{2}11)$ plane distance variations $\Delta d/d_{\text{GaAs}}$ along [111] (the reference is unstrained GaAs).
respectively. Here, because of the presence of several stacking faults in the InAs, the experimental map has been obtained by fitting 2D Gaussians to the intensity peaks in order to locate the atomic column positions and performing peak-pairs analysis (PPA). Extracted line profiles of the relative plane distance variations along the [111] direction indicate agreement between experiment and simulation, and demonstrate that the lattice parameter transition between GaAs and InAs occurs in a remarkably narrow zone with a 20–80% width of 1.2 nm in the experiment or 1.1 nm in the simulation (Figure 3e). This result points to a very effective elastic relaxation of misfit strains at the nearby sidewall surfaces. The relatively strong data scatter in Figure 3c can be primarily attributed to the TEM specimen preparation, i.e., a large ratio of the focused ion beam damaged and oxidized InAs surface layer thicknesses to the transmitted thickness of pristine InAs.

In order to address the question of how the finite-diameter nanopillar substrate affects the strain relaxation, the map and profile of Figure 3d,e are compared with those obtained from analogous simulations for an identically sized InAs island on a smaller and on a larger diameter GaAs nanopillar (Figure 4). While the data for the system with a larger, 42 nm diameter pillar hardly differs from the case of Figure 3 (diameter 25 nm), a larger dilatation of the GaAs and a lower compression of the InAs in close vicinity of the heterointerface are observed, if the GaAs pillar diameter is reduced to that of the InAs island. Owing to the higher compliancy of the GaAs in the latter case the strain partitions more symmetrically around the heterointerface.

In the literature, several approaches have been described for calculating the critical dimensions of heteroepitaxial islands grown on either planar or on nanopillar substrates. First, the analytical model of Tillmann and Förster considers conical quantum dots with a variable height-to-diameter ratio on a planar substrate, resulting in a critical diameter (at the island base) of 15 nm for a height-to-diameter ratio value of 1, corresponding to a sidewall angle of 63.4°.[27] However, this value might be inaccurate because the correction function employed for approximating the strain relaxation has been fitted only for sidewall angles ≤45°. Second, the approach of Ye and Yu refers to a mismatched nanowire growing either on a nanowire substrate having the same diameter or on a planar substrate.[28] The coherent-to-incoherent transition observed in our data (Figures 2 and 3) occurs for InAs diameters between 6 and 11 nm, in line with the critical diameter of 9 nm predicted by the model of Ye and Yu for the case of a nanowire on a planar substrate.
3. Conclusion

In summary, the size-dependent strain relaxation in InAs quantum dots grown on (111)A GaAs nanopillar tops has been studied at the atomic scale by means of aberration-corrected HAADF-STEM and molecular static simulations. It is found that InAs islands with a width larger than 10 nm and a height larger than 7 nm reveal one or more 60° dislocations directly at the GaAs/InAs heterointerface, while smaller ones are dislocation-free. Islands with sizes close to the critical dimensions show both significant elastic and plastic relaxation, indicating a gradual transition between the two relaxation regimes. Strain analysis yields a 20–80% Δd/d_{GaAs} strained transition zone width of 1.2 nm in the experiment and 1.1 nm in simulation at the dislocation-free GaAs/InAs interface, demonstrating the effectiveness of elastic lattice relaxation at the free surfaces.

Due to their steeper sidewalls and a larger volume fraction of fully relaxed lattice as compared to InAs quantum dots on planar GaAs(001), the InAs dots on GaAs(111)A nanopillars are promising for achieving narrower infrared emission linewidths.

4. Experimental Section

Experimental Details: Nanopillar arrays were patterned on GaAs(111)A wafers using nanosphere lithography and reactive ion etching. The pillar height amounted to 80–90 nm, the diameter to 20–45 nm. Next, the patterned substrate was in situ cleaned using atomic hydrogen, and InAs was heteroepitaxially grown by solid source molecular beam epitaxy. For the formation of InAs islands on top of the nanopillars, a low growth temperature of 150 °C at a rate of 0.011 nm s⁻¹ under As-rich conditions (V/III ratio ≈400) was chosen. The nominally deposited InAs thickness was between 2 and 15 nm. Structural characterization was performed by aberration-corrected HAADF-STEM imaging in a JEOL JEM-ARM200F instrument operated at 200 kV. TEM cross-sectional specimens were prepared by mechanical grinding followed by dimpling and ion polishing using a Gatan PIPS Model 691. In addition, one TEM specimen was prepared by focused ion beam milling using 5 keV Ga⁺ ions in the last thinning step. Strain maps were evaluated by GPA using the plugin of HREM Research Inc. and by PPA using self-written scripts in the Gatan Microscopy Suite software.[30–32]

Calculational Details: Atomistic models consisting of a centered InAs island on top of a GaAs nanopillar were generated by self-written script programs in the DigitalMicrograph software. For both InAs and GaAs cylindrical morphologies with dimensions corresponding to the experiments for deposited InAs thicknesses of 2 and 5 nm were chosen (Figure 5). A cylindrical shape has been assumed because it is similar to the rounded triangular InAs island shape found by scanning electron microscopy imaging for 15 nm deposited thickness,[31] and because for smaller deposited thicknesses, i.e., smaller island size the triangular shape is expected to be less stable from an energetic viewpoint. If present in the experiment, 60° misfit dislocations were inserted at the heterointerface as follows. First, the atoms in an inclined InAs (111) half plane intersecting the nanopillar axis are removed and placed on the top (111) surface of the island. Second, the formed gap in InAs is closed by applying static atom displacements, followed by imposing an analytical approximation.

![Figure 3.](image-url)
of the dislocation strain field (Figure 5b, graphics at the right).\[21\] As the potential used for the relaxation better reproduces a Ga terminated than an As terminated dislocation core, the considered configuration was a Ga terminated shuffle-set dislocation. Third, the structures were iteratively relaxed by minimizing their total energy with the conjugate gradient method in the LAMMPS software.\[33\] The energy computation relies on the Tersoff potential using the parametrization of Hammerschmidt et al.,\[34–36\] which considers the interaction energies of atom triples including the effect of atomic coordination on bond strength. This parametrization results in equilibrium lattice parameters of $a_{\text{GaAs}} = 5.653$ Å for GaAs and $a_{\text{InAs}} = 6.050$ Å for InAs, corresponding to a misfit of $f = 6.8\%$, compared to reported experimental bulk lattice parameters of $a_{\text{GaAs}} = 5.653$ Å for GaAs and $a_{\text{InAs}} = 6.058$ Å for InAs ($f = 6.9\%$).\[37,38\] As stopping criterion for the iterations a relative energy difference between successive steps of $<10^{-9}$ was applied. Lattice plane distance maps and profiles were extracted from relaxed atom coordinates by means of self-written DigitalMicrograph scripts. The InAs wetting layer on the GaAs nanopillar detected in the experiment was not included in the simulation because it does not significantly influence the lattice strain.\[31\]

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords

III–V semiconductors, heteroepitaxy, misfit dislocations, strain, nanostructures

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