Formation of linear InAs quantum dot arrays on InGaAsP/InP (100) by self-organized anisotropic strain engineering and their optical properties

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Formation of linear InAs quantum dot arrays on InGaAsP/InP (100) by self-organized anisotropic strain engineering and their optical properties

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The formation of linear InAs quantum dot (QD) arrays based on self-organized anisotropic strain engineering of an InGaAsP/InP (100) superlattice (SL) template in chemical beam epitaxy is demonstrated, and the optimized growth window is determined. InAs QD formation, thin InGaAsP capping, annealing, InGaAsP overgrowth, and stacking in SL template formation produce wirelike InAs structures along [001] due to anisotropic surface migration and lateral and vertical strain correlations. InAs QD ordering is governed by the corresponding lateral strain field modulation on the SL template surface. Careful optimization of InGaAsP cap layer thickness, annealing temperature, InAs amount and growth rate, and number of SL periods results in straight and well-separated InAs QD arrays. The InAs QD arrays exhibit excellent photoluminescence (PL) emission up to room temperature which is tuned into the 1.55 μm telecommunications wavelength region through the insertion of ultrathin GaAs interlayers. Temperature dependent PL measurements and the linear polarization behavior indicate lateral electronic coupling of the QDs in the arrays.


I. INTRODUCTION

Many applications of epitaxial semiconductor quantum dots (QDs) in future quantum functional devices require lateral ordering of QDs in well-defined arrangements. To overcome the random arrangement of QDs formed in the Stranski-Krastanov (SK) growth mode, the QD growth on multilayer high stepped vicinal substrates or on artificially patterned substrates has been pursued.1–5 The structural and optical properties of such ordered QDs, however, are often degraded due to step edge roughness or pattern imperfections at the nanometer scale. We have previously introduced a concept for the lateral ordering of InAs QDs based on self-organized anisotropic strain engineering of InGaAs/GaAs superlattice (SL) templates in molecular beam epitaxy (MBE) producing well-ordered QD arrays with excellent optical properties due to the inherent smoothness of the lateral strain field modulations on the SL template surface at the nanometer scale.6–8

In this work, we transfer our concept of self-organized anisotropic strain engineering to the InAs/InGaAsP material system on InP (100) by chemical beam epitaxy (CBE) and optimize the growth conditions for formation of well-ordered, linear InAs QD arrays with photoluminescence (PL) emission in the technologically important 1.55 μm telecommunications wavelength region. InAs/InGaAsP SL template formation comprises InAs QD formation, thin InGaAsP capping, annealing, InGaAsP overgrowth, and stacking. This produces wirelike InAs structures due to anisotropic adatom surface migration during annealing, together with lateral and vertical strain correlations and strain gradient driven In adatom migration during stacking. The corresponding lateral strain field modulation on the SL template surface governs InAs QD ordering due to local strain recognition. The presence of linear QD arrays depends on the substrate miscut altering the adatom surface migration. The optimum substrate miscut is 2° toward (110) for which the InGaAsP cap layer thickness, annealing temperature, InAs amount and growth rate, and number of SL periods are optimized for the formation of straight and well-separated QD arrays along [001]. The PL emission of the optimized InAs QD arrays is tuned into the 1.55-μm-wavelength region through the insertion of an ultrathin GaAs interlayer beneath the InAs QD arrays.9,10 Excellent PL emission from the QD arrays is observed up to room temperature (RT). In temperature dependent PL measurements the characteristic minimum of the PL full width at half maximum (FWHM) and the enhanced low-energy shift of the PL peak due to thermally activated carrier redistribution typical for randomly arranged QDs (Refs. 11 and 12) are hardly observed. This indicates minimized carrier localization in the QD arrays due to the lateral electronic coupling of the closely spaced QDs in the arrays which is supported by the linear polarization of the PL along the arrays.

II. EXPERIMENT

The samples were grown by CBE using pressure controlled trimethylindium (TMI), triethylgallium (TEG), AsH3, and PH3 as precursors. The AsH3 and PH3 gases were thermally decomposed in a high temperature injector at 900 °C. The InP (100) substrates with different miscuts summarized in Table I were mounted by In on Mo blocks. The sample structure commenced with a 200 nm InP buffer layer and 100 nm lattice-matched InGaAsP with band gap at 1.27 μm (Q1.27), which is a typical waveguide core material in InP-based optoelectronic devices. Each period of the SL template consisted of 2.1–3.2 ML (monolayer) InAs, 10 s growth in
TABLE I. Denotation of the InP (100) substrates with various miscuts.

<table>
<thead>
<tr>
<th>Substrate</th>
<th>Miscut toward</th>
<th>Miscut angle</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>(110)</td>
<td>0.2°</td>
</tr>
<tr>
<td>B</td>
<td>(111)A</td>
<td>0.5°</td>
</tr>
<tr>
<td>C</td>
<td>(111)B</td>
<td>0.5°</td>
</tr>
<tr>
<td>D</td>
<td>(110)</td>
<td>0.5°</td>
</tr>
<tr>
<td>E</td>
<td>(111)B</td>
<td>2.0°</td>
</tr>
<tr>
<td>F</td>
<td>(110)</td>
<td>2.0°</td>
</tr>
</tbody>
</table>

terruption in As flux, a 0.3–1.0 nm thin Q1.27 cap layer, 2 min annealing, and a 15.3 nm Q1.27 spacer layer. The number of SL periods was between 5 and 11. On top of the SL template a 2.1–3.2 ML InAs QD layer was grown either directly on the Q1.27 layer or on a 0.8 ML GaAs interlayer inserted beneath the QDs. For PL measurements the InAs QDs were capped by 100 nm Q1.27 and 50 nm InP. The growth temperature was 515 °C for all layers and the annealing temperature was between 530 and 550 °C. The growth rate of InAs was varied from 0.23 to 0.47 ML/s, calibrated by high-resolution x-ray diffraction (XRD).

The surface morphologies of uncapped samples were characterized by tapping-mode atomic force microscopy (AFM) in air. For the PL studies, a neodymium-doped yttrium aluminum garnet (Nd:YAG) laser (532 nm) with excitation power density of 256 mW/cm² was used as excitation source. The samples were mounted in a He-flow cryostat with temperature control between 4.8 K and RT. The PL was dispersed by a single monochromator and recorded by a cooled InGaAs linear array detector.

III. FORMATION OF LINEAR QD ARRAYS

A. Substrate miscut

The AFM images of 2.6 ML InAs grown on a five period 2.1 ML InAs/15.6 nm Q1.27 SL template (0.23 ML/s InAs growth rate, 0.3 nm Q1.27 cap layer, and 530 °C annealing temperature) on InP (100) substrates with different miscuts are shown in Fig. 1. For low miscut <0.5° and for miscut toward (111)A (step edges along [0–11]) dense quantum dashes (QDashes) elongated along [0–11] form, as shown in Figs. 1(a) and 1(b). For miscut toward (111)B (step edges along [011]) QD arrays appear in a zigzag arrangement, alternately oriented along [001] and [010], shown in Figs. 1(c) and 1(e). Linear and well-separated QD arrays are produced on substrates miscut toward (110) (step edges along [001]) whose ordering improves with the miscut angle, shown in Figs. 1(d) and 1(f).

Obviously the preferential surface reconstruction and step driven adatom surface migration along [0–11] on substrates with low miscut or miscut toward (111)A hinder the formation of well-separated QD arrays, which is attributed to a too small surface migration length along [011]. As a result, dense QDashes elongated along [0–11] form during SL growth. Once the direction of adatom surface migration is altered by the presence of a sufficient density of steps along [011], QD arrays along [010] or [001], which are the elastically soft directions, form due to a large enough surface migration length perpendicular to the arrays. Linear and well-separated QD arrays along one of these directions, e.g., [001], are then formed for steps along [011]. The orientation of the QD arrays along the elastically soft directions implies that it is determined by in-plane strain correlation governed by the cubic anisotropy of the elastic moduli. This is different from the case of InAs/GaAs based linear QD arrays where the orientation is determined by the direction of adatom surface migration during annealing, which can be influenced by the presence of steps. In the InAs/InP system the anisotropic adatom surface migration during annealing is essential only to smoothen the InAs QD arrays in each SL period toward uniform wirelike structures oriented along the elastically soft directions to minimize the strain energy. It is important to note that the ordering of the InAs QDs is purely governed by the vertical and lateral strain correlations during stacking rather than by morphological features related to steps. This is demonstrated by the unchanged orientation and lateral periodicity of the QD arrays as a function of substrate miscut, number of SL periods, and the smoothness of the SL template surface discussed below, with the lateral periodicity determined by the strain gradient driven In adatom surface migration perpendicular to the QD arrays in successive layers.

![AFM images of 2.6 ML InAs on the five period 2.1 ML InAs/0.3+15.3 nm Q1.27 SL template (InAs growth rate of 0.23 ML/s and annealing temperature of 500 °C) on InP (100) substrates with different miscuts denoted in Table I. (a) Substrates A, (b) B, (c) C, (d) D, (e) E, and (f) F. The scan field is 2.0 × 2.0 μm² and the height contrast is 20 nm.](image)
This is in contrast to other reports\textsuperscript{17,18} where QDashes have been observed for substrates miscut toward substrates miscut by 2° toward linear and well-separated InAs QD arrays is performed on a 100 nm Q1.27 buffer layer. For all substrates with different miscuts denoted in Table \textsuperscript{1}, ordering of InAs QDs in linear arrays along [001] is clearly observed for 0.3–0.7 nm cap layer thickness together with annealing temperatures of 530–540 °C, as shown in Figs. 3(a)–3(b) and 3(d)–3(e). Outside this growth window ordering is strongly degraded or absent. Similar to the case of InAs/GaAs, the combination of cap layer thickness and annealing temperature balances In desorption during annealing. This is essential for a smooth and straight connection of the wirelike structures requiring strain reduction, maintenance of sufficient In for vertical strain correlation, and sufficient lateral mass transport during annealing to smoothen the InAs QD arrays. For too thin cap layer and too high annealing temperature [Figs. 3(c) and 3(f)], the In desorption is too high to maintain a vertical strain correlation. On the other hand, for too thick cap layer and too low growth temperature, In desorption is insufficient. The excess strain accumulation is too large that straight wirelike structures cannot form and a high density of kinks and branches is introduced in the QD arrays to relieve the strain. Moreover, smoothening of the QD arrays during annealing is suppressed and finally hindered resulting in high-density stacked QDs [Figs. 3(g)–3(i)] which eventually develop a rough morphology for increasing excess strain accumulation during stacking, in particular, when the QDs are fully capped or no annealing step is performed.

**B. Thin cap layer thickness and annealing temperature**

Optimization of the growth conditions for formation of linear and well-separated InAs QD arrays is performed on substrates miscut by 2° toward (110). In the series of samples shown in Fig. 3, the thickness of the thin Q1.27 cap layer above the InAs QDs is varied between 0.3 and 1.0 nm and the annealing temperature between 530 and 550 °C. The number of SL periods of 5, InAs amount of 3.2 ML, InAs growth rate of 0.47 ML/s, and Q1.27 spacer layer thickness of 15.3 nm are kept constant. Ordering of InAs QDs in linear arrays along [001] is clearly observed for 0.3–0.7 nm cap layer thickness together with annealing temperatures of 530–550 °C. The scan field is 2.0×2.0 \( \mu \text{m}^2 \) and the height contrast is 10 nm.

**C. InAs amount and growth rate**

In the series of samples shown in Fig. 4, the InAs amount is decreased from 3.2 to 2.1 ML and the InAs growth rate from 0.47 to 0.23 ML/s. Other growth conditions are unchanged with an optimized Q1.27 cap layer thickness of 0.3 nm and annealing temperature of 530 °C. The number of
SL periods is 5. With a decrease of the InAs amount and growth rate the ordering of the QD arrays is improved, as shown in Figs. 4(a)–4(e). The reduction of the InAs amount mainly lowers the QD height and diameter to further reduce excess strain accumulation, while the decreasing InAs growth rate improves the uniformity of the QD arrays which is attributed to enhancement of the In adatom surface migration length.

In the SL template, the critical thickness for InAs QD formation is less than that on bare InGaAsP surfaces and the InAs QDs are larger due to the strain correlated stacking. Therefore, reduction of the InAs amount in the SL template maintains a high QD density, while excess strain accumulation is further suppressed to create straight QD arrays without bends and branches and to reduce the probability of multiple QD array formation and QD coalescence.

D. Number of SL periods

In the series of samples shown in Fig. 5, the number of SL periods is changed between 5 and 11 to fully optimize QD ordering. Other growth conditions are unchanged with an optimized Q1.27 cap layer thickness of 0.3 nm, annealing temperature of 530 °C, 2.1 ML InAs amount, and 0.23 ML/s InAs growth rate. The amount of InAs for QD formation on the SL template surface is 2.6 ML which slightly increases the QD height.

With an increase of the number of SL periods from 5 to 7 [Figs. 5(a) and 5(b)], the QD ordering improves further due to the evolution of the self-organizing process during stacking. Accompanied with the increasing strain accumulation the QDs become larger. No significant improvement is observed when the number of SL periods is increased from 9 to 11 revealing the stability of the SL template. However, the QDs within the arrays become more connected, as depicted in Figs. 5(c) and 5(d), most probably due to a deeper lateral strain field modulation.

FIG. 4. (Color online) [(a)–(e)] AFM images of 2.1–3.2 ML InAs QD arrays on the five period 2.1–3.2 ML InAs/0.3+15.3 nm Q1.27 SL template (annealing temperature of 530 °C) with InAs amounts and growth rates of (a) 3.2 ML and 0.47 ML/s, (b) 2.8 ML and 0.47 ML/s, (c) 2.6 ML and 0.35 ML/s, (d) 2.4 ML and 0.23 ML/s, and (e) 2.1 ML and 0.23 ML/s. The scan field is 2.0×2.0 μm² and the height contrast is 20 nm.

FIG. 5. (Color online) [(a)–(d)] AFM images of 2.6 ML InAs QD arrays on the 2.1 ML InAs/0.3+15.3 nm Q1.27 SL template (InAs growth rate of 0.23 ML/s and annealing temperature of 530 °C) with numbers of SL periods of (a) 5, (b) 7, (c) 9, and (d) 11. The scan field is 2.0×2.0 μm² and the height contrast is 20 nm.

E. SL template surface and GaAs interlayer

The surface of the optimized SL template providing the highest degree of QD ordering is shown in Fig. 6(a). The number of SL periods is 7. The surface of the SL template shows a very shallow height modulation with mounds elongated along [001] and a lateral periodicity of 130–180 nm, corresponding to the lateral periodicity of the QD arrays. This shallow height modulation is superimposed on the typical height modulation extending over micrometer length scales, shown in Fig. 6(b). Hence, though the SL template surface does not perfectly planarize, the small height modulation of only 0.7–1.0 nm, corresponding to 3.0–4.0 ML, excludes the QD ordering due to preferential nucleation at multilayer high step edges formed due to step bunching. This is further confirmed by the unchanged lateral periodicity of the QD arrays as functions of the number of SL periods, i.e., layer thickness and substrate miscut, discussed above, which both would affect the lateral periodicity of multilayer high steps.

For QD arrays with PL emitting in the 1.55 μm wavelength region a 0.8 ML GaAs interlayer is inserted beneath the QDs on the SL template surface. The GaAs interlayer does not affect the QD ordering, as shown in Fig. 6(c). The role of the GaAs interlayer is to suppress As/P exchange during InAs growth to reduce the QD height from...
7–11 to 4–5 nm and, therefore, the emission wavelength from far above 1.6 to 1.55 μm at RT into the 1.55 μm region.9,10

IV. OPTICAL PROPERTIES OF LINEAR QD ARRAYS

A. Temperature dependence

Figure 7 shows the PL spectra taken at 4.8 K and RT of the capped InAs QD arrays on the optimized SL template with GaAs interlayer and of the SL template without QDs on top for reference. The PL spectrum measured at 4.8 K reveals a PL peak of the QD arrays at 1475 nm and of the SL template at 1379 nm. The PL peak of the QD arrays at RT is redshifted to 1548 nm with a shoulder at the short wavelength side stemming from the SL template which reveals a PL peak at 1478 nm. The PL efficiency drops by about three orders of magnitude between 4.8 K and RT.

The temperature dependences of the PL peak energy and FWHM of the capped 2.6 ML InAs QD arrays with 0.8 ML GaAs interlayer on the optimized SL template and of the capped 2.6 ML single InAs QD layer on 100 nm Q1.27. The dashed line is the temperature dependence of the InAs band gap energy according to the Varshni law.

redshifted to 1548 nm with a shoulder at the short wavelength side stemming from the SL template which reveals a PL peak at 1478 nm. The PL efficiency drops by about three orders of magnitude between 4.8 K and RT.

The temperature dependences of the PL peak energy and FWHM of the InAs QD arrays are plotted in Fig. 8(a) and those of a single InAs QD layer grown on a bare 100 nm Q1.27 layer in Fig. 8(b) for comparison. InAs amount and growth rate of 2.6 ML and 0.23 ML/s, respectively, are the same for both cases. The QD arrays reveal a PL peak energy which follows closely the band gap energy dependence [Varshni law for InAs Ref. 20] and a FWHM which monotonically increases with temperature. On the other hand, the PL peak energy of the single QD layer reveals an increased low-energy shift in the temperature range of 100–200 K which is accompanied by a pronounced minimum of the FWHM. This behavior is typical for inhomogeneous QD ensembles due to thermally activated carrier redistribution in this temperature range preferentially from smaller (higher energy) to larger (lower energy) QDs and the equilibration of the carrier distribution at higher temperatures.11,12 The much less evident thermally activated carrier redistribution in the QD arrays is an indication of lateral electronic coupling of the closely spaced QDs within the arrays allowing carrier redistribution independent of the temperature by providing a quasicontinuous density of states such as in quantum wires.

FIG. 6. (Color online) (a) AFM image of the seven period 2.1 ML InAs/0.3+15.3 nm Q1.27 optimized SL template surface (InAs growth rate of 0.23 ML/s and annealing temperature of 530 °C). (b) AFM line scan along [010] of the SL template surface, as indicated in (a). (c) AFM image of the 2.6 ML InAs QD arrays on the optimized SL template with 0.8 nm GaAs interlayer inserted beneath the QD arrays. The scan field is 2.0 × 2.0 μm² and the height contrast is 10 nm.

FIG. 7. (Color online) PL spectra of the capped 2.6 ML InAs QD arrays on the optimized SL template and bare SL template without QD arrays taken at 4.8 K and room temperature.

FIG. 8. (Color online) (a) Temperature dependences of the PL peak energy and PL linewidth of the capped 2.6 ML InAs QD arrays with 0.8 ML GaAs interlayer on the optimized SL template and (b) of the capped 2.6 ML single InAs QD layer on 100 nm Q1.27. The dashed line is the temperature dependence of the InAs band gap energy according to the Varshni law.
A small degree of carrier localization accounts for the small deviation of the PL peak energy from the Varshni law and the small plateau in the FWHM around 150 K.

The integrated PL intensities of both the QD arrays and the single QD layer, shown in Figs. 9a and 9b, are roughly constant up to 130 K and exponentially decrease at a higher temperature due to thermal quenching. The activation energy of the PL quenching is derived by fitting the integrated PL intensity to

$$I_{PL} = C/[1 + A \exp(-E_a/k_B T)]$$

where $k_B$ is the Boltzmann constant, $T$ is the temperature, and $A$ and $C$ are the fitting parameters. The activation energies are 183 meV for the QD arrays and 197 meV for the single QD layer. These are consistent with the energy differences of the QD ground state energy and the Q1.27 barrier band gap energy, which are 188 meV for the QD arrays and 203 meV for the single QD layer. Hence, the thermal quenching of the PL intensity is due to thermally activated escape of carriers from the QDs into the Q1.27 barrier. This reveals the absence of nonradiative recombination centers in the QDs contributing to the thermal quenching, evidencing the excellent optical and structural quality of the InAs QD arrays similar to single layers of QDs.

B. Polarization dependence

The linear polarization-dependent PL spectra taken from the sample surface of the QD arrays and single QD layer are shown in Figs. 10a and 10b. The PL of the QD arrays is polarized along [001] in (a) and along [01-1] in (b).
V. CONCLUSION

We have realized linear InAs QD arrays based on self-organized anisotropic strain engineering of an InAs/InGaAsP SL template on InP (100) in CBE and determined the optimized growth conditions for ordering. During SL template formation, InAs QD growth, thin InGaAsP capping, annealing, InGaAsP overgrowth, and stacking produce wire-like InAs structures due to anisotropic surface migration and lateral and vertical strain correlations. InAs QD ordering is governed by the lateral strain field modulation which builds up during SL template formation. The presence of linear QD arrays depends on the substrate miscut altering the adatom surface migration. The optimum substrate miscut is 2° toward [001] for which the InGaAsP cap layer thickness, annealing temperature, InAs amount and growth rate, and number of SL periods are optimized for the formation of straight and well-separated arrays along [001]. The InAs QD arrays exhibit excellent PL emission up to room temperature which is tuned into the 1.55 µm telecommunications wavelength region through the insertion of ultrathin GaAs interlayers. Temperature dependent PL measurements indicate lateral electronic coupling of the QDs in the arrays which is supported by the linear polarization of the PL along the arrays.

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