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Direct imaging of self-organized anisotropic strain engineering for improved one-dimensional ordering of (In,Ga)As quantum dot arrays

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Single (In,Ga)As quantum dot (QD) arrays are formed on GaAs (100) substrates by self-organized anisotropic strain engineering of an (In,Ga)As/GaAs quantum wire (QWR) superlattice (SL) template in molecular beam epitaxy. The crucial steps in QWR template evolution, i.e., elongated QD formation at elevated temperature, thin GaAs capping, annealing, and stacking, are directly imaged by atomic force microscopy (AFM). AFM reveals a very smooth connection of the QDs into QWRs upon annealing. In addition, AFM shows the presence of height and width fluctuations of the QWRs with a significant number of bends and branches. These are attributed to excess strain accumulation during formation of the QWR template. By reducing the amount of (In,Ga)As and increasing the GaAs separation layer thickness in each SL period, a dramatic improvement of the uniformity of the QWR template is achieved. On the improved QWR template, well-defined one-dimensional single (In,Ga)As QD arrays are formed which are straight over more than 1 μm and extended to over 10 μm length with a small number of branches. After capping, the QD arrays exhibit clear photoluminescence emission up to room temperature without increase of the peak width. © 2004 American Institute of Physics. [DOI: 10.1063/1.1631069]

I. INTRODUCTION

Epitaxial growth of strained (In,Ga)As on GaAs (100) above the critical layer thickness results in nucleation of coherent quantum dots (QDs) in the Stranski–Krastanov (S–K) mode which are randomly distributed on the surface.^{1–3} For many applications,^{4,5} however, it is desirable to arrange these QDs into laterally ordered arrays without degradation of their structural, electronic, and optical properties. We have recently introduced a concept for the lateral ordering of QDs in one-dimensional arrays on planar singular GaAs (100) substrates by molecular beam epitaxy (MBE), which fulfils these requirements.⁶ The concept is based on self-organized anisotropic strain engineering of an (In,Ga)As quantum wire (QWR) superlattice (SL) template.^{6–9} Single InAs QD arrays were created due to the lateral surface strain field modulation generated by the underlying template, which exhibit excellent optical properties at low temperature.⁶ Hence, this self-organized ordering process does not introduce any defects in the QDs which often occurs when applying artificial patterning techniques.^{10–16} The crucial steps for template formation and QD ordering, previously deduced from *in-situ* reflection high-energy electron diffraction (RHEED) and high-resolution x-ray diffraction (XRD), are schematically illustrated in Figs. 1(a)–1(e).^{6–9} They comprise

- (1) Random formation of elongated (In,Ga)As QDs at elevated temperature on the GaAs buffer layer in the S–K growth mode [Fig. 1(a)].¹⁷

- (2) Growth of a thin GaAs capping layer. The spotty RHEED pattern indicates that the surface does not planarize [Fig. 1(b)].⁸
- (3) Annealing at higher temperature. The QDs further elongate and become connected due to anisotropic adatom surface migration and In desorption. The latter is balanced by the thin GaAs cap layer of step (2) to reduce the strain for a uniform QD connection.^{8,9} The streaky RHEED pattern indicates planarization of the surface. QWRs form [Fig. 1(c)].
- (4) Growth of the GaAs separation layer. The thickness is chosen to preserve the lateral strain field modulation from the buried QWRs at the surface [Fig. 1(d)] due to vertical strain mediation.¹⁸
- (5) Growth of the subsequent (In,Ga)As QD layer. The lateral strain minima on the GaAs surface reduce the lattice mismatch and induce strain gradient driven In adatom migration. The QDs preferentially nucleate in the center above the QWRs underneath.¹⁸ One-dimensional QD arrays form [Fig. 1(e)].

By repeating these steps in (In,Ga)As/GaAs SL growth, the length of the QWRs increases and their lateral ordering improves due to the vertical, strain correlated stacking. For more than ten SL periods, the length of the QWRs exceeds several microns and a well-defined lateral periodicity of the order of 100 nm is developed.⁶

In this study, we directly image each of these steps by atomic force microscopy (AFM) providing first hand insight in the template formation process. In addition, AFM reveals the presence of height and width fluctuations of the QWRs with a significant number of bends and branches. These fluctuations are attributed to excess strain accumulation in the

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FIG. 1. (a)–(e) Schematic illustration of (In,Ga)As QWR template formation and QD ordering. (a) Random formation of elongated (In,Ga)As QDs, (b) growth of the thin GaAs cap layer, (c) annealing at higher temperature, (d) growth of the GaAs separation layer, and (e) growth of the subsequent (In,Ga)As QD layer.

QWR template. Hence, we achieve a dramatic improvement of the uniformity of the QWR template by minimizing the strain accumulation through reduction of the amount of (In,Ga)As to a minimum above the critical thickness for island formation and increase of the GaAs separation layer thickness in each SL period, which is confirmed by high-resolution XRD and photoluminescence (PL) measurements. On the improved QWR template, the long-range ordering of the one-dimensional single (In,Ga)As QD arrays is dramatically improved as well. They are straight over more than $1\ \mu\text{m}$ and extended to over $10\ \mu\text{m}$ length with a small number of branches. These QD arrays, capped with GaAs, exhibit clear PL emission up to room temperature without increase of the peak width.

II. EXPERIMENTAL DETAILS

The samples were grown by conventional solid-source MBE on GaAs (100) substrates with miscut smaller than 0.05° . Before the (In,Ga)As/GaAs SL template, a 200 nm thick GaAs buffer layer was grown at 580°C . For template formation, each SL period comprised the following sequence: (1) growth of 2.3 nm (or 1.8 nm) $\text{In}_{0.41}\text{Ga}_{0.59}\text{As}$ (nominal supply) at 540°C , (2) growth of 0.7 nm GaAs cap at 540°C without growth interruption, (3) annealing at 580°C for 2 min, and (4) growth of 12.3 nm (or 15.3 nm) GaAs at 580°C . The two-dimensional to three-dimensional growth mode transition was observed for both, the 2.3 and 1.8 nm (In,Ga)As supply. The number of SL periods was 15–16. For formation of the QD arrays 2.3 nm (or 1.8 nm)

FIG. 2. (a)–(d) AFM images (scan field: $1\ \mu\text{m}\times 1\ \mu\text{m}$, black-to-white height contrast: 20 nm) of the surfaces during formation of the 16th period of the (In,Ga)As 2.3 nm/GaAs 13.0 nm QWR template (a) 2.3 nm $\text{In}_{0.41}\text{Ga}_{0.59}\text{As}$ grown at 540°C , (b) 0.7 nm GaAs cap deposited at 540°C without growth interruption, (c) annealing at 580°C for 2 min, and (d) 12.3 nm GaAs grown at 580°C .

$\text{In}_{0.41}\text{Ga}_{0.59}\text{As}$ was deposited on top of the last GaAs layer at 540°C . The QD arrays were capped with 100 nm GaAs (20 nm at 540°C plus 80 nm at 580°C) for the PL measurements. The growth rates of GaAs and $\text{In}_{0.41}\text{Ga}_{0.59}\text{As}$ were 0.054 and 0.092 nm/s, respectively, which were calibrated by high-resolution XRD of (In,Ga)As/GaAs SL structures grown at 480°C . The As_4 beam equivalent pressure was kept at 1.0×10^{-5} Torr. The structural properties of the samples were characterized by AFM and high-resolution XRD in air. For the PL measurements, the 532 nm line of a Nd–YAG laser was used as excitation source with an excitation power density of $0.2\ \text{W}/\text{cm}^2$. The PL signal was dispersed by a single monochromator and detected by a cooled InGaAs charge-coupled device.

III. DIRECT IMAGING OF SELF-ORGANIZED ANISOTROPIC STRAIN ENGINEERING

Figure 2 shows the AFM images of the surfaces after the different stages of formation of the 16th period of the (In,Ga)As 2.3 nm/GaAs 13.0 nm QWR SL template: (a) $\text{In}_{0.41}\text{Ga}_{0.59}\text{As}$, (b) GaAs cap, (c) annealing, and (d) GaAs separation layer. The corresponding AFM line scans in Figs. 3(a)–3(d) are taken along [011] (left side, perpendicular to the QD arrays and QWRs) and along [0-11] (right side, following the top of the QD arrays and QWRs). One-dimensional QD arrays are observed in Fig. 2(a), indicating fully developed QWRs in the 15th SL period underneath.⁶

FIG. 3. (a)–(d) AFM line scans (scan length: 500 nm) of the surfaces imaged in Figs. 2(a)–(d). Left side: line scans along [011] perpendicular to the QD arrays and QWRs. Right side: line scans along [0-11] following the top of the QD arrays and QWRs.

The AFM image of the QDs in the first SL period (not shown here) reveals a random site distribution as commonly observed. The uniformity of the QWRs and QD arrays improves with the number of SL periods, as shown previously.^{6,7} After growth of the GaAs cap layer [Fig. 2(b)], the QDs are still visible with only marginal changes in shape and size, as quantified from the comparison of the AFM line scans in Figs. 3(a) and 3(b). After annealing [Fig. 2(c)], the surface becomes rather flat with QWR like structures along the [0-11] direction. The root-mean square (RMS) roughness over a 500 nm $\times$ 500 nm area is reduced from 2.2 nm [Fig. 2(b)] to 0.6 nm [Fig. 2(c)]. Most notably, the RMS roughness along a 500-nm-line on top of the QWRs [Fig. 3(c)] is only 0.2 nm. The smoothness of the QWR like structures affirms that the QDs in the array have uniformly connected. After growth of the GaAs separation layer, the surface develops asymmetric mounds elongated along [0-11] [Figs. 2(d) and 3(d)], which is typical for the reported growth instability on singular GaAs (100).¹⁹ The much larger length and lateral ordering of the QD arrays on top [closing the sequence to Fig. 2(a)] confirm that the QD ordering is not caused by morphological features like step edge induced nucleation but originates from the uniform lateral strain field modulation on the surface of the QWR template.

IV. IMPROVEMENT OF QWR TEMPLATE UNIFORMITY

Although the QDs become smoothly connected into QWRs after annealing, large width and height fluctuations of the QWRs together with a significant number of bends and branches are visible in the case of the (In,Ga)As 2.3 nm/GaAs 13.0 nm QWR template [Fig. 2(c)]. These are attributed to excess strain accumulation in the QWRs. Since elastic strain relaxation cannot occur along straight QWRs, an excess strain field will result in the observed fluctuation in width and height (and ultimately breaking up of the QWRs into QDs) and/or bends and branches to release the strain.

FIG. 4. (a)–(c) XRD spectra around the asymmetric (311) glancing exit reflection of the (In,Ga)As QWR templates A–C. The black (gray) spectra are measured with the x-ray beam parallel to the [011] ([0-11]) direction.

The excess strain accumulation is mainly caused by supply of (In,Ga)As far above the critical thickness and too thin GaAs separation layers between successive (In,Ga)As layers. Moreover, the excess supply of (In,Ga)As results in the nucleation of QDs not only at the most preferable sites on top of the center of the QWRs underneath but also aside, generating regions of multiple QD arrays and/or QD coalescence. In these regions the strain field in the QWRs after annealing is locally enhanced even more and, in addition, they promote the formation of multiple QD arrays in the next (In,Ga)As layer due to vertical strain mediation. The excess strain field is therefore most effectively carried on and accumulated in SL growth through thin GaAs separation layers favoring strong strain coupling between subsequent (In,Ga)As layers. Hence, formation of straight and uniform QD arrays requires the reduction of the strain field by reducing the supply of (In,Ga)As to a minimum above the critical thickness for island formation and by increasing the GaAs separation layer thickness while preserving sufficient vertical strain correlation between the (In,Ga)As layers for ordering.

Figure 4 shows the high-resolution XRD spectra in the vicinity of the (311) glancing exit reflection (a) of the 15-periods $\text{In}_{0.41}\text{Ga}_{0.59}\text{As}$ 2.3 nm/GaAs 13.0 nm QWR template A, (b) of the 15-periods $\text{In}_{0.41}\text{Ga}_{0.59}\text{As}$ 1.8 nm/GaAs 13.0 nm QWR template B with reduced (In,Ga)As thickness, and (c) of the 15-periods $\text{In}_{0.41}\text{Ga}_{0.59}\text{As}$ 1.8 nm/GaAs 16.0 nm QWR template C with reduced (In,Ga)As and increased GaAs

FIG. 5. (a)–(c) PL spectra taken at 5 K of the QWR templates A–C.

separation layer thickness. In this geometry, XRD is most sensitive to detect the lateral periodicity of modulated structures with maximized coherent path and can be evaluated like a multiple-slit Fraunhofer diffraction.²⁰ With the x-ray beam parallel to the [011] direction clear satellite peaks are observed due to the lateral periodicity together with the vertical correlation of the stacked QWRs in all samples. As the strain accumulation is reduced from template A to C the satellite peaks sharpen and their peak-to-valley ratio increases. This clearly indicates the improved uniformity of the QWRs with respect to lateral periodicity and strain distribution. Also in the direction of the QWRs, the narrower 0th order peaks with the x-ray beam parallel to [0-11] for templates B and C compared to that of template A support the improved homogeneity of the QWRs along their length. The low temperature PL spectra taken at 5 K of samples A–C are shown in Figs. 5(a)–5(c). The PL peak energies and peak widths are (a) 1.33 eV, 81 meV, (b) 1.39 eV, 51 meV, and (c) 1.41 eV, 42 meV. The weak PL lines around 1.49 eV stem from GaAs bulk. The decrease of the PL peak width with reduced strain accumulation confirms the improved uniformity of the QWR templates.

V. FORMATION OF SINGLE QD ARRAYS

The AFM images of the (In,Ga)As QD arrays formed at 540 °C on the 15-periods (In,Ga)As QWR templates A–C are depicted in Figs. 6(a)–6(c). The amount of supplied $\text{In}_{0.41}\text{Ga}_{0.59}\text{As}$ for QD array formation is 2.3 nm on template A and 1.8 nm on templates B and C. The improved uniformity of the QWR templates directly relates to that of the QD arrays from (a) to (c) with reduced number of multiple QD arrays and bends. In case of template B with less amount of (In,Ga)As and 13 nm GaAs separation layer thickness, however, a rather large number of the branches [see one in the

FIG. 6. AFM images (a) of the (In,Ga)As 2.3 nm QD arrays grown at 540 °C on QWR template A, (b) of the (In,Ga)As 1.8 nm QD arrays grown at 540 °C on QWR template B, and (c) of the (In,Ga)As 1.8 nm QD arrays grown at 540 °C on QWR template C. The scan field is $1\text{ }\mu\text{m} \times 1\text{ }\mu\text{m}$.

center of Fig. 6(b)] is still visible with an area density of $2.2\text{ }\mu\text{m}^{-2}$. For increased GaAs separation layer thickness of 16.0 nm [template C, Fig. 6(c)] the area density of the branches is significantly reduced to $1.4\text{ }\mu\text{m}^{-2}$. This is additionally reflected by the two-dimensional fast Fourier transform analysis of $5\text{ }\mu\text{m} \times 5\text{ }\mu\text{m}$ AFM images of the QD arrays on templates A–C, shown in Figs. 7(a)–7(c). From (a) to (c), the two bright spots in the center, due to the lateral periodicity of the QD arrays, become better resolved and the faint background is reduced. The single QD arrays in Figs. 6(c) and 7(c) are perfectly straight over more than $1\text{ }\mu\text{m}$ and most of the QD arrays are extended over $10\text{ }\mu\text{m}$ length with a small number of branches.

VI. OPTICAL PROPERTIES OF SINGLE QD ARRAYS

Figure 8 shows the temperature-dependent PL spectra of the capped (In,Ga)As (1.8 nm) QD arrays on template C. The corresponding PL intensity and peak width are plotted in Figs. 9(a) and 9(b). At 5 K, the PL emission from the QD arrays is centered at 1.21 eV with peak width of 72 meV. The PL line at 1.37 eV stems from the QWR template. The PL of the QWR template vanishes around 100 K which is accompanied by a slight increase of the PL intensity of the QD arrays, indicating thermally activated carrier transfer from

FIG. 7. Two-dimensional fast Fourier transform analysis of $5\ \mu\text{m} \times 5\ \mu\text{m}$ AFM images (a) of the (In,Ga)As 2.3 nm QD arrays on QWR template A, (b) of the (In,Ga)As 1.8 nm QD arrays on QWR template B, and (c) of the (In,Ga)As 1.8 nm QD arrays on QWR template C.

FIG. 8. Temperature-dependent PL spectra of the capped (In,Ga)As 1.8 nm QD arrays grown at $540\ ^\circ\text{C}$ on QWR template C.

FIG. 9. (a) PL peak intensity and (b) PL peak width of the capped (In,Ga)As 1.8 nm QD arrays on QWR template C as a function of temperature.

the QWR template to the QD arrays. The PL emission from the QD arrays is clearly visible up to room temperature with a drop in intensity of about four orders of magnitude. The PL peak width undergoes a minimum around 200 K which is commonly observed for ensembles of inhomogeneous QDs due to carrier redistribution.^{21,22} Most notably, the PL peak width of the QD arrays at room temperature of 70 meV does not exceed the low temperature value (72 meV) as is expected for QDs with strong carrier confinement.

VII. CONCLUSION

We have directly imaged by atomic force microscopy (AFM) the formation of (In,Ga)As quantum wire (QWR) superlattice (SL) templates based on self-organized anisotropic strain engineering for the creation of uniform one-dimensional single (In,Ga)As quantum dot (QD) arrays by molecular beam epitaxy (MBE) on GaAs (100) substrates. In addition to the very smooth connection of the QDs into QWRs upon annealing, AFM revealed the presence of excess strain accumulation to produce fluctuations in height and width of the QWRs and a significant number of the bends and branches, which directly affect the uniformity of the QD arrays. High-resolution x-ray diffraction (XRD) and photoluminescence (PL) measurements proved that the uniformity of the QWR template is drastically improved by reducing the amount of (In,Ga)As and increasing the GaAs separation layer thickness in SL growth. The QD arrays formed on top of the optimized template are well separated, perfectly straight over more than $1\ \mu\text{m}$ and extended to over $10\ \mu\text{m}$ length with a small number of branches. The QD arrays exhibit clear PL emission up to room temperature with the PL peak width not exceeding that at low temperature. Hence, self-organized anisotropic strain engineering is established as a route for formation of well-defined QD arrays with high structural and optical quality.

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