Complex quantum dot arrays formed by combination of self-organized anisotropic strain engineering and step engineering on shallow patterned substrates

Citation for published version (APA):

Mano, T., Nötzel, R., Zhou, D., Hamhuis, G. J., Eijkemans, T. J., & Wolter, J. H. (2005). Complex quantum dot arrays formed by combination of self-organized anisotropic strain engineering and step engineering on shallow patterned substrates. *Journal of Applied Physics*, 97(1), 014304-1/5. [014304].
<https://doi.org/10.1063/1.1823578>

DOI:

[10.1063/1.1823578](https://doi.org/10.1063/1.1823578)

Document status and date:

Published: 01/01/2005

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.
- The final author version and the galley proof are versions of the publication after peer review.
- The final published version features the final layout of the paper including the volume, issue and page numbers.

[Link to publication](#)

General rights

Copyright and moral rights for the publications made accessible in the public portal are retained by the authors and/or other copyright owners and it is a condition of accessing publications that users recognise and abide by the legal requirements associated with these rights.

- Users may download and print one copy of any publication from the public portal for the purpose of private study or research.
- You may not further distribute the material or use it for any profit-making activity or commercial gain
- You may freely distribute the URL identifying the publication in the public portal.

If the publication is distributed under the terms of Article 25fa of the Dutch Copyright Act, indicated by the "Taverne" license above, please follow below link for the End User Agreement:

www.tue.nl/taverne

Take down policy

If you believe that this document breaches copyright please contact us at:

openaccess@tue.nl

providing details and we will investigate your claim.

Complex quantum dot arrays formed by combination of self-organized anisotropic strain engineering and step engineering on shallow patterned substrates

T. Mano,^{a)} R. Nötzel, D. Zhou, G. J. Hamhuis, T. J. Eijkemans, and J. H. Wolter
eiTT/COBRA, Inter-University Research Institute, Eindhoven University of Technology, 5600MB Eindhoven, The Netherlands

(Received 26 May 2004; accepted 28 September 2004; published online 9 December 2004)

One-dimensional (In,Ga)As quantum dot (QD) arrays are created on planar singular, vicinal, and shallow mesa-patterned GaAs (100) substrates by self-organized anisotropic strain engineering of an (In,Ga)As/GaAs quantum wire (QWR) superlattice template in molecular beam epitaxy. On planar singular substrates, highly uniform single QD arrays along $[0-11]$ are formed. On shallow $[0-11]$ and $[011]$ stripe-patterned substrates, the generated type-*A* and -*B* steps distinctly affect the surface migration processes which are crucial for QWR template development, i.e., strain-gradient-driven In adatom migration along $[011]$ and surface-reconstruction-induced Ga/In adatom migration along $[0-11]$. In the presence of both type-*A* and -*B* steps on vicinal substrates misoriented towards $[101]$, the direction of adatom migration is altered to rotate the QD arrays. This establishes the relationship between self-organized anisotropic strain and step engineering, which is exploited on shallow zigzag-patterned substrates for the realization of complex QD arrays and networks with well-positioned bends and branches, exhibiting high structural and optical quality.
 © 2005 American Institute of Physics. [DOI: 10.1063/1.1823578]

I. INTRODUCTION

The fabrication of self-assembled quantum dots (QDs) has been intensively investigated in the last decade for basic physics and device applications.^{1,2} Among the hurdles which have to be overcome, lateral ordering of self-assembled QDs is one of the most challenging issues. Commonly, artificial patterning techniques are applied for the position control of QDs.³⁻⁶ These techniques, however, often degrade the structural and optical properties of the QDs. To solve this problem, we have introduced a concept for the lateral ordering of QDs based on self-organized anisotropic strain engineering of an (In,Ga)As/GaAs quantum wire (QWR) superlattice (SL) template in molecular beam epitaxy (MBE).⁷⁻¹¹ Multiple and single one-dimensional InAs and (In,Ga)As QD arrays along $[0-11]$, which exhibit excellent optical properties up to room temperature, have been realized on planar singular GaAs (100) substrates due to the laterally modulated strain field generated by the underlying QWR SL templates. For clarity, we briefly recall the essential steps for template formation and QD ordering.^{8,9}

(1) Randomly distributed (In,Ga)As QDs form in the Stranski-Krastanov (SK) growth mode on a GaAs buffer layer.

(2) A thin GaAs cap layer is grown.

(3) The next step is annealing at higher temperature. The QDs elongate and connect due to preferential anisotropic Ga/In adatom surface migration along $[0-11]$ on the

(2×4) reconstructed GaAs (100) surface, accompanied by In desorption. QWRs along $[0-11]$ form.

(4) Growth of a GaAs separation layer takes place.

(5) This is followed by growth of the subsequent (In,Ga)As QD layer. The QDs preferentially nucleate in the lateral strain field minima above the QWRs due to the strain-gradient-driven In adatom migration preferentially along $[011]$. This is orthogonal to the surface-reconstruction-induced adatom migration along $[0-11]$. One-dimensional well-separated QD arrays along $[0-11]$ form.

(6) Then there is repetition of steps (1)–(5) in SL growth. The length of the QWRs increases and the lateral ordering improves due to the vertical-strain-correlated stacking.

In this study, we establish the relationship of self-organized anisotropic strain engineering with step engineering on vicinal and shallow mesa-patterned GaAs (100) substrates for the realization of advanced, complex QD arrays and networks. Type-*A* and -*B* steps, which are generated on the shallow $[0-11]$ and $[011]$ stripe-patterned substrates, differently affect the adatom surface migration processes during QWR SL template development. While type-*A* steps along $[0-11]$ have no significant effect on the strain-gradient-driven In adatom migration along $[011]$, type-*B* steps along $[011]$ strongly hinder the surface-reconstruction-induced Ga/In adatom migration along $[0-11]$ to prevent QWR formation and QD ordering.¹² When both type-*A* and -*B* steps are present on vicinal substrates which are misoriented towards $[101]$, the surface-reconstruction-induced adatom migration is altered to rotate the QD arrays. On shallow zigzag-patterned substrates, this leads to complex QD arrays and networks with well-positioned bends and branches, which exhibit excellent structural and optical quality.

^{a)}Present address: Nanomaterials Laboratory, National Institute for Materials Science, 1-2-1 Sengen, Tsukuba, Ibaraki 305-0047, Japan; electronic mail: MANO.Takaaki@nims.go.jp

FIG. 1. Scheme of the shallow (a) stripe- and (b) zigzag- patterned GaAs (100) substrates.

II. EXPERIMENTAL DETAILS

The samples were grown by conventional solid-source MBE on planar singular, vicinal (2° off towards [101]), and shallow-patterned GaAs (100) substrates. The patterned substrates were prepared by optical lithography and wet chemical etching in the $\text{H}_2\text{SO}_4:\text{H}_2\text{O}_2:\text{H}_2\text{O}$ (1:8:1000) solution.¹³ On the stripe-patterned substrates, the periodic mesas with $5\ \mu\text{m}$ width, $5\ \mu\text{m}$ separation, and $30\ \text{nm}$ height were oriented along $[0-11]$ and $[011]$, as schematically depicted in Fig. 1(a). For the periodic zigzag patterns, $10\ \mu\text{m}$ wide mesas with $30\ \text{nm}$ height were separated by $10\ \mu\text{m}$. The $10\ \mu\text{m}$ long sidewalls were alternately rotated by $+30^\circ$ and -30° off $[0-11]$ [Fig. 1(b)]. After substrate cleaning in concentrated H_2SO_4 and rinsing in ultrapure water, the growth commenced with a $200\ \text{nm}$ thick GaAs buffer layer at 580°C . For the QWR SL template formation, each SL period comprised the following sequence. (1) growth of $1.8\text{--}2.2\ \text{nm}$ $\text{In}_{0.41}\text{Ga}_{0.59}\text{As}$ QDs in the SK mode at 540°C ; (2) growth of a thin $0.7\text{--}0.8\ \text{nm}$ GaAs cap at 540°C ; (3) annealing at 580°C for 2 min under As_4 flux; and (4) growth of a $10.8\text{--}12.3\ \text{nm}$ GaAs separation layer at 580°C . The number of SL periods was 15. On top of the last GaAs layer of the SL, the (In,Ga)As layer was repeated without annealing for QD array formation. For photoluminescence (PL) studies, the (In,Ga)As QD arrays were capped with $50\ \text{nm}$ GaAs at 540°C followed by $100\ \text{nm}$ GaAs at 580°C . The growth rates of GaAs and (In,Ga)As were $0.054\text{--}0.062$ and $0.092\text{--}0.110\ \text{nm/s}$. The As_4 beam equivalent pressure was 1.0×10^{-5} Torr. The structural properties of the samples were characterized by atomic force microscopy (AFM) and high-resolution x-ray diffraction (XRD) in air. For the micro-PL measurements with a spatial resolution of $\sim 5\ \mu\text{m}$, a He-Ne laser with a power of $30\ \mu\text{W}$ was used for excitation through an optical microscope objective. The PL emission was collected through the same optical microscope objective, dispersed by a single monochromator, and detected by a cooled (In,Ga)As charge-coupled device.

FIG. 2. (a) AFM image of the (In,Ga)As QD arrays on planar singular GaAs (100). The black-to-white height contrast is $15\ \text{nm}$. (b) FFT analysis of a $5 \times 5\ \mu\text{m}^2$ AFM image.

III. RESULTS AND DISCUSSION

A. Planar singular GaAs (100) for reference

Figure 2(a) shows the AFM image of the $1.8\ \text{nm}$ $\text{In}_{0.41}\text{Ga}_{0.59}\text{As}$ QD arrays on the 15-period $(1.8\ \text{nm}\ \text{In}_{0.41}\text{Ga}_{0.59}\text{As})/[(0.7+12.3)\ \text{nm}\ \text{GaAs}]$ QWR SL template on planar singular GaAs (100). Growth rates of (In,Ga)As and GaAs are $0.054\ \text{nm/s}$ and $0.092\ \text{nm/s}$. The QD arrays are oriented along $[0-11]$ with an average lateral periodicity of $160\ \text{nm}$ in the perpendicular $[011]$ direction, which is deduced from the fast Fourier transform (FFT) analysis shown in Fig. 2(b). This is consistent with the $180\ \text{nm}$ lateral periodicity of the QWRs in the SL template determined by XRD recorded in the vicinity of the glancing exit (311) reflection. In the $[0-11]$ direction along the arrays, the QDs are closely spaced with an average center-to-center distance of $60\ \text{nm}$.

B. Shallow $[0-11]$ and $[011]$ stripe-patterned GaAs (100)

Figures 3 and 4 show the $1.8\ \text{nm}$ $\text{In}_{0.41}\text{Ga}_{0.59}\text{As}$ QD arrays grown on the 15-period $(1.8\ \text{nm}\ \text{In}_{0.41}\text{Ga}_{0.59}\text{As})/$

FIG. 3. (a) AFM image of the (In,Ga)As QD arrays on $[0-11]$ stripe-patterned GaAs (100). (b)–(d) Magnified images of the top, bottom, and slope areas. The black-to-white height contrast is 15 nm.

$[(0.7+12.3) \text{ nm GaAs}]$ QWR SL template on the $[0-11]$ and $[011]$ stripe-patterned substrates. After the GaAs buffer layer growth, the angles between the mesa-slopes and the (100) surface are already below 5° and change only slightly during SL growth [Figs. 3(a) and 4(a)]. This indicates that the slopes consist of (100) terraces and monolayer-height type-A (parallel to $[0-11]$) or type-B (parallel to $[011]$) steps rather than of facets.¹⁴ On the mesa stripes along $[0-11]$, well-ordered QD arrays are found all over the top, bottom, and slope areas [Figs. 3(b)–3(d)]. The size of the QDs is similar to that on the planar singular surface shown in Fig. 2(a) and also the PL spectra are unchanged.¹² The average lateral periodicities

FIG. 4. (a) AFM image of the (In,Ga)As QD arrays on $[011]$ stripe-patterned GaAs (100). (b)–(d) Magnified images of the top, bottom, and slope areas. The black-to-white height contrast is 15 nm.

of the QD arrays are 170 nm (top), 180 nm (bottom), and 190 nm (slope), which are all similar to the average lateral periodicity of the QD arrays in planar areas. In the case of the mesa-stripes along $[011]$, well-ordered QD arrays along $[0-11]$ are found only in the top areas [Fig. 4(b)]. The lateral periodicity of 150 nm is almost the same as that on the planar substrate. Towards the bottom areas of the mesas, the QD distribution becomes rather random, as seen in Fig. 4(c) and the left-hand side of Fig. 4(d), which is accompanied by distinct changes of the PL spectra.¹²

These results provide the basic information on the impact of steps on the development of the QWR SL template, i.e., on the strain-gradient-driven In adatom migration along $[011]$ during QD formation, and on the surface-reconstruction-induced Ga/In adatom migration along $[0-11]$ during annealing.^{10,15,16} The former causes the QWRs to be well-separated and the latter causes the QWRs to be uniform along their length. Hence, since the lateral periodicities of the QD arrays in the top, bottom, and slope areas are similar on the $[0-11]$ mesas, it is evident that the generated type-A steps (parallel to $[0-11]$) do not affect the strain-gradient-driven In migration along $[011]$. Although the ordering of QDs by type-A steps has been reported,^{14,17} the probability of adatom trapping at type-A step edges is low.^{18,19} This explains that the strain field fully governs the QD nucleation sites in the presence of type-A steps, which may not affect much the surface-reconstruction-induced Ga/In migration along $[0-11]$. On the contrary, the QDs are arranged rather randomly in the slope areas of the mesas along $[011]$, indicating that QWRs are not developed in the presence of type-B steps. On (2×4) reconstructed GaAs (100) surfaces, type-B steps provide effective sites for adatom adsorption, unlike to type-A steps.^{18,19} Moreover, the density of type-B steps becomes larger towards the bottom areas of the mesas, where the angle between the slopes and the (100) surface is steeper. As the result, the surface-reconstruction-induced Ga/In migration along $[0-11]$ is hindered during the annealing process and uniform QWRs cannot be formed, although the strain-gradient-driven In migration along $[011]$ may not be affected.¹⁵

C. Vicinal GaAs (100)

The 2.2 nm $\text{In}_{0.41}\text{Ga}_{0.59}\text{As}$ QD arrays formed on the 15-period $(2.2 \text{ nm } \text{In}_{0.41}\text{Ga}_{0.59}\text{As})/[(0.8+10.8) \text{ nm GaAs}]$ QWR SL template on the vicinal (2° off towards $[101]$) substrate are shown in Fig. 5(a). The growth rates of (In,Ga)As and GaAs are 0.062 nm/s and 0.110 nm/s. The QD arrays are rotated by 20° off the $[0-11]$ direction with an average lateral periodicity of 170 nm, determined by the FFT analysis shown in Fig. 5(b). For the present misorientation, the average step direction is along $[001]$, i.e., 45° off $[0-11]$. Indeed, (In,Ga)As QDs, which are directly grown on the GaAs buffer, show some alignment along $[001]$ due to the step-edge-induced QD nucleation. Hence, the smaller rotation angle of the QD arrays on the QWR SL template cannot be simply attributed to the average step-edge direction and related QD nucleation.¹⁶ Microscopically, the $[001]$ step edges are composed of alternating active type-B steps sepa-

FIG. 5. (a) AFM image of the (In,Ga)As QD arrays on vicinal (2° off towards $[101]$) GaAs (100). The black-to-white height contrast is 15 nm. (b) FFT analysis of a $5 \times 5 \mu\text{m}^2$ AFM image.

rated by nonactive type-A steps of equal fractions.^{18,19} During the annealing process, the preferential capture of surface adatoms at the portions of type-B steps, will therefore add a component of migration along $[001]$ [or $[011]$] to the surface-reconstruction-induced migration along $[0-11]$. This results in a net rotation of the direction of the surface adatom migration and, hence, of the QWRs and QD arrays in between the $[0-11]$ and $[001]$ directions. On the other hand, the QD nucleation sites and the strain-gradient-driven migration are again solely governed by the strain field and unaffected by the presence of the type-A and -B steps, as evidenced by the lateral periodicity of the QD arrays, which is comparable to that on planar singular substrates.

D. Shallow zigzag-patterned GaAs (100)

The rotation of the QD arrays in the presence of both type-A and -B steps as a function of the misorientation is utilized for the formation of complex QD arrays and networks. On the shallow zigzag-patterned substrates, stepped slopes inclined by few degrees and oriented by $+30^\circ$ and -30° off $[0-11]$ are developed during GaAs buffer layer

FIG. 6. (a) AFM image of the (In,Ga)As QD arrays on zigzag-patterned GaAs (100). (b) Magnified image of the slope intersections. The black-to-white height contrasts are (a) 40 nm and (b) 20 nm.

growth, in addition to planar (100) surface areas, as depicted in Fig. 6(a). After QWR SL template formation and QD growth, it is clearly observed that the one-dimensional QD arrays on the slopes are rotated by $+16^\circ$ and -16° off $[0-11]$ [Fig. 6(b)], in analogy to the behavior on the vicinal substrate. The smaller rotation angles of the QD arrays, compared to those of the slopes, and the unchanged lateral periodicity, confirm that the direction of the QD arrays is determined by the rotation of the surface migration during annealing in QWR SL template formation due to the presence of both type-A and -B steps, while the QD nucleation sites are controlled by the strain field. The rotation of the QD arrays on the slopes naturally produces the basic building blocks of complex QD networks in a well-defined way. Bends of the QD arrays by $\sim 32^\circ$ are formed at the slope intersections, and periodic arrangements of branches are generated at the intersections of the slopes and the planar areas, which contain QD arrays oriented along $[0-11]$, as demonstrated in Fig. 6(b).

The optical properties of the capped QD arrays on the zigzag-patterned substrate are assessed by micro-PL measurements. Figure 7 shows the PL spectra which are taken from the planar and slope areas. The PL lines at longer and shorter wavelengths in each spectrum stem from the QD arrays and the QWR SL template, respectively. The peak energy and width of the PL emission from the QD arrays in the planar (slope) area are 1.279 eV (1.265 eV) and 52 meV (77 meV). The origin of the small PL redshift and line broadening for the QD arrays in the slope area is attributed to the mixture of the rotated arrays, the bends and branches, gen-

FIG. 7. Micro-PL spectra at 5 K of the capped (In,Ga)As QD arrays on zigzag-patterned GaAs (100) taken from the planar area (black line) and slope area (gray line).

erating larger entities. The integrated PL intensity of the QD arrays on the slope is the same as that of the QD arrays in the planar area. Thus, the optical quality of the complex QD arrays, which are realized on shallow patterned substrates, is not degraded compared to that of QD arrays on planar ones. This is attributed to the fact that the QD nucleation and ordering are exclusively governed by the strain field, which is inherently smooth on the dot-diameter and dot-to-dot distance length scales.⁸ The role of steps induced by the shallow pattern is to modify (rotate) the adatom surface migration during annealing in QWR SL template formation, allowing to design the strain field. The demonstrated high complexity and optical quality of the QD arrays on shallow-patterned substrates highlight the key advantage of self-organized anisotropic strain engineering for QD ordering.

IV. CONCLUSIONS

The formation of ordered (In,Ga)As QD arrays by self-organized anisotropic engineering of an (In,Ga)As/GaAs QWR SL template in MBE has been investigated on planar singular, vicinal, and shallow mesa-patterned GaAs (100) substrates. On planar singular substrates, highly uniform single QD arrays along $[0-11]$ are formed. On shallow $[0-11]$ and $[011]$ stripe-patterned substrates, well-ordered QD arrays are developed with the same lateral periodicity, except on the slopes of the mesas along $[011]$. This indicates that, for QWR SL template formation, type-A steps along $[0-11]$ do not affect the strain-gradient-driven In adatom migration along $[011]$ during QD growth. On the contrary,

type-B steps along $[011]$ drastically hinder the surface-reconstruction-induced Ga/In adatom migration along $[0-11]$ during annealing to prevent QWR formation and QD ordering. On vicinal substrates 2° off towards $[101]$ with alternating type-A and -B steps along $[001]$, the direction of adatom migration during annealing in QWR SL growth is altered to rotate the QD arrays. This has been exploited for the creation of complex QD arrays on shallow zigzag-patterned substrates, where well-positioned bends and branches are naturally introduced at the intersections of the slopes. The optical properties of the QD arrays on the shallow-patterned substrates are not degraded compared to those of QD arrays formed on planar substrates. Hence, the potential of self-organized anisotropic strain engineering, which is guided by step engineering on shallow patterned substrates, has been proven for the creation of advanced QD arrays and networks of high complexity.

¹Y. Arakawa and H. Sakaki, Appl. Phys. Lett. **40**, 939 (1982).

²H. Sakaki, Jpn. J. Appl. Phys., Part 1 **28**, L314 (1989).

³D. S. L. Mui, D. Leonard, L. A. Coldren, and P. M. Petroff, Appl. Phys. Lett. **66**, 1620 (1995).

⁴A. Konkar, A. Madhukar, and P. Chen, Appl. Phys. Lett. **72**, 220 (1998).

⁵W. Seifert, N. Carlsson, A. Petersson, L. E. Wernersson, and L. Samuelson, Appl. Phys. Lett. **68**, 1684 (1996).

⁶Y. Nakamura, O. G. Schmidt, N. Y. Jin-Phillipp, S. Kiravittaya, C. Müller, K. Eberl, H. Gräbeldinger, and H. Schweizer, J. Cryst. Growth **242**, 339 (2002).

⁷W. Ma, R. Nötzel, A. Trampert, M. Ramsteiner, H. Zhu, H.-P. Schönherr, and K. H. Ploog, Appl. Phys. Lett. **78**, 1297 (2001).

⁸T. Mano, R. Nötzel, G. J. Hamhuis, T. J. Eijkemans, and J. H. Wolter, Appl. Phys. Lett. **81**, 1705 (2002).

⁹T. Mano, R. Nötzel, G. J. Hamhuis, T. J. Eijkemans, and J. H. Wolter, J. Appl. Phys. **95**, 109 (2004).

¹⁰T. Mano, R. Nötzel, G. J. Hamhuis, T. J. Eijkemans, and J. H. Wolter, J. Cryst. Growth **251**, 264 (2003).

¹¹Yu. I. Mazur, W. Q. Ma, X. Wang, Z. M. Wang, G. J. Salamo, M. Xiao, T. D. Mishima, and M. B. Johnson, Appl. Phys. Lett. **83**, 987 (2003).

¹²T. Mano, R. Nötzel, G. J. Hamhuis, T. J. Eijkemans, E. Smalbrugge, and J. H. Wolter, Mater. Res. Soc. Symp. Proc. **794**, T4.5.1 (2004).

¹³D. W. Shaw, J. Electrochem. Soc. **128**, 874 (1981).

¹⁴Q. Gong, R. Nötzel, H.-P. Schönherr, and K. H. Ploog, Physica E (Amsterdam) **13**, 1176 (2002).

¹⁵T. Mano, R. Nötzel, G. J. Hamhuis, T. J. Eijkemans, and J. H. Wolter, J. Appl. Phys. **92**, 4043 (2002).

¹⁶Kasu and N. Kobayashi, J. Cryst. Growth **170**, 246 (1997).

¹⁷M. Kitamura, M. Nishioka, J. Oshinowo, and Y. Arakawa, Appl. Phys. Lett. **66**, 3663 (1995).

¹⁸H. Yamaguchi and Y. Horikoshi, Jpn. J. Appl. Phys., Part 1 **30**, 802 (1991).

¹⁹S. Tsukamoto and N. Koguchi, J. Cryst. Growth **201/202**, 118 (1999).