

Ordered quantum dot molecules and single quantum dots formed by self-organized anisotropic strain engineering

Citation for published version (APA):

Lippen, van, T., Nötzel, R., Hamhuis, G. J., & Wolter, J. H. (2005). Ordered quantum dot molecules and single quantum dots formed by self-organized anisotropic strain engineering. *Journal of Applied Physics*, 97(4), 044301-1/6. Article 044301. <https://doi.org/10.1063/1.1840098>

DOI:

[10.1063/1.1840098](https://doi.org/10.1063/1.1840098)

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.

Ordered quantum dot molecules and single quantum dots formed by self-organized anisotropic strain engineering

T. van Lippen,^{a)} R. Nötzel, G. J. Hamhuis, and J. H. Wolter

European Institute of Telecommunication Technologies/Communication Technology, Basic Research and Applications (eITT/COBRA) Inter-University Research Institute, Eindhoven University of Technology, 5600 MB Eindhoven, The Netherlands

(Received 19 October 2004; accepted 2 November 2004; published online 20 January 2005)

An ordered lattice of lateral InAs quantum dot (QD) molecules is created by self-organized anisotropic strain engineering of an (In,Ga)As/GaAs superlattice (SL) template on GaAs(311)B by molecular-beam epitaxy, constituting a Turing pattern in solid state. The SL template and InAs QD growth conditions, such as the number of SL periods, growth temperatures, amount and composition of deposited (In,Ga)As, and insertion of Al-containing layers, are studied in detail for an optimized QD ordering within and among the InAs QD molecules on the SL template nodes, which is evaluated by atomic force microscopy. The average number of InAs QDs within the molecules is controlled by the thickness of the upper GaAs separation layer on the SL template and the (In,Ga)As growth temperature in the SL. The strain-correlated growth in SL template formation and QD ordering is directly confirmed by high-resolution x-ray diffraction. Ordered arrays of single InAs QDs on the SL template nodes are realized for elevated SL template and InAs QD growth temperatures together with the insertion of a second InAs QD layer. The InAs QD molecules exhibit strong photoluminescence (PL) emission up to room temperature. Temperature-dependent PL measurements exhibit an unusual behavior of the full width at half maximum, indicating carrier redistribution solely within the QD molecules. © 2005 American Institute of Physics.

[DOI: 10.1063/1.1840098]

I. INTRODUCTION

Self-assembled quantum dots (QDs) formed in the Stranski–Krastanov (SK) growth mode are usually randomly distributed over the wafer surface. The lateral ordering of semiconductor QDs is, however, required for the realization of quantum functional devices for applications such as quantum computing and quantum communication.¹ To control the QD sites, epitaxial growth on artificially patterned substrates has been widely investigated^{2–4} which, however, imposes strong limitations due to the spatial resolution of the lithography and/or etching steps, and easily introduces size fluctuations and defects in the QDs. We have established a concept for the lateral ordering of QDs in one- and two-dimensional arrays by molecular-beam epitaxy (MBE) on planar GaAs(100) (Refs. 5 and 6) and (311)B (Ref. 7) substrates which overcomes these limitations. The concept is based on self-organized anisotropic strain engineering of (In,Ga)As/GaAs superlattice (SL) templates and the lateral ordering of (In,Ga)As QDs on top by local strain recognition. It benefits from the inherent smoothness of the lateral strain field modulations generated on the SL template surfaces on the nanometer length scale, producing ordered QD arrays of excellent structural and optical quality.

Here we present a systematic growth study of the (In,Ga)As/GaAs SL template and of the InAs QDs on GaAs(311)B substrates for optimized ordering into a lattice of lateral InAs QD molecules with a controlled number of QDs. During stacking, the SL template self-organizes into a

highly ordered, two-dimensional (In,Ga)As and, thus, strain field modulation on a mesoscopic length scale. It constitutes a Turing pattern in solid state, which is stable after ten SL periods. The QD molecules formed on the nodes of the SL template exhibit the highest degree of ordering for slightly reduced InAs growth temperature and medium total strain, i.e., In composition and thickness of the (In,Ga)As layers in the SL template, as evaluated by atomic force microscopy (AFM). Insertion of Al-containing layers in the SL template inhibits the evolution of strain modulation and InAs QD ordering. The average number of QDs within the molecules is controlled by the thickness of the upper GaAs separation layer on the SL template and the growth temperature of the (In,Ga)As and thin GaAs cap layers in the SL. For optimized SL template formation and QD ordering the strain-correlated growth is consistently confirmed by high-resolution x-ray diffraction (XRD) in various scattering geometries. In addition, ordered arrays of single InAs QDs on the SL template nodes are realized at elevated temperatures for SL template formation and InAs QD growth together with the insertion of a second InAs QD layer. The QD molecules exhibit excellent photoluminescence (PL) properties up to room temperature (RT). Most interesting, temperature-dependent PL measurements exhibit an unusual behavior of the full width at half maximum (FWHM), indicating carrier redistribution solely within the QD molecules.

II. EXPERIMENTAL DETAILS

The samples were grown by solid source MBE on GaAs (311)B substrates. If not mentioned otherwise, each SL pe-

^{a)}Electronic mail: t.v.lippen@tue.nl

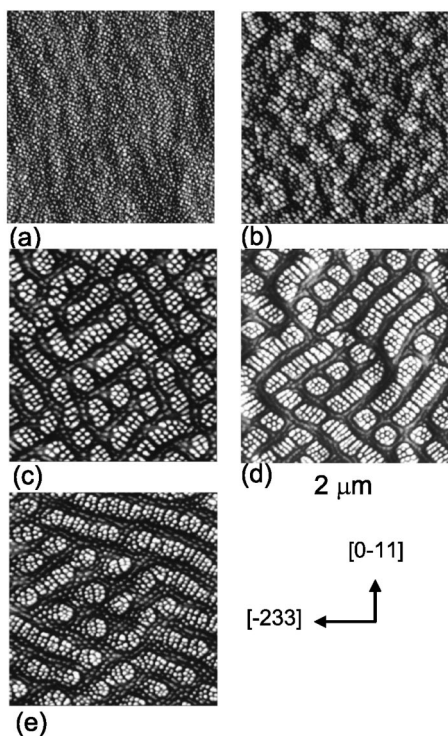


FIG. 1. AFM images of 3.2-nm (In,Ga)As on the (a) 1-, (b) 5-, (c) 10-, and (d) 15-period SL templates. In (e) the growth temperature of the (In,Ga)As and thin GaAs cap layers in the ten-period SL template is increased to 550°C, and the temperature for annealing and GaAs spacer layer growth to 610°C. The scan field is $2 \times 2 \mu\text{m}^2$ and the black-to-white height contrast is 15 nm for all images.

riod comprised 3.2-nm $\text{In}_{0.37}\text{Ga}_{0.63}\text{As}$ grown at 500°C, thin capping by 0.7-nm GaAs at 500°C, annealing for 2 min at 580°C, and growth of a 5.5-nm GaAs spacer layer at 580°C. The number of SL periods was between 1 and 15 in different samples. The growth rates of GaAs and $\text{In}_{0.37}\text{Ga}_{0.63}\text{As}$ were 0.073 and 0.116 nm/s. In further experiments, the In composition of the (In,Ga)As SL template layers was varied between 25% and 45%, and the (In,Ga)As thickness between 3.0 and 5.0 nm. For InAs QD formation on the ten-period SL template, the growth temperature was varied between 450 and 520°C, and the upper GaAs separation layer thickness from 5.5 to 20 nm. InAs was deposited to a nominal thickness of 0.5–0.6 nm at a growth rate of 0.0013 nm/s. The structural properties of the InAs QDs and SL templates were characterized by AFM and XRD in air. For the PL studies the QDs were capped by 200-nm GaAs (20 nm at the InAs QD growth temperature plus 180 nm at 580°C without annealing). The PL was excited by the 512-nm line of a neodymium: yttrium aluminum garnet (Nd:YAG) laser with a power density of 0.2 W/cm². For the temperature-dependent PL measurements a He-flow cryostat was used to control the temperature between 5 and 300 K.

III. SL TEMPLATE EVOLUTION AND STABILITY

The SL template formation, as described in Ref. 7, is briefly recalled for clarity. The AFM images in Figs. 1(a)–1(c) depict the 3.2-nm $\text{In}_{0.37}\text{Ga}_{0.63}\text{As}$ layers on the (In,Ga)As/GaAs SL templates with 1, 5, and 10 periods. Upon stacking, the nanoscale two-dimensional (In,Ga)As

surface modulation for the first SL period⁸ evolves into the distinct mesoscopic mesalike arrangement when the number of SL periods is increased from 5 to 10 due to anisotropic surface migration during annealing (smoothing the mesas to form mesoscopic nodes) and strain-correlated (In,Ga)As stacking [governed by preferential (In,Ga)As accumulation on the nodes due to strain-gradient driven In adatom migration]. The most important result here is the stability of the mesoscopic surface pattern after ten SL periods, as evidenced in Fig. 1(d) for the (In,Ga)As layer on the 15-period SL template with mesas of ~ 200 -nm width, 300-nm lateral periodicity, and 8–10-nm average height.

The morphology in Figs. 1(c) and 1(d) constitutes the Turing pattern⁹ of buried (In,Ga)As quantum disks obtained by metal organic vapor-phase epitaxy (MOVPE).¹⁰ In contrast to MOVPE, where the strain-driven materials reorganization is completed for the first (In,Ga)As layer due to the higher growth temperature, the development of the SL template in MBE provides snapshots of the pattern evolution, which have been rarely observed experimentally.¹¹ Within the general description of reaction–diffusion systems, in the present case, the reaction term may be associated with strain-induced island formation during (In,Ga)As growth and the diffusion term with adatom surface migration during annealing and (In,Ga)As growth guided by the lateral strain field modulation. This is supported by altering the balance between reaction and diffusion when the temperature for (In,Ga)As growth and thin GaAs capping is increased to 550°C and the temperature for annealing and GaAs spacer layer growth to 610°C. This reduces the strain due to enhanced In desorption and increases the adatom surface migration length. As a result, the mesalike pattern aligned $\pm 45^\circ$ off [0-11] transforms into a zigzag or stripelike pattern oriented preferentially along the directions $\sim 70^\circ$ off [0-11], as shown in Fig. 1(e). A quantitative analysis of the pattern formation and pattern transition¹¹ as functions of the growth conditions is, however, beyond the scope of the present evaluation.

IV. INAS QD MOLECULES

InAs QDs form on top of the SL template (i.e., on the upper GaAs layer) in dense and well separated groups, as shown in Fig. 2(a) for 0.6-nm InAs deposited at 500°C on the ten-period SL template. The QDs arrange on the nodes of the SL template where the underlying (In,Ga)As accumulation establishes tensile strain field minima and the related strain-gradient driven In adatom migration for strain-correlated stacking.⁷ The ordering and size uniformity of the QDs within the groups are significantly improved by lowering the InAs growth temperature to 470°C [see the 0.5-nm InAs QD molecules in Fig. 2(b)]. When the temperature is lowered further to 450°C the QD ordering decreases due to the too short In adatom migration length [Fig. 2(c)]. For an InAs growth temperature of 520°C large, elongated mounds are observed in Fig. 2(d), indicating coalescence of the QDs, which is already recognized in Fig. 2(a) for the QDs grown at 500°C in the center of the nodes. Hence, a growth temperature of 470°C is identified for optimum InAs QD order-

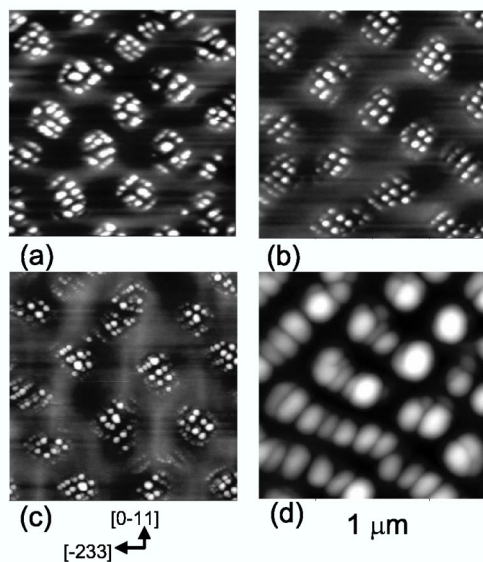


FIG. 2. AFM images of the InAs QDs grown at (a) 500°C, (b) 470°C, (c) 450°C, and (d) 520°C on the ten-period SL template. The scan field is $1 \times 1 \mu\text{m}^2$ for all images and the black-to-white height contrast is 10 nm for (a)–(c) and 20 nm for (d).

ing within the molecules while the ordering is decreased at higher temperature due to QD coalescence and at lower temperature due to the too short In adatom migration length.

V. SL TEMPLATE PROPERTIES

The In composition and thickness of the (In,Ga)As layers in the ten-period SL template are varied and Al-containing layers are inserted in the GaAs spacer layers to elucidate the SL template evolution for QD ordering. The two-dimensional strain field modulation and, hence, (In,Ga)As distribution is probed by the location of the optimized InAs QDs grown at 470°C on top. The scan field of the AFM images is enlarged to provide the best overview on the ordering among the QD molecules. Regarding In composition, the highest degree of ordering of the QD molecules in well separated and ordered groups is observed in a window between 37% and 29% when adjusting the respective (In,Ga)As layer thickness to 3.2 and 4.0 nm to keep the total amount of In constant [see Figs. 3(a) and 3(b), respectively].

Increasing the (In,Ga)As layer thickness to 4.0 nm for an In composition of 37% [Fig. 3(c)], or increasing the In composition to 45% for a layer thickness of 3.0 nm [Fig. 3(d)] leads to less separated QD molecules with more and smaller disordered QDs per group. Also for the low In composition of 25% and 5.0-nm (In,Ga)As layer thickness, disorderly connected QD groups are produced together with larger InAs clusters [Fig. 3(e)]. Therefore, the highest degree of ordering of the QD molecules is achieved at medium total strain, i.e., In composition and thickness of the (In,Ga)As layers in the SL template. A too high total strain opposes the build up of sufficient (In,Ga)As accumulation on the SL template nodes and, hence, the magnitude of the strain field modulation, leading to less separated QD groups with more and smaller QDs of low internal ordering. Similarly, for a too low total strain, a more homogeneous (In,Ga)As layer is

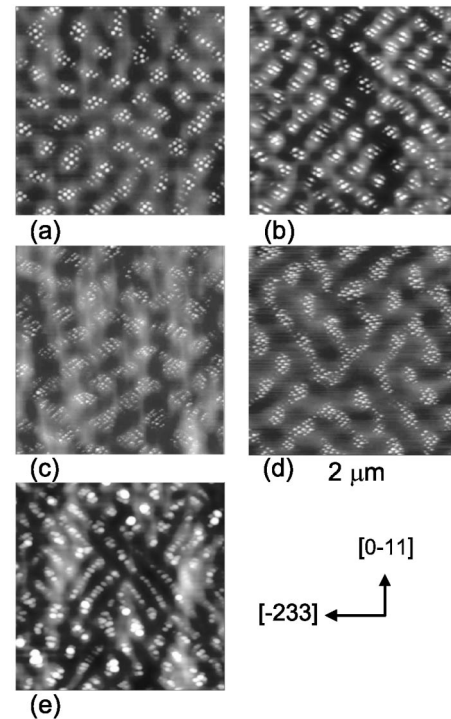


FIG. 3. AFM images of the InAs QDs grown at 470°C on the ten-period SL template with (In,Ga)As layers of (a) 37% (3.2 nm), (b) 29% (4.0 nm), (c) 37% (4.0 nm), (d) 45% (3.0 nm), and (e) 25% (5.0 nm) In composition and thickness. The scan field is $2 \times 2 \mu\text{m}^2$ and the black-to-white height contrast is 15 nm for all images.

maintained and the strain-correlated stacking for the formation of well-defined SL nodes and strain field modulation is not supported.

Inserting AIAs at various locations in the GaAs layers of the SL template significantly alters its formation and the QD ordering. The total spacer layer thickness is kept at 5.5 nm. Depositing two-monolayer (ML) AIAs on top of the SL template directly underneath the InAs QD layer leads to more and smaller QDs per group with less ordering and a significant number in-between [Fig. 4(a)]. This is attributed to a decrease in the In adatom migration length on AIAs, as is the case for lower growth temperature [see Fig. 2(c) for comparison]. When two-ML AIAs covered by 1.0-nm GaAs are introduced beneath each (In,Ga)As layer of the SL template and the InAs QDs on top, a rather smooth mesalike morphology is observed, superimposed by shallow elongated QDs [Fig. 4(b)]. The mesa diameter and periodicity are slightly larger compared to those of the SL template without AIAs. The incorporation of two-ML AIAs on top of the thin GaAs cap layer after annealing in each SL period results in mesas with a slightly smaller diameter and periodicity, covered with shallow InAs QDs and a small number of large InAs clusters [Fig. 4(c)]. Replacing the whole GaAs spacer layer by $\text{Al}_{0.24}\text{Ga}_{0.76}\text{As}$ produces a low density of InAs QDs together with large InAs clusters, which are randomly distributed on the sample surface [Fig. 4(d)].

The diminished InAs QD ordering in the presence of AIAs in the SL template indicates that a well-defined (In,Ga)As and strain field modulation is not established. This is attributed to the short surface migration length of Al

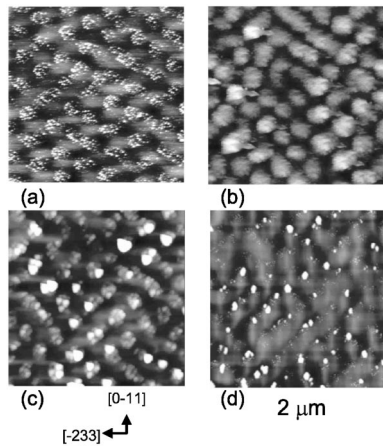


FIG. 4. AFM images of InAs grown at 470°C on the ten-period SL template with Al-containing GaAs spacer layers. (a) InAs QDs directly grown on 2-ML AlAs on the SL template. The black-to-white height contrast is 5 nm. (b) InAs grown on the SL template with 2-ML AlAs covered by 1.0-nm GaAs underneath each (In,Ga)As layer in the SL template and the InAs layer on top. The black-to-white height contrast is 10 nm. (c) InAs grown on the SL template with 2-ML AlAs on each thin GaAs cap layer after annealing. The black-to-white height contrast is 10 nm. (d) InAs grown on the SL template with $\text{Al}_{0.24}\text{Ga}_{0.76}\text{As}$ spacer layers. The black-to-white height contrast is 20 nm. The total spacer layer thickness in the SL template is $5.5\ \mu\text{m}^2$ for all samples. The scan field is $2 \times 2\ \mu\text{m}^2$ for all images.

and the reduced surface migration lengths of In [revealed in Fig. 4(a)] and Ga on AlAs, hindering the strain-induced materials reorganization. The result of Fig. 4(b) with 2-ML AlAs covered by 1.0-nm GaAs beneath the (In,Ga)As SL template layers indicates that a considerable lateral mass transport of GaAs underneath the (In,Ga)As layers is involved in the formation of the strain-modulated (In,Ga)As/GaAs SL template of Fig. 1, which is hindered by the 2-ML AlAs layer. The result of Fig. 4(c) with the 2-ML AlAs deposited on the thin GaAs cap after annealing furthermore indicates that also a significant lateral mass transport during GaAs spacer layer growth is essential for building up the strain-modulated SL template nodes, which is likewise hindered on AlAs. Finally, for (Al,Ga)As spacer layers [Fig. 4(d)] a two-dimensional mesoscopic ordering is entirely suppressed due to the short surface migration length of Al. It is interesting to note that despite of the insertion of thin AlAs layers a two-dimensional mesalike surface morphology is still observed in Figs. 4(b) and 4(c) though with a reduced height, whereas InAs QD ordering is strongly degraded. This underlines the lateral strain field modulation established in Al-free (In,Ga)As/GaAs SL templates to fully govern QD ordering independent of morphological features.

VI. QD NUMBER PER MOLECULE

To control the average number of InAs QDs within the molecules, a reduction of the amount of InAs rather reduces the QD height, leaving the QD number unchanged. On the other hand, the average number of QDs is controlled by the upper GaAs separation layer thickness on the SL template. The number is reduced from 11 for the 5.5-nm-thick separation layer [Fig. 5(a)] to 8.5 for a thickness of 9.5 nm, and to 7.5 for a thickness of 15 nm, depicted in Fig. 5(b). The InAs growth temperature is 470°C for optimized QD ordering

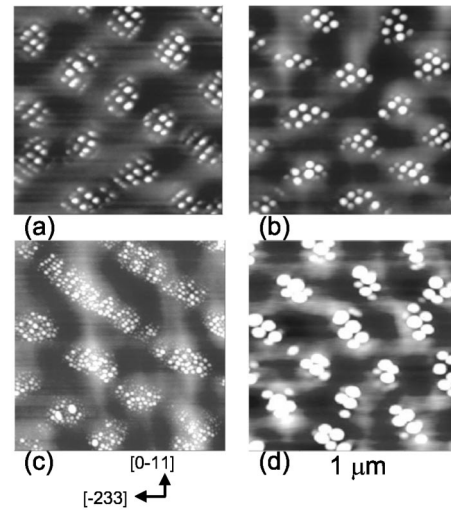


FIG. 5. AFM images of the InAs QD molecules grown at 470°C on the ten-period SL template with an upper GaAs separation layer thickness of (a) 5.5, (b) 15, and (c) 20 nm. In (d) the growth temperature of the (In,Ga)As and thin GaAs cap layers in the ten-period SL template with a 15-nm upper GaAs separation layer thickness is increased to 520°C . The scan field is $1 \times 1\ \mu\text{m}^2$ and the black-to-white contrast is 10 nm for all images.

within the molecules on the ten-period SL template. The reduction of the average number of QDs per molecule with the increasing upper GaAs separation layer thickness on the SL template is attributed to a decrease of the tensile strain field minima, shrinking the effective area for preferred QD formation. When, however, the upper GaAs separation layer thickness is increased to 20 nm, the QD ordering degrades for too shallow strain field minima, shown in Fig. 5(c).

The effective area for QD formation and, thus, the average number of QDs per molecule is further decreased by increasing the (In,Ga)As growth temperature in the SL template, which is attributed to enhanced In migration and In accumulation on the nodes. Figure 5(d) shows the InAs QD molecules deposited at 470°C on the SL template with the (In,Ga)As layers grown at 520°C and a 15-nm-thick upper GaAs separation layer. The average number of QDs per molecule is 4. Hence, by adjusting the thickness of the upper GaAs separation layer and the (In,Ga)As growth temperature in the SL template, well-separated groups of ordered InAs QDs with an adjustable number are realized, which are themselves self-organized into an ordered lattice.

VII. X-RAY DIFFRACTION

High-resolution XRD performed in various scattering geometries consistently confirms the lateral strain field modulation and strain-correlated growth in SL template formation and QD ordering. The XRD spectra of the optimized ten-period SL template of Fig. 1(c) [without the (In,Ga)As layer on top] are recorded in the vicinity of the symmetric (311) reflection with the x-ray beam 45° off [0-11] and along [0-11]; see Fig. 6. Both spectra reveal clear satellite peaks close to the substrate reflection whose spacing provides the periodicity of the lateral strain field modulation in the respective directions.¹² The lateral periodicity along the direction 45° off [0-11] amounts to 350 nm and that along [0-11] to 420 nm. For completeness, grazing exit XRD spectra re-

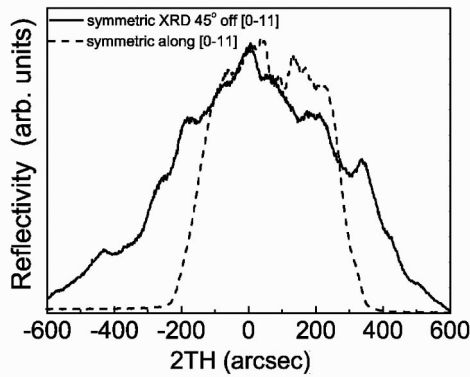


FIG. 6. XRD spectra recorded in the vicinity of the symmetric (311) reflection of the ten-period SL template of Fig. 1(c) without an upper (In,Ga)As layer with the x-ray beam 45° off (solid line) and along (dashed line) [0-11].

recorded in the vicinity of the asymmetric (004) reflection reveal a lateral periodicity along [0-11] of 400 nm. These values coincide well with the lateral periodicity along the direction 45° off [0-11] of the InAs QD molecules on the SL template of 300 nm, which is derived from the two-dimensional fast-Fourier-transform analysis of the AFM images in Figs. 2(a) and 2(b).

VIII. SINGLE QDS

In order to realize single InAs QDs on the nodes of the ten-period SL template, not only the growth temperature of the (In,Ga)As layers in the SL template is increased but also the temperatures for annealing and GaAs spacer layer growth and that for InAs QD deposition. Figure 7(a) shows the AFM image of the InAs QDs grown at 500°C on the SL template with (In,Ga)As and thin GaAs cap growth temperature of 530°C , and annealing and GaAs spacer layer growth at 610°C . The upper GaAs separation layer thickness is 15 nm. The number of QDs on the nodes, however, does not change compared to that in Fig. 5(d) where only the growth of the (In,Ga)As layers is at a higher temperature. The QD groups are less separated and ordered due to the enhanced In desorption in SL growth and a more stripelike pattern with less defined nodes discussed for the SL template in Fig. 1(e).

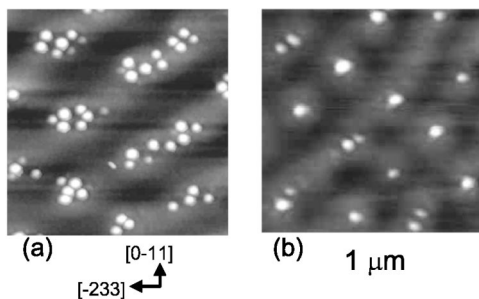


FIG. 7. AFM images of the InAs QDs grown at 500°C on the ten-period SL template with a growth temperature of 530°C of the (In,Ga)As and thin GaAs cap layers, and 610°C for annealing and the GaAs spacer layers. InAs QDs (a) without and (b) with an additional InAs QD layer, thin GaAs capped and annealed, on the SL template with a 15-nm upper GaAs separation layer and overgrown by 15-nm GaAs. The scan field is $1 \times 1 \mu\text{m}^2$ and the black-to-white height contrast is 20 nm for both images.

Single InAs QDs on the SL template nodes are realized at the elevated growth and annealing temperatures when an additional 0.5-nm InAs QD layer is inserted on the SL template with a 15-nm upper GaAs separation layer and overgrown by 15-nm GaAs. The InAs QD layer is thin GaAs capped and annealed, as the (In,Ga)As layers in the SL template. Due to the fact that the InAs QDs in this interlayer solely form on the SL nodes, the lateral strain field modulation most effectively concentrates after thin GaAs capping and annealing. Together with the enhanced In adatom migration length at elevated temperatures, supported by the tendency for QD coalescence [discussed for the case of Fig. 2(a)], the resulting shrinkage of the effective area of the tensile strain field minima for preferred QD formation produces single InAs QDs in the center of the nodes, as shown in Fig. 7(b). Only the combination of elevated temperatures for SL template formation and InAs QD growth and the insertion of an additional InAs QD layer realizes single QDs. Applied separately, the average QD number per group is not changed significantly.

IX. OPTICAL PROPERTIES OF QD MOLECULES

The high structural and optical quality of the InAs QD molecules manifests itself in the excellent PL properties up to RT. The FWHM of the capped InAs QD molecules of Fig. 5(b) at RT of 60 meV is the same as that at low temperature and the PL peak intensity drops by a factor of only 1500 due to thermally activated escape of carriers from the QD molecules.⁷

Detailed temperature-dependent PL measurements between 5 K and RT reveal a constant value of the FWHM of the QD molecules up to 80 K, as shown in Fig. 8(a). The FWHM then undergoes a distinct minimum at 140 K due to thermally activated redistribution of carriers preferentially from smaller (higher energy) to larger (lower energy) QDs.¹³ This is accompanied by a characteristic enhancement of the low-energy shift of the PL peak position as a function of temperature, depicted in Fig. 8(b). The FWHM increases steeply to 57 meV at 190 K due to equilibration of the carrier distribution in large and small QDs when the probability of carrier escape from the large QDs increases. Remarkably, the steep increase of the FWHM is followed by a very weak increase above 190 K to a value of 59 meV at RT. The distinct inflection point at 190 K indicates equilibration of the carrier redistribution solely within the QD molecules, which is followed by thermal broadening resembling that of isolated QDs,¹⁴ hence, indicating that the QD molecules are electronically isolated. Such an unusual behavior of the FWHM is not observed for the capped (In,Ga)As QD layer of Fig. 1(c) and for the SL template of Fig. 1(c) without an upper (In,Ga)As layer. After the minimum of the FWHM and enhanced low-energy shift of the PL peak position due to thermally activated carrier redistribution, the FWHM continuously increases with temperature, which is attributed to a continuous increase of the carrier spread over areas fully covered with QDs.

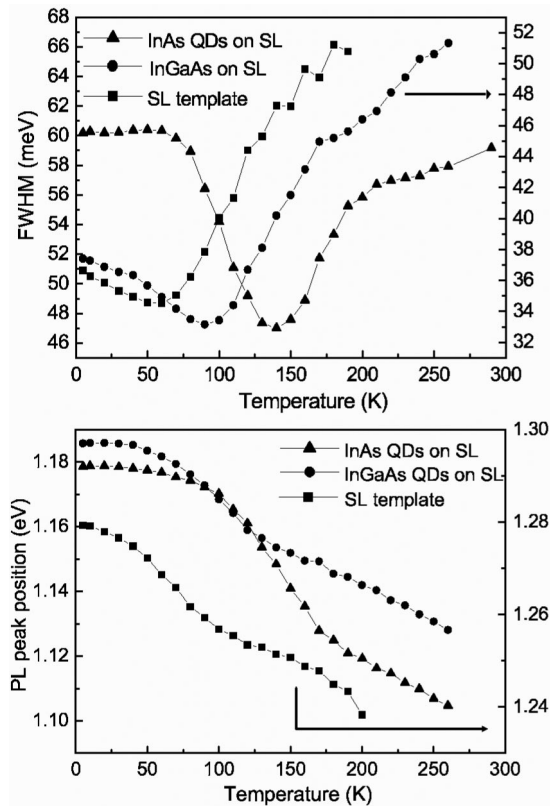


FIG. 8. Temperature dependence of (a) the full width at half maximum (FWHM) and (b) the PL peak position of the capped InAs QD molecules of Fig. 5(b) (solid triangles), the capped (In,Ga)As QDs of Fig. 1(c) (solid circles), and the SL template of Fig. 1(c) without upper (In,Ga)As layer (solid squares).

X. CONCLUSIONS

In conclusion, we have studied the formation of ordered lateral InAs quantum dot (QD) molecules by self-organized anisotropic strain engineering of an (In,Ga)As/GaAs superlattice (SL) template on GaAs (311)B substrates by molecular-beam epitaxy (MBE). During stacking the SL template self-organizes into a highly ordered, two-dimensional (In,Ga)As and, thus, strain field modulation on a mesoscopic length scale due to anisotropic surface migration and strain-correlated growth, constituting a Turing pattern in solid state. InAs QDs form a lattice of ordered groups on the SL template nodes due to local strain recognition. The SL template and QD growth conditions, such as the number of SL peri-

ods, growth temperatures, amount and composition of deposited (In,Ga)As, and insertion of Al-containing layers, have been varied for optimized InAs QD ordering within and among the QD molecules, as evaluated by atomic force microscopy (AFM). The average number of QDs per molecule has been controlled by the upper GaAs separation layer thickness on the SL template and the growth temperature of the (In,Ga)As layers in the SL template. The strain-correlated growth in the SL template formation and QD ordering has been consistently confirmed by high-resolution x-ray diffraction (XRD) in various scattering geometries. Single InAs QDs on the SL template nodes have been realized at elevated temperatures for SL template formation and InAs QD growth together with the insertion of an additional InAs QD layer. The optical properties of the QD molecules are excellent up to room temperature. Temperature dependent photoluminescence (PL) measurements have revealed an unusual behavior of the full width at half maximum indicating carrier redistribution solely within the QD molecules. Hence, fascinating applications of our QD molecules for quantum functional devices are foreseen.

- ¹R. Nötzel, T. Mano, Q. Gong, and J. H. Wolter, see Special Issue on *Nanoelectronics and Nanoscale Processing* [Proc. IEEE**91**, 1898 (2003)].
- ²T. Ishikawa, T. Nishimura, S. Kohmoto, and K. Asakawa, *Appl. Phys. Lett.* **76**, 167 (2000).
- ³H. Lee, J. A. Johnson, M. Y. He, J. S. Speck, and P. M. Petroff, *Appl. Phys. Lett.* **78**, 105 (2001).
- ⁴Y. 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).
- ⁵T. Mano, R. Nötzel, G. J. Hamhuis, T. J. Eijkemans, and J. H. Wolter, *Appl. Phys. Lett.* **81**, 1705 (2002).
- ⁶T. Mano, R. Nötzel, G. J. Hamhuis, T. J. Eijkemans, and J. H. Wolter, *J. Appl. Phys.* **95**, 109 (2004).
- ⁷T. van Lippen, R. Nötzel, G. J. Hamhuis, and J. H. Wolter, *Appl. Phys. Lett.* **85**, 118 (2004).
- ⁸Q. Gong, R. Nötzel, G. J. Hamhuis, T. J. Eijkemans, and J. H. Wolter, *Appl. Phys. Lett.* **81**, 3254 (2002).
- ⁹J. Temmyo, R. Nötzel, and T. Tamamura, *Appl. Phys. Lett.* **71**, 1086 (1997).
- ¹⁰R. Nötzel, J. Temmyo, and T. Tamamura, *Nature (London)* **369**, 131 (1994).
- ¹¹T. Leppänen, M. Karttunen, R. A. Barrio, and K. Kaski, *Phys. Rev. E* **70**, 066202 (2004).
- ¹²T. Mano, R. Nötzel, G. J. Hamhuis, T. J. Eijkemans, and J. H. Wolter, *J. Appl. Phys.* **92**, 4043 (2002).
- ¹³D. I. Lubyshchev, P. P. Gonzalez-Borrero, E. Marega Jr., E. Petitprez, N. La Scala, Jr., and P. Basmaji, *Appl. Phys. Lett.* **68**, 205 (1996).
- ¹⁴I. Favero *et al.*, *Phys. Rev. B* **68**, 233301 (2003).