Self-organized template formation for quantum dot ordering

Richard Nötzel, a) Takaaki Mano, and Joachim H. Wolter
eiT/COBRA Inter-University Research Institute, Eindhoven University of Technology, 5600 MB Eindhoven, The Netherlands

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Ordered arrays of quantum dots (QDs) are created by self-organized anisotropic strain engineering of (In,Ga)As/GaAs quantum wire (QWR) superlattice (SL) templates on exactly oriented GaAs (100) substrates by molecular beam epitaxy (MBE). The well-defined one-dimensional arrays of (In,Ga)As QDs formed on top of these templates due to local strain recognition are of excellent structural and optical quality up to room temperature. The QD arrays thus allow for fundamental studies and device operation principles based on single- and multiple carrier- and photon-, and coherent quantum interference effects. © 2004 American Vacuum Society.

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I. INTRODUCTION

The realization of semiconductor quantum dot (QD) arrays and networks in well-defined lateral arrangements is essential for the development of future quantum functional devices.1 We have recently introduced a new concept for creating these kinds of ordered QD arrays by self-organized epitaxy. The concept is based on self-organized anisotropic strain engineering of an (In,Ga)As/GaAs quantum wire (QWR) superlattice (SL) template on exactly oriented GaAs (100) substrates.2 During molecular beam epitaxy (MBE) at elevated temperature elongated (In,Ga)As QDs develop into very uniform and long QWR arrays with a well-defined lateral periodicity.3 QWR formation relies on the anisotropic adatom surface migration and In desorption during annealing of the layers of elongated QDs after capping with a thin GaAs layer.4,5 The improvement of the uniformity of the QWRs and related uniformity and accumulation of the anisotropic strain field in SL growth provides a well-defined template for the ordering of single InAs (Ref. 2) and (In,Ga)As (Ref. 4) QDs formed on top due to local strain recognition in one-dimensional arrays as illustrated in Fig. 1.

Excellent structural perfection and optical properties are established for these ordered (In,Ga)As QD arrays by atomic force microscopy (AFM), high-resolution x-ray diffraction, and photoluminescence (PL) spectroscopy. Temperature-dependent PL measurements reveal efficient carrier transfer from the QWR template to the QDs and within the QD arrays. Hence, self-organized anisotropic strain engineering provides a unique route for the realization of well-ordered and functional QD arrays of high quality.

II. TEMPLATE FORMATION

The crucial steps for template formation and QD ordering in the growth by solid-source MBE on epi-ready, singular GaAs (100) substrates are schematically depicted in Figs. 2(a)–2(e). They comprise (after deposition of a 200 nm GaAs buffer layer at 580 °C at a growth rate of 0.054 nm/s and an As$_4$ beam equivalent pressure of 1.0 × 10$^{-5}$ Torr)

(a) Formation of randomly distributed, elongated (In,Ga)As (1.8–2.6 nm) QDs at elevated temperature (540 °C) with In composition between 36% and 41% in the Stranski–Krastanov (SK) growth mode [Fig. 1(a)].6

(b) Growth of the thin GaAs cap layer (0.7–0.9 nm at 540 °C).

(c) Annealing at higher temperature (2 min at 580 °C). The QDs elongate further and connect due to anisotropic adatom surface migration preferentially along [0–11] related to the (2 × 4) GaAs surface reconstruction. Simultaneous In desorption allows uniform QD connection due to strain reduction and is balanced by the thin GaAs cap layer of step (2).4,5

(d) Growth of the GaAs separation layer (to a total thickness of 13–16 nm at 580 °C). The lateral strain field modulation from the buried QWRs is preserved at the surface [Fig. 1(d)].

(e) Growth of the subsequent (In,Ga)As QD layer (at 540 °C). The QDs preferentially nucleate above the QWRs where the lateral strain field minima reduce the lattice mismatch and induce strain gradient-driven In adatom migration preferentially along [011] towards the minima.7 One-dimensional QD arrays form [Fig. 1(e)].

(f) 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.2

Figure 3 shows the corresponding AFM images taken in tapping mode in air for the 16th period of the In$_{0.41}$Ga$_{0.59}$As 2.3 nm/GaAs 13.0 nm QWR template: (a) In$_{0.41}$Ga$_{0.59}$As QDs; (b) GaAs cap; (c) annealing; and (d) GaAs separation layer. The In$_{0.41}$Ga$_{0.59}$As growth rate is 0.092 nm/s. One-dimensional QD arrays are observed in Fig. 3(a). The growth of the GaAs cap layer [Fig. 3(b)] induces only marginal changes in shape and size of the QDs. After annealing [Fig. 3(c)], the surface becomes rather flat, with QWR-like structures along the [0–11] direction. The root-mean-square (rms) roughness over a 500×500 nm$^2$ area is reduced from 2.2 nm [Fig. 3(b)] to 0.6 nm [Fig. 3(c)] and the rms roughness along a 500 nm line on top of the QWRs is only 0.2 nm, affirming...
that the QDs have uniformly connected. After growth of the GaAs separation layer, asymmetric mounds elongated along [0–1 1] are developed [Fig. 3(d)] due to the growth instability on singular GaAs (100). The much larger length of the QD arrays compared to that of the mounds and their lateral ordering [closing the sequence to Fig. 3(a)] confirm that the QD ordering is not caused by morphological features like step edges but originates from the uniform lateral strain field modulation on the QWR template.

The evolution of the (In,Ga)As/GaAs QWR template is further demonstrated by the distinct dependence of the ordering of InAs QDs grown on top on the number of SL periods. The locations of the InAs QDs mark the lateral strain field minima at the GaAs surface induced by the underlying (In,Ga)As QWR structure. Figures 4(a)–4(e) show the AFM images of the QDs formed by 2.1 ML InAs at 480 °C at a growth rate of 0.037 nm/s (a) directly on GaAs (100) and (b)–(e) on the In0.36Ga0.64As 2.6 nm/GaAs 16 nm QWR template with the number of SL periods of (b) 1; (c) 5; (d) 10; and (e) 15. The InAs QDs are arranged randomly on the GaAs buffer layer [Fig. 4(a)]. For one SL period [Fig. 4(b)], QD ordering is hardly observed, indicating incomplete QWR formation. A weak modulation of the QD density along [011] appears when the number of SL periods is increased to 5 [Fig. 4(c)]. For 10 and 15 SL periods [Figs. 4(d) and 4(e)], a clear ordering in arrays of multiple QDs along [0–1 1] takes place. The length of the arrays easily exceeds 3 μm with a lateral periodicity of 140 nm, which agrees with the QWR periodicity determined from x-ray diffraction. The ordering of the InAs QDs and, hence, the uniformity of the QWR template, improves with increasing number of SL periods, generating a well-defined lateral strain field modulation at the GaAs surface with sufficiently deep minima. Single InAs QDs are formed on this QWR template for reduced InAs thickness of 1.5 ML and a very low growth rate of 0.0007 nm/s.

Although fairly long and uniform QWRs are clearly formed, a significant number of bends and branches is still evident. The bends and branches are attributed to excess...
strain accumulated in the QWR template which cannot be relaxed along straight QWRs. Excess strain is accumulated by supply of \( \text{In}, \text{Ga} \)As too far above the critical thickness and too-thin GaAs separation layers between successive \( \text{In}, \text{Ga} \)As layers. Moreover, too much \( \text{In}, \text{Ga} \)As results in the nucleation of QDs not only at the most preferable sites on top of the center of the QWRs but also aside, generating regions of multiple QD arrays and 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 \( \text{In}, \text{Ga} \)As layer due to vertical strain mediation. Hence, the uniformity of the QWR template will improve for reduced supply of \( \text{In}, \text{Ga} \)As to a minimum above the critical thickness for island formation and increased GaAs separation layer thickness, while preserving sufficient vertical strain correlation between successive \( \text{In}, \text{Ga} \)As layers for ordering. This improvement of the QWR template is demonstrated for a series of samples comprising 2.3 or 1.8 nm \( \text{In}_{0.41} \text{Ga}_{0.59} \)As separated by 13 or 16 nm thick GaAs layers in each of the 15 SL periods.

Figure 5 shows the high-resolution x-ray diffraction spectra in the vicinity of the asymmetric (311) glancing exit reflection (a) of the 15-period \( \text{In}_{0.41} \text{Ga}_{0.59} \)As 2.3 nm/GaAs 13 nm QWR template A; (b) of the 15-period \( \text{In}_{0.41} \text{Ga}_{0.59} \)As 1.8 nm/GaAs 13 nm QWR template B; and (c) of the 15-period \( \text{In}_{0.41} \text{Ga}_{0.59} \)As 1.8 nm/GaAs 16 nm QWR template C. The black (gray) spectra are measured with the x-ray beam parallel to the [011] ([0-11]) direction. With the x-ray beam parallel to the [011] direction, clear satellite peaks are observed for all samples due to the lateral periodicity of the stacked QWRs. As the strain accumulation is reduced from template A to C, the satellite peaks sharpen and their peak-to-valley ratio increases, indicating the improved uniformity of the QWRs. This is supported by the narrower zeroth-order peaks with the x-ray beam parallel to \( [0-11] \) for samples B and C. Additional confirmation of the improved uniformity of the QWR templates A to C is gained from the low-temperature PL spectra shown in Figs. 6(a)–6(c). The PL peak width decreases from 81 meV for QWR template A to 42 meV for QWR template C.

III. QD ORDERING

The improved uniformity of the QWR templates A to C directly relates to that of the single \( \text{In}, \text{Ga} \)As QD arrays grown on top. The AFM images of the \( \text{In}, \text{Ga} \)As QD arrays formed at 540 °C on the 15-period \( \text{In}, \text{Ga} \)As QWR templates A–C are depicted in Figs. 7(a)–7(c). The QDs are formed by 2.3 nm \( \text{In}_{0.41} \text{Ga}_{0.59} \)As on template A and 1.8 nm \( \text{In}_{0.41} \text{Ga}_{0.59} \)As on templates B and C. Though improved compared to template A [Fig. 7(a)], after reduction of the \( \text{In}, \text{Ga} \)As supply for template B [Fig. 7(b)], a relatively large number of branches is still visible with an area density of 2.2 \( \mu \text{m}^{-2} \). For the additional increase of the GaAs separation...
layer thickness in template C [Fig. 7(c)], the area density of the branches is significantly reduced to 1.4 \( \mu \text{m}^2 \). The single QD arrays are perfectly straight over more than 1 mm and most of the QD arrays are extended over 10 \( \mu \text{m} \) length.

The high structural quality of the 1.8-nm-In\(_{0.41}\)Ga\(_{0.59}\)As single QD arrays on the QWR template C is highlighted by its optical properties. For the PL measurements the QD arrays are capped with 100 nm GaAs (20 nm at 540 °C plus 80 nm at 580 °C after 10 s growth interruption). The PL is excited by the 532 nm line of a Nd–YAG laser with an excitation power density of 0.2 W/cm\(^2\), dispersed by a single monochromator and detected by a cooled InGaAs charge-coupled device. Figure 8 shows the temperature-dependent PL spectra and Figs. 9(a) and 9(b) the corresponding PL intensity and peak width. At low temperature, the PL emission from the QD arrays, centered at 1.21 eV, exhibits a peak width of 72 meV. The PL line at 1.37 eV stems from the QWR template. The high-energy shift is due to the In desorption during template formation which is independently confirmed by x-ray diffraction.\(^4,5\) When the PL of the QWR template vanishes around 100 K, the PL intensity of the QD arrays slightly increases [Fig. 9(a)], indicating thermally activated carrier transfer from the QWRs to the QDs. Carrier redistribution within the QD arrays is revealed from the PL intensity and peak width.

**Fig. 6.** Low temperature PL spectra (a) of the 15-period In\(_{0.41}\)Ga\(_{0.59}\)As 2.3 nm/GaAs 13 nm QWR template A; (b) of the 15-period In\(_{0.41}\)Ga\(_{0.59}\)As 1.8 nm/GaAs 13 nm QWR template B; and (c) of the 15-period In\(_{0.41}\)Ga\(_{0.59}\)As 1.8 nm/GaAs 16 nm QWR template C.

**Fig. 7.** AFM images (a) of the 2.3 nm In\(_{0.41}\)Ga\(_{0.59}\)As QD arrays on the 15-period In\(_{0.41}\)Ga\(_{0.59}\)As 2.3 nm/GaAs 13 nm QWR template A; (b) of the 1.8 nm In\(_{0.41}\)Ga\(_{0.59}\)As QD arrays on the 15-period In\(_{0.41}\)Ga\(_{0.59}\)As 1.8 nm/GaAs 13 nm QWR template B; and (c) of the 1.8 nm In\(_{0.41}\)Ga\(_{0.59}\)As QD arrays on the 15-period In\(_{0.41}\)Ga\(_{0.59}\)As 1.8 nm/GaAs 16 nm QWR template C. The scan field is \( 1 \times 1 \mu \text{m}^2 \) and the black-to-white height contrast is 30 nm in all images.

**Fig. 8.** Temperature-dependent PL spectra of the capped 1.8 nm In\(_{0.41}\)Ga\(_{0.59}\)As single QD arrays on the 15-period In\(_{0.41}\)Ga\(_{0.59}\)As 1.8 nm/GaAs 16 nm QWR template C.

**Fig. 9.** (a) PL peak intensity and (b) PL peak width of the capped 1.8 nm In\(_{0.41}\)Ga\(_{0.59}\)As single QD arrays on the 15-period In\(_{0.41}\)Ga\(_{0.59}\)As 1.8 nm/GaAs 16 nm QWR template C as a function of temperature.
peak width, which exhibits a minimum around 200 K [Fig. 9(b)]. This minimum reflects preferential carrier occupation of larger QDs at intermediate temperatures followed by equilibration of the carrier distribution at higher temperatures.\textsuperscript{10} The PL efficiency of the QD arrays is strong up to room temperature. Most notably, the PL peak width of the QD arrays at room temperature of 70 meV does not exceed that at low temperature (72 meV) as is expected for QDs with strong carrier confinement. This high structural and optical quality of the QD arrays on the QWR template is attributed to the smoothness of the strain field modulation on the dot-diameter and dot-to-dot distance length scales which does not introduce any defects or irregularities in the QD arrays, often occurring when artificial patterning techniques are applied.\textsuperscript{11–17} This is the key advantage of our method for QD ordering based on self-organized anisotropic strain engineering.

IV. CONCLUSION

Well-ordered quantum dot (QD) arrays have been created by self-organized anisotropic strain engineering of an (In,Ga)As/GaAs quantum wire (QWR) superlattice (SL) template. During molecular beam epitaxy (MBE) of a strained (In,Ga)As/GaAs SL on exactly oriented GaAs (100) substrates, elongated (In,Ga)As QDs evolve into very uniform and long QWR arrays with a well-defined lateral periodicity. The related anisotropic strain field provides a unique template for the ordering of (In,Ga)As QDs on top in one-dimensional arrays. The high uniformity of the QWR template is achieved by reducing the amount of (In,Ga)As and increasing the GaAs separation layer thickness in SL growth. The QD arrays formed on these templates are well separated, perfectly straight over more than 1 μm, and extended to over 10 μm length with a very small number of branches. The QD arrays exhibit strong PL emission up to room temperature with the PL peak width not exceeding that at low temperature. Temperature-dependent PL spectroscopy reveals efficient carrier transfer from the QWR template to the QDs and within the QD arrays. Self-organized anisotropic strain engineering is thus established for the creation of well-defined QD arrays and networks of high structural and optical quality, being excellent candidates for the realization of quantum functional devices for quantum communication and computing applications in solid state.