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Self-organized anisotropic strain engineering for quantum dot ordering

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Well-defined quantum dot arrays are formed by self-organized anisotropic strain engineering on GaAs (100) and (311)B substrates. Superior structural and optical properties are established for these ordered quantum dot arrays. The quantum dot arrays are, thus, excellent candidates for the realization of novel quantum functional devices based on single and multiple carrier- and photon-, and coherent quantum interference effects.
Key words: quantum dot array, self-organized epitaxy, strain engineering

1. INTRODUCTION

Lateral ordering of semiconductor quantum dots (QDs) in well-defined arrangements is one of the most challenging prerequisites to be fulfilled for the realization of future quantum functional devices. We have established a new concept for creating ordered arrays of QDs by self-organized epitaxy. The concept is based on self-organized anisotropic strain engineering of strained layer templates and is demonstrated for (In,Ga)As/GaAs superlattice structures on GaAs (100) substrates and strain induced (In,Ga)As growth instability on high-index GaAs (311)B substrates [1]. The well-defined one- and two-dimensional arrays and networks of InAs QDs formed on top of these templates due to local strain recognition are of excellent structural perfection and optical quality up to room temperature.

Temperature dependent photoluminescence (PL) measurements reveal efficient carrier transfer from the templates - which themselves constitute distinct one- and zero-dimensional quantum nanostructure arrays - to the QDs and within the QD arrays. It are these types of complex self-organized semiconductor nanostructure networks which are regarded to provide functional building blocks for full control of the optoelectronic materials properties at the single charge and photon level required for future quantum communication and computing applications in solid state [1].

2. GAAS (100): TEMPLATE FORMATION

On GaAs (100) substrates, during molecular beam epitaxy (MBE) of a strained (In,Ga)As/GaAs superlattice (SL), elongated (In,Ga)As QDs deposited at elevated temperature (540 °C) develop into very uniform and long quantum wire (QWR) arrays with a well-defined lateral periodicity. QWR formation relies on the anisotropic adatom surface migration and In desorption during annealing at 580 °C of the layers of elongated QDs after capping with a thin (0.7-0.9 nm) GaAs layer. By repetition in (In,Ga)As/GaAs (13-16 nm at 580 °C) SL growth the accumulation and improvement of the uniformity of the generated anisotropic strain field provides a well-defined template for the ordering of (In,Ga)As QDs grown on top of the last GaAs layer in one-dimensional arrays [2,3]. The one-dimensional ordering of the QDs originates from the preferential QD nucleation at locations of local strain minima at the GaAs surface, which are induced by the (In,Ga)As QWR structure below. There, the lattice mismatch between (In,Ga)As and GaAs is reduced, making QD nucleation energetically more favorable, and the lateral strain gradient causes directional In surface migration and In accumulation in these minima.

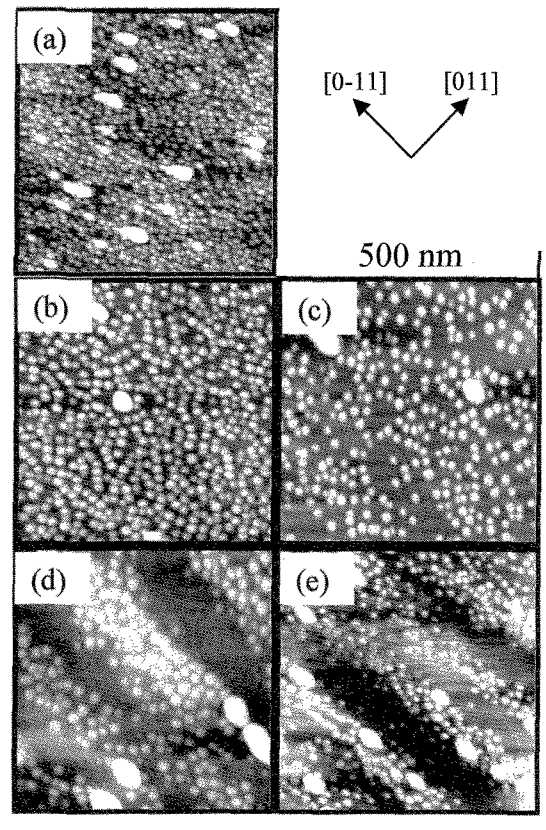

Fig. 1. AFM images of the 2.1-ML-InAs QDs (a) on GaAs (100) and (b) on the 1, (c) 5, (d) 10, and (e) 15-periods $In_{0.36}Ga_{0.64}As$ 2.6 nm/GaAs 16 nm QWR templates (InAs growth temperature 480 °C, growth rate 0.037 nm/s). The scan field is 500 nm × 500 nm and the black-to-white height contrast is 7 nm in all images.

The template effect is demonstrated by the distinct dependence of the InAs QD ordering on the number of SL periods. Figures 1 (a)-(e) show the atomic force microscopy (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 $In_{0.36}Ga_{0.64}As$ 2.6 nm/GaAs 16 nm QWR template with the number of SL periods of (b) 1, (c) 5, (d) 10, and (e) 15. While on the GaAs buffer layer the InAs QDs are arranged randomly, a distinct tendency towards QD ordering occurs on the QWR templates when the number of SL periods is increased from 1 to 15. For one period (Fig. 1 (b)), QD ordering is hardly observed, indicating a large disorder or incomplete QWR formation of the template. When the number of SL periods is increased to 5, a weak modulation of the QD density along [011] appears (Fig 1 (c)). For 10 and 15 SL periods (Figs. 1 (d) and (e)), a clear ordering in arrays of multiple QDs along [0-11] takes place. The length of the arrays easily exceeds 3 μm with a lateral periodicity of 140 nm. This periodicity is consistent with that of the (In,Ga)As QWRs determined from X-ray diffraction [3]. Thus, the ordering of the InAs QDs develops with the improved uniformity of the QWR template and the related uniformity and accumulation of the lateral strain field modulation at the GaAs surface with sufficiently deep minima upon increasing the number of SL periods.

3. GAAS (100): ONE-DIMENSIONAL QD ARRAYS

The formation of one-dimensional single QD arrays is achieved by reducing the InAs coverage and enhancing the In adatom migration length at low growth rate for InAs QDs and/or by the growth of larger (In,Ga)As QDs at higher temperature. The nucleation of the QDs then occurs solely at the most favored locations above the centers of the QWRs. Figure 2 shows the AFM images of the QDs formed (a) by 1.5 ML InAs at 480 °C at a growth rate of 0.0007 nm/s and (b) by 2.6 nm $In_{0.36}Ga_{0.64}As$ at 540 °C at a growth rate of 0.104 nm/s on the 15-periods $In_{0.36}Ga_{0.64}As$ 2.6 nm/GaAs 16 nm QWR template. The formation of single InAs and (In,Ga)As QD arrays with average lateral spacing of again ~140 nm and QD distance of only a few nanometers is evident.

Although single QD arrays are clearly formed on the QWR template, a significant number of bends and branches, in particular for the longer (In,Ga)As QD arrays in Fig. 2 (b) is still visible. The bends and branches are attributed to excess strain accumulation in the QWR template. Since elastic strain relaxation cannot occur along straight QWRs, the excess strain field will result in fluctuations in width and height (and ultimately breaking up of the QWRs into QDs) and/or bends and branches to release the strain. The excess strain accumulation is mainly caused by supply of (In,Ga)As too 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. Hence, formation of straight and uniform single 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.

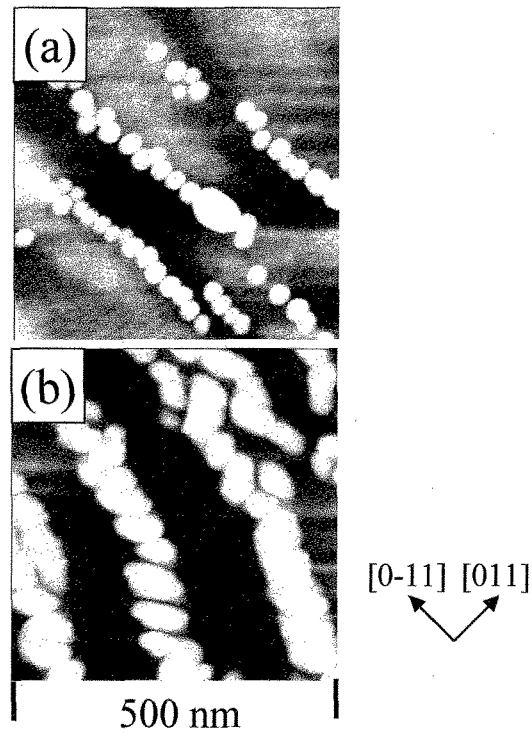

Fig. 2. AFM images of (a) the 1.5-ML-InAs single QD arrays (growth temperature 480 °C, growth rate 0.0007 nm/s) and (b) the 2.6-nm-$In_{0.36}Ga_{0.64}As$ single QD arrays (540 °C, 0.104 nm/s) on the 15-periods $In_{0.36}Ga_{0.64}As$ 2.6 nm/GaAs 16 nm QWR templates. The scan field is 500 nm × 500 nm in both images and the black-to-white height contrast is (a) 10 nm and (b) 15 nm.

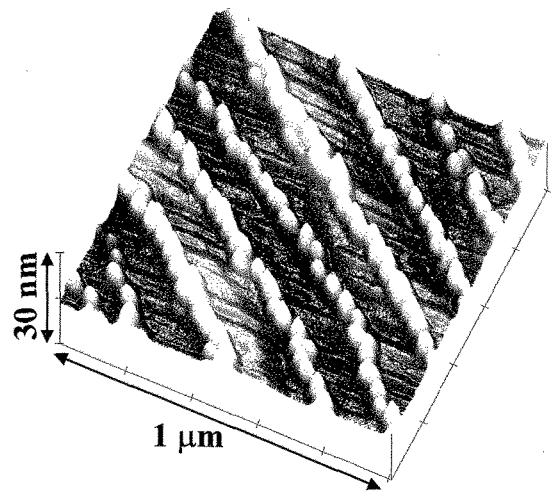

Fig. 3. Three-dimensional AFM image of the 1.8-nm-$In_{0.41}Ga_{0.59}As$ single QD arrays (growth temperature 540 °C, growth rate 0.092 nm/s) on the 15-periods $In_{0.41}Ga_{0.59}As$ 1.8 nm/GaAs 16 nm QWR template. The scan field is 1 μm × 1 μm.

This improvement of the QD arrays is shown for a different set of samples comprising QWR templates with 2.3 or 1.8 nm $In_{0.41}Ga_{0.59}As$ separated by 13 or 16 nm thick GaAs layers in each of the 15 SL periods [4]. The AFM image of the 1.8-nm-$In_{0.41}Ga_{0.59}As$ QD layer on top of the $In_{0.41}Ga_{0.59}As$ 1.8 nm/GaAs 16 nm QWR template is shown in Fig. 3. The one-dimensional single (In,Ga)As QD arrays are now straight over more than 1 μm and most of the QD arrays are extended over 10 μm length with a very small number of branches.

4. GAAS (100): OPTICAL PROPERTIES

To assess the optical properties, the QD arrays are capped with 100 nm GaAs (20 nm at 540 °C plus 80 nm at 580 °C). The temperature dependent PL spectra of the optimized 1.8-nm-$In_{0.41}Ga_{0.59}As$ QD arrays on the 15-periods $In_{0.41}Ga_{0.59}As$ 1.8 nm/GaAs 16 nm QWR template are shown in Fig. 4. The corresponding PL intensity and peak width are plotted in Fig 5 (a) and (b). At low temperature, 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 high-energy shift is due to the In desorption during template formation [5]. 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 the QWRs to the QDs. The PL of 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 among the QDs. 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. The high structural and optical quality of the QD arrays on the template is attributed to the smoothness of the strain field modulation on the dot-diameter and dot-to-dot distance length scales. This is the key advantage of our method for QD ordering based on self-organized anisotropic strain engineering.

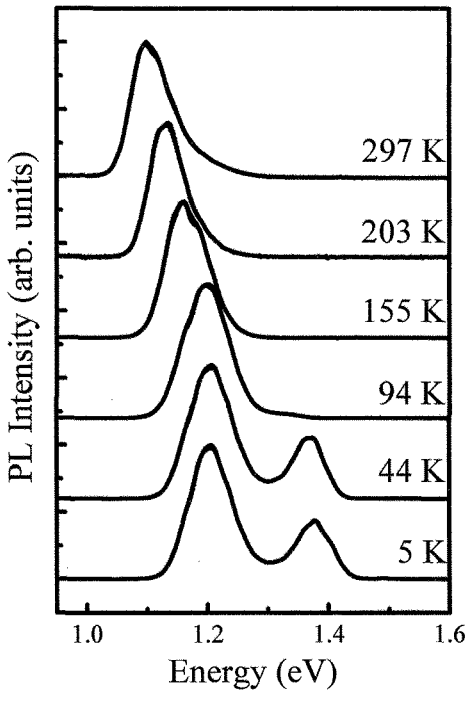

Fig. 4. Temperature dependent PL spectra of the capped 1.8-nm-$In_{0.41}Ga_{0.59}As$ single QD arrays on the 15-periods $In_{0.41}Ga_{0.59}As$ 1.8 nm/GaAs 16 nm QWR template.

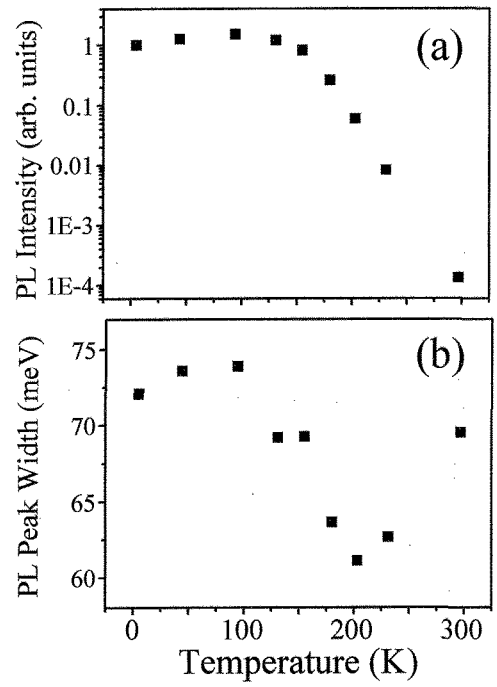

Fig. 5. (a) PL peak intensity and (b) PL peak width of the capped 1.8-nm-$In_{0.41}Ga_{0.59}As$ single QD arrays on the 15-periods $In_{0.41}Ga_{0.59}As$ 1.8 nm/GaAs 16 nm QWR template as a function of temperature.

5. GAAS (311)B: TEMPLATE FORMATION

On high-index GaAs (311)B substrates self-organized strain engineering for QD ordering is accomplished by strain induced growth instability of thin (In,Ga)As layers. The resulting two-dimensional strain field modulation generates a unique template for full control of the nucleation and growth of InAs QDs in a connected network [6,7]. Fig. 6 shows the AFM images of the $In_{0.35}Ga_{0.65}As$ layers on GaAs (311)B with thickness between 1.3 and 2.1 nm grown at 500 °C at a rate of 0.37 μm/h. While the surface of the 1.3-nm-$In_{0.35}Ga_{0.65}As$ layer is flat, without any pronounced features (Fig. 6 (a)), the 1.5 nm thick layer (Fig. 6 (b)) develops a matrix of cells with an average height modulation of 1 nm. Upon further growth the height of the cells increases gradually from 1.5 nm in Fig. 6 (c) (thickness 1.7 nm) to 2.5 nm in Fig. 6 (d) (thickness 2.1 nm). The area density of the cells remains unchanged. The gradual height increase of the surface modulation with constant lateral periodicity clearly identifies strain induced growth instability in contrast to QD formation in the Stranski-Krastanov (SK) growth mode where the height increases and saturates

abruptly at the critical layer thickness and further growth mainly increases the QD density.

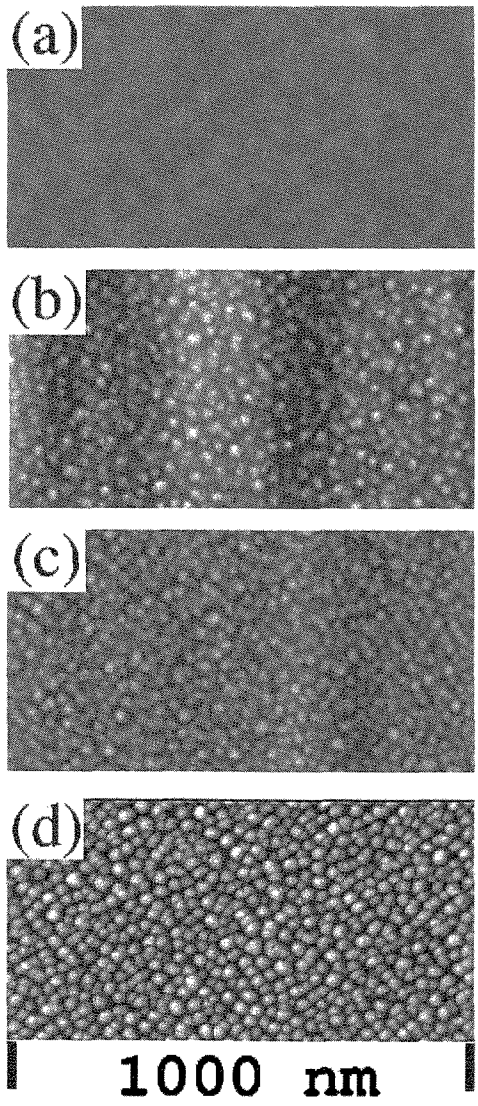

Fig. 6. AFM images of (a) the 1.3 nm, (b) 1.5 nm, (c) 1.7 nm, and (d) 2.1 nm thick $In_{0.35}Ga_{0.65}As$ layers on GaAs (311)B (growth temperature 500 °C, growth rate 0.37 μm/h). The scan field is 1000 nm x 500 nm and the black-to-white height contrast is 5 nm in all images.

6. GAAS (311)B: TWO-DIMENSIONAL QD ARRAY

The formation of two-dimensional InAs QD arrays on the (In,Ga)As template is shown in the AFM image in Fig. 7 (a). The QDs are formed by 0.23-nm-InAs at 500 °C at the low growth rate of 0.0028 μm/h on the 1.4 nm thick $In_{0.35}Ga_{0.65}As$ layer. The InAs QDs (three of them are marked by arrows in the lower left corner for clarity) are located exclusively on top, in the center of the cells of the template. This evidences full control of the QD nucleation in the SK mode, which occurs on GaAs (311)B for InAs due to the large lattice mismatch. Only a portion of the cells is occupied. The QD density can easily be enlarged from 2.4 x 10^{10} cm^{-2} to almost full cell coverage of 6 x 10^{10} cm^{-2} when the InAs layer thickness is increased to 0.38 nm. For comparison, QDs formed directly on GaAs by 0.46-nm-InAs at the same conditions (Fig. 7 (b)) exhibit a density of only 6 x 10^{9} cm^{-2} with random site distribution and large size fluctuations.

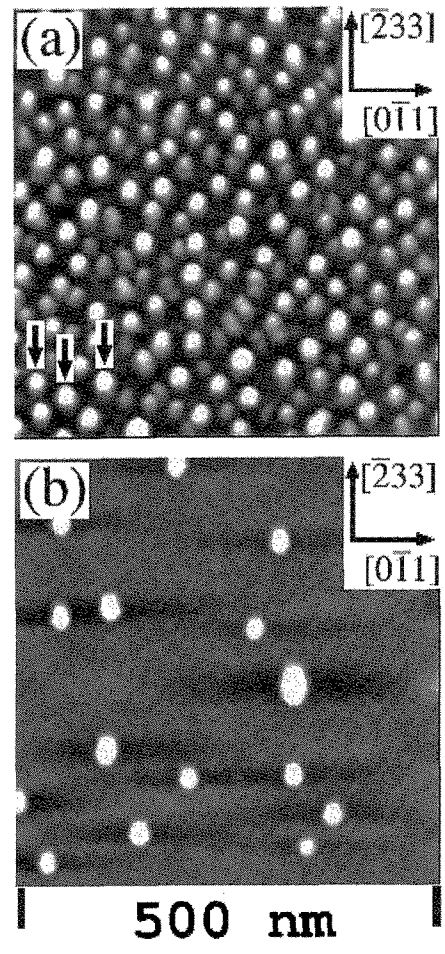

Fig. 7. (a) AFM image of the 0.23-nm-InAs QDs on the 1.4-nm-$In_{0.35}Ga_{0.65}As$ template, and (b) of the 0.46-nm-InAs QDs on GaAs (InAs growth temperature 500 °C, growth rate 0.0028 μm/h). The scan field is 500 nm x 500 nm and the black-to-white height contrast is 5 nm in both images.

The nucleation control of the InAs QDs on the template is assigned to the lateral strain field modulation at the surface of the matrix of cells. The partial strain relief in the center of each cell generates the local strain field minima where QD nucleation occurs, together with the directed, strain-gradient driven In surface migration. Moreover, the corresponding strain maxima at the borders of each cell provide barriers for In migration, limiting the diameter of the collection area below the In adatom diffusion length. This relates the QD size uniformity to that of the cell area. Hence, the template governs the nucleation site and collection area plus the directed adatom migration, thereby suppressing the random nature of the nucleation process and, thus, enabling the growth of uniform QDs of high density.

7. GAAS (311)B: OPTICAL PROPERTIES

The excellent structural properties are underlined by the PL measurements of the capped InAs QDs shown in Fig. 8. The InAs QDs formed on the 1.4-nm-$In_{0.35}Ga_{0.65}As$ template exhibit one strong PL line (solid line) with 943 nm peak wavelength and 32 meV line width. On the contrary, the PL spectrum of the InAs QDs directly grown on GaAs (dashed line), reveals a line at 869 nm which is assigned to the wetting layer and a rather broad line from the InAs QDs centered at 916 nm with 42 meV line width. Moreover, the PL peak intensity of the QDs on the template is more than 20 times higher than that of the InAs QDs on GaAs and no emission is detected from the (In,Ga)As template at 880 nm, revealing efficient carrier transfer from the template to the QDs. Considering the InAs QD area density, this suggests strong electronic coupling between the closely packed cells of the template. The structure realized here thus demonstrates self-organization of a two-dimensionally connected nanoscale network of isolated QDs with, moreover, controllable probability of the node occupation.

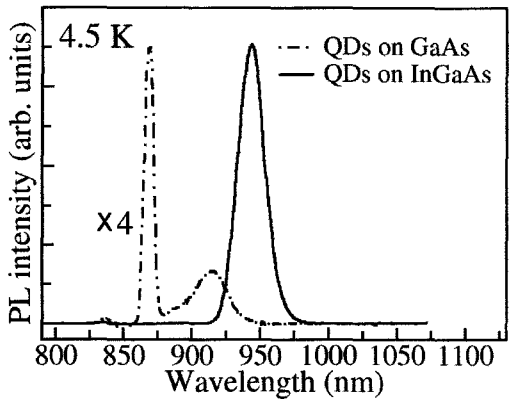

Fig. 8. PL spectrum (solid line) of the 0.23-nm-InAs QD network on the 1.4-nm-$In_{0.35}Ga_{0.65}As$ template taken at 4.5 K. The dashed line shows the PL spectrum of the 0.46-nm-InAs QDs on GaAs.

8. CONCLUSION

Well-defined quantum dot (QD) arrays have been created by self-organized anisotropic strain engineering of (In,Ga)As/GaAs templates. During molecular beam epitaxy (MBE) of a strained (In,Ga)As/GaAs superlattice (SL) on GaAs (100), 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 uniform template for InAs QD ordering on top in one-dimensional arrays. On high-index GaAs (311)B substrates, strain induced growth instability of (In,Ga)As generates a matrix of closely packed cells. The established strain distribution constitutes a unique template for the full control of InAs QD nucleation in a two-dimensionally connected network. The ordered InAs QD arrays exhibit superior structural perfection and optical properties up to room temperature with efficient carrier transfer from the template to the QDs and within the QD arrays. These well-defined and functional QD arrays and networks are, thus, excellent candidates for the realization of novel quantum functional devices for future quantum communication and computing applications.

9. REFERENCES

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