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Wavelength controlled multilayer-stacked linear InAs quantum dot arrays on InGaAsP/InP (100) by self-organized anisotropic strain engineering: A self-ordered quantum dot crystal

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Multilayer-stacked linear InAs quantum dot (QD) arrays are created on InAs/InGaAsP superlattice templates formed by self-organized anisotropic strain engineering on InP (100) substrates in chemical beam epitaxy. Stacking of the QD arrays with identical emission wavelength in the 1.55 μm region at room temperature is achieved through the insertion of ultrathin GaAs interlayers beneath the QDs with increasing interlayer thickness in successive layers. The increment in the GaAs interlayer thickness compensates the QD size/wavelength increase during strain correlated stacking. This is the demonstration of a three-dimensionally self-ordered QD crystal with fully controlled structural and optical properties. © *2008 American Institute of Physics*.
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Future quantum functional devices with well-designed control of the quantum mechanical and electromagnetic interactions of single and multiple electrons and photons require the lateral ordering of semiconductor quantum dots (QDs),[1] ultimately together with vertical ordering toward three-dimensional QD crystals. Lateral ordering of QDs formed in the Stranski–Krastonov growth mode, otherwise producing randomly arranged QDs, has been achieved by artificial substrate patterning[2–4] and combined with vertical strain correlated stacking.[5,6] Artificial patterning techniques, however, often introduce defects and irregularities in the QDs given by the spatial resolution of the lithography and etching steps which degrade the electronic and optical qualities. Recently we have introduced a concept for the lateral ordering of InGaAs QDs in linear arrays on planar GaAs (100) substrates by molecular beam epitaxy[7,8] and planar InP (100) substrates by chemical beam epitaxy (CBE),[9] on which we concentrate here. Based on anisotropic adatom surface migration and lateral and vertical strain correlations, wirelike InAs nanostructures are created during growth of an InAs/InGaAsP superlattice (SL), which acts as a template for the formation of linear InAs QD arrays due to local strain field recognition. This self-organization process has been proven to produce ordered QD arrays with excellent optical properties up to room temperature (RT) and whose emission wavelength was tuned into the technologically important 1.55 μm telecommunication wavelength region through the insertion of ultrathin GaAs interlayers beneath the QDs.[9]

In this letter, we demonstrate the formation of multilayer-stacked linear InAs QD arrays on the InAs/InGaAsP SL template on InP (100) demonstrating a fully self-ordered three-dimensional QD crystal. Identical emission wavelength of the stacked QD arrays is achieved by increasing the thickness of the GaAs interlayer in successive layers. The increment in the GaAs interlayer thickness compensates the QD size/wavelength increase encountered in strain correlated stacking for relatively thin separation layers.[10–12] The stacked QD arrays exhibit strong photoluminescence (PL) emission in the 1.55 μm wavelength region up to RT.

The samples were grown by CBE on semi-insulating InP (100) substrates with 2° miscut toward (110). Pressure controlled trimethylindium, triethylgallium, AsH_3, and PH_3 were used as precursors. The AsH_3 and PH_3 gases were thermally decomposed in a high temperature injector at 900 °C. After growth of a 200 nm InP buffer layer and 100 nm lattice-matched InGaAsP with band gap of 1.25 μm (Q1.25), the SL template was grown consisting of 2.1 monolayers (ML) InAs, 10 s growth interruption under As flux, 0.3 nm Q1.25 cap layer, 2 min annealing, and 15.3 nm Q1.25 separation layer, and repeated for seven periods. On top of the SL template a 2.6 ML InAs QD layer was grown with 0.6 ML GaAs interlayer inserted beneath. The multilayer-stacked 2.6 ML InAs QD arrays were separated by 16 nm Q1.25. The increment in the GaAs interlayer thickness in successive 2.6 ML InAs QD layers was 0.12 or 0 ML for comparison. The growth temperature was 505 °C for all layers and the annealing temperature was 520 °C. The growth rate of InAs was 0.24 ML/s. The surface morphology of the uncapped samples was characterized by tapping mode atomic force microscopy (AFM) in air. For the PL studies, the samples were capped by 100 nm Q1.25 and 50 nm InP, and excited by a neodymium doped yttrium aluminum garnet laser (532 nm) with excitation power density of $256\ \mathrm{mW/cm^2}$. The PL was dispersed by a single monochromator and recorded by a cooled InSb detector.

Figures 1(a)–1(c) show the AFM images of the uncapped single-layer QD arrays (described in detail in Ref. 9), the uncapped twofold-stacked, and the uncapped threefold-stacked QD arrays with increment in the GaAs interlayer thickness. For comparison, the AFM image of the uncapped threefold-stacked QD arrays without increment in the GaAs interlayer thickness is shown in Fig. 1(d). The QD arrays are

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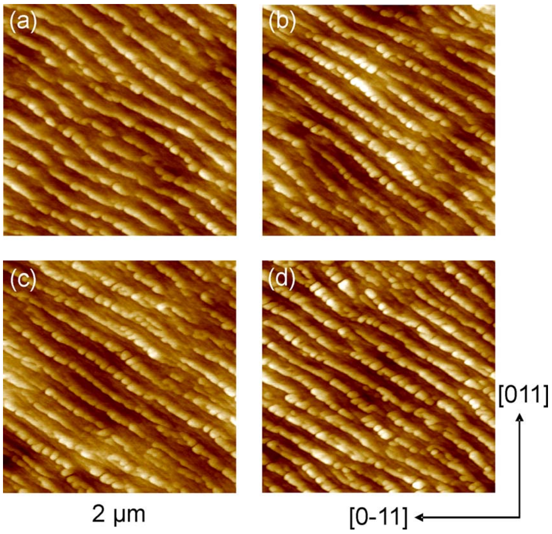

FIG. 1. (Color online) (a) AFM image of uncapped single-layer 2.6 ML InAs QD arrays on the seven period 2.1 ML InAs/0.3+15.3 nm InGaAsP SL template on InP (100) substrate with 0.6 ML GaAs interlayer beneath the QDs. [(b) and (c)] AFM images of uncapped twofold-stacked and threefold-stacked 2.6 ML InAs QD arrays with 0.12 ML increment in the GaAs interlayer thickness in successive layers. (d) AFM image of uncapped threefold-stacked 2.6 ML InAs QD arrays without increment in the GaAs interlayer thickness. The scan field is $2.0 \times 2.0\ \mu m^2$ and the height contrast is 10 nm.

aligned along the elastically soft [001] direction to minimize the strain energy,[13,14] which is selected by the miscut of the substrate with steps in the same direction.[9] InAs amount and growth rate, cap layer thickness, annealing temperature, and number of SL template periods are optimized for self-organized ordering due to anisotropic adatom surface migration during annealing and lateral and vertical strain correlations during stacking. Strain correlated stacking of the QD arrays separated by 16 nm Q1.25 manifests itself by the maintenance of linear ordering. With increment in the GaAs interlayer thickness, the height of the top twofold-stacked QD arrays (3.5 ± 0.5 nm height, with 0.72 ML GaAs interlayer) and that of the threefold-stacked QD arrays (3.2 ± 0.5 nm height, with 0.84 ML GaAs interlayer) are comparable to that of the single-layer QD arrays (3.3 ± 0.5 nm height, with 0.6 ML GaAs interlayer). The QD diameters remain almost unchanged with the increase in the number of stacked layers (single layer: 87.6 ± 8.0 nm, twofold stacked: 87.6 ± 10.1 nm, and threefold stacked: 87.1 ± 9.7 nm), as plotted in Fig. 2.

The role of the GaAs interlayer is to suppress As/P exchange encountered during the deposition of InAs on InGaAsP. This reduces the QD height as a function of interlayer thickness, and therefore the emission wavelength from far above 1.6 μm at RT into the 1.55 μm telecommunication wavelength region.[15] For the stacked QD arrays the increase in the GaAs interlayer thickness in successive layers compensates the well-known QD height and diameter increase,[10–12] to maintain the QD height and diameter throughout the entire structure. This is underlined by the pronounced increase in the QD height (4.2 ± 0.6 nm) and diameter (99.9 ± 10.2 nm) for the top threefold-stacked QD arrays without increment in the GaAs interlayer thickness [see Figs. 1(d) and 2].

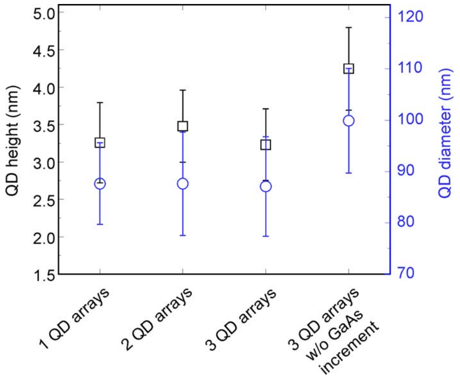

FIG. 2. (Color online) Average height (squares) and diameter (circles) of the QDs within the arrays as a function of the number of stacked layers.

The PL measurements taken at RT of the capped multilayer-stacked QD arrays shown in Fig. 3 corroborate the structural properties. The PL peak wavelength of the threefold-stacked QD arrays (1553 nm) is almost identical compared to that of the single-layer QD arrays (1551 nm). The PL efficiency of the threefold-stacked QD arrays increases by 44% compared to that of single-layer QD arrays indicating good crystal quality of the multilayer-stacked structure. Without increment in the GaAs interlayer thickness, the PL peak wavelength redshifts to 1582 nm due to the gradual increase in the QD size upon stacking (see inset in Fig. 3).

In summary, the formation of multilayer-stacked linear InAs QD arrays on InAs/InGaAsP SL templates formed by self-organized anisotropic strain engineering on InP (100) substrates in CBE has been achieved. Identical emission wavelength of the stacked QD arrays in the 1.55 μm telecommunication region at RT has been accomplished through the insertion of ultrathin GaAs interlayers beneath the QDs

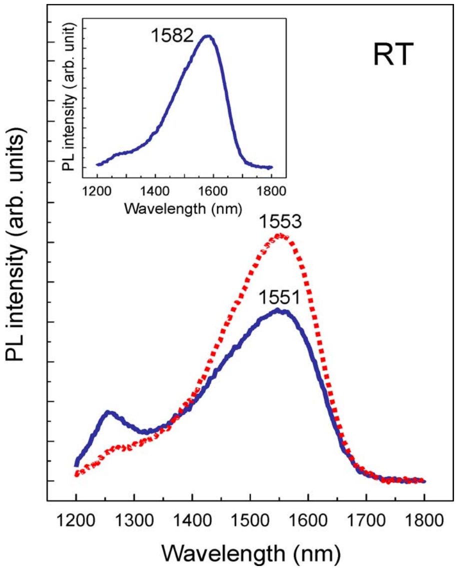

FIG. 3. (Color online) PL spectra taken at RT of capped single-layer (solid line) and threefold-stacked (dashed line) 2.6 ML InAs QD arrays with 0.12 ML increment in the GaAs interlayer thickness in successive layers. Inset: PL spectrum of capped threefold-stacked 2.6 ML InAs QD arrays without increment in the GaAs interlayer thickness.

with increasing thickness in successive layers, demonstrating a three-dimensionally self-ordered QD crystal.

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