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Self-organized strain engineering on GaAs (311)*B*: Template formation for quantum dot nucleation control

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A matrix of closely packed cells develops during molecular-beam epitaxy of $In_{0.35}Ga_{0.65}As$ on GaAs (311)*B*, due to strain-driven growth instability. The established lateral strain distribution generates a unique template that controls the nucleation and growth of InAs quantum dots (QDs). The QDs exhibit pronounced improvement of the structural and optical properties with efficient carrier transfer from the template. Thus, self-organization of a two-dimensionally connected quantum dot network is demonstrated. © *2002 American Institute of Physics.* [DOI: 10.1063/1.1516637]

Strained layer epitaxy in the Stranski–Krastanov (SK) growth mode to fabricate self-assembled quantum dots (QDs) has been the subject of intense study over the last decade. The major drawback of this method is the random nature of the nucleation process, usually leading to undesirable QD size fluctuations and uncontrollable QD positions. Selective growth on patterned substrates has been explored to control QD nucleation,[1–3] which is, however, strongly limited by the spatial resolution of lithography. Improvement of the uniformity has been achieved by stacking QD layers due to strain mediation.[4–7] But stacking multilayers allows only for gradual size uniformity improvement.

In this letter, we present a method to control nucleation and growth of InAs QDs by creation of a strain modulated (In,Ga)As template on GaAs (311)*B* substrate which is based on strain-driven growth instability. Growth instability of thin alloy films has been theoretically studied[8,9] and experimentally verified[10,11] in the epitaxy of strained $Si_{1-x}Ge_x$ on Si (100). Growth instability is characterized by nucleationless evolution of surface undulation with periodicity mainly given by the lattice mismatch. During growth the undulation height continuously increases while its periodicity is kept constant. On the contrary, the SK growth mode involves formation of a two-dimensional wetting layer followed by random island nucleation. The island height increases and saturates very abruptly and further growth mainly increases the island density. On GaAs (100), the growth of strained (In,Ga)As follows the SK mode while instable growth has been reported recently for the GaAs (311)A surface[12] to produce an undulated surface with nanometer-scale wire-like structures. QD nucleation then occurs preferentially on top of the wires but is random along their length. On GaAs (311)*B*, the undulation of the surface morphology is two dimensional, in the form of a matrix of closely packed cells. Due to the well defined nature of evolution with constant periodicity, the related two-dimensional strain modulation generates a uniform template for full control of the nucleation of InAs QDs.

The samples were grown by solid source molecular-beam epitaxy (MBE) on GaAs (311)*B* substrates. After oxide desorption a 300 nm thick GaAs buffer layer was deposited at 580 °C. The substrate temperature was then lowered to 500 °C for growth of $In_{0.35}Ga_{0.65}As$ with thicknesses between 1.3 and 2.1 nm. The $In_{0.35}Ga_{0.65}As$ layer was capped by 10 nm GaAs at 500 °C and an additional 100 nm thick GaAs layer at 580 °C. Thereafter, the same $In_{0.35}Ga_{0.65}As$ layer was repeated on the surface at 500 °C for morphological characterization. In a different sample, 0.23 nm InAs was deposited on the 1.4 nm thick $In_{0.35}Ga_{0.65}As$ layer at 500 °C for QD growth. For comparison, a sample with QDs formed by 0.46 nm InAs directly on the GaAs surface was grown under the same conditions. The growth rate of GaAs was 0.24 μm/h and that of InAs was 0.13 μm/h for (In,Ga)As growth while it was lowered to 0.0028 μm/h for InAs QD formation. The arsenic beam equivalent pressure was $8-9 \times 10^{-6}$ Torr. Photoluminescence (PL) measurements were carried out with the samples cooled down to 4.5 K in a cryostat and excited by a Nd:YAG laser at an excitation power density of 256 mW/cm^2. The surface morphology of the samples was measured in air by atomic force microscopy (AFM) in tapping mode.

The AFM images and line scans of the $In_{0.35}Ga_{0.65}As$ layers deposited on GaAs (311)*B* with different layer thicknesses are shown in Fig. 1. There is a marked change of the surface morphology when the layer thickness is increased from 1.3 [Fig. 1(a)] to 1.5 nm [Fig. 1(b)]. The surface of 1.3 nm $In_{0.35}Ga_{0.65}As$ is flat, without any pronounced features while 1.5 nm the surface develops a matrix of cells with average height modulation of 1 nm determined from the AFM line scan. Upon further growth the height of the cells increases gradually from 1.5 nm in Fig. 1(c) (thickness 1.7 nm) to 2.5 nm in Fig. 1(d) (thickness 2.1 nm). The area density of the cells remains unchanged. This evolution of the surface morphology clearly identifies strain induced growth instability, additionally confirmed by the PL results shown in Fig. 2. The PL peak position shifts continuously from 868 to 942 nm when the $In_{0.35}Ga_{0.65}As$ layer thickness is increased from 1.3 to 2.1 nm. The shift in the PL peak is accompanied by gradual broadening of the PL line due to fluctuations in cell height. In contrast, QD nucleation is commonly accompanied by abrupt changes of the PL peak position and linewidth due to the abrupt increase in island height. This sheds

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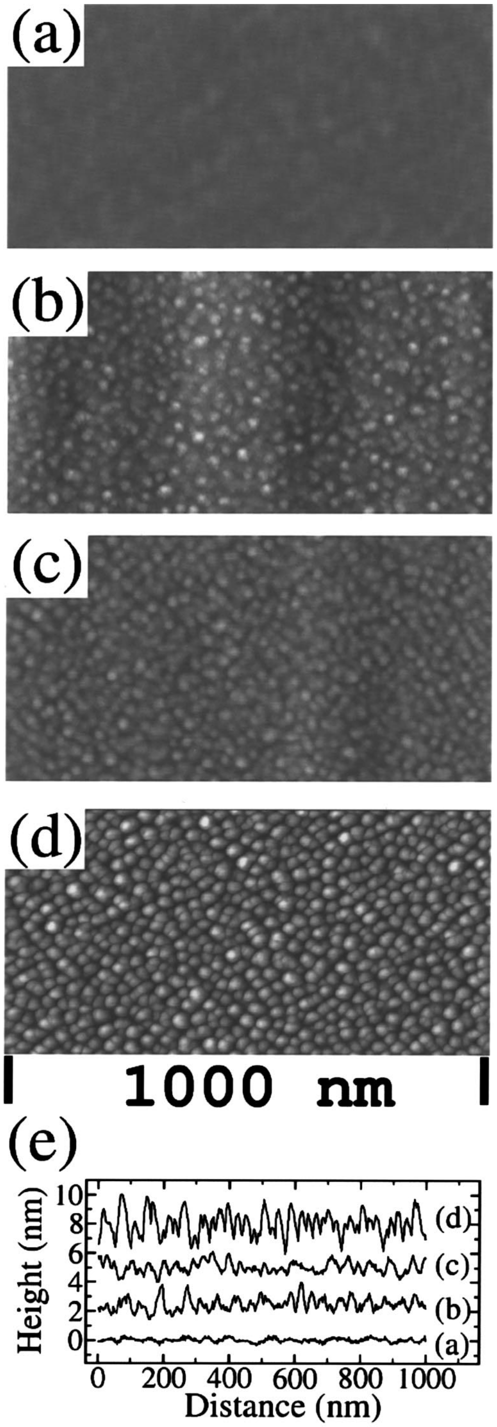

FIG. 1. AFM images of (a) 1.3, (b) 1.5, (c) 1.7, and (d) 2.1 nm thick $In_{0.35}Ga_{0.65}As$ deposited on GaAs (311)*B*. The height scale is 5 nm. AFM line scans along $[01\overline{1}]$ for (a)–(d) are shown in (e).

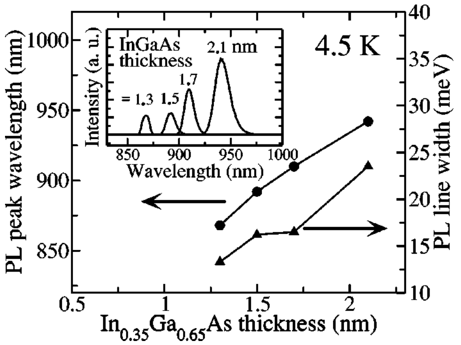

FIG. 2. Dependence of the PL peak wavelength and linewidth (at 4.5 K) on the $In_{0.35}Ga_{0.65}As$ layer thickness. The inset shows measured PL spectra.

new light on the formation mechanism and the nature of highly packed connected (In,Ga)As QDs on GaAs (311)*B* reported previously,[13–15] which represent a matrix of connected cells, formed due to strain-driven growth instability.

In the following this surface is explored as a template for InAs QD nucleation. Figure 3(a) shows an AFM image of InAs dots formed by 0.23 nm InAs at 0.0028 μm/h on the 1.4 nm thick $In_{0.35}Ga_{0.65}As$ layer. For comparison, InAs QDs formed directly on GaAs by 0.46 nm InAs are shown in Fig. 3(b). The corresponding AFM line scans are shown in Fig. 3(c). The dots on the GaAs surface with a density of only 6×10^9 cm^{-2} are randomly distributed and have large fluctuations in size. The surface between the QDs is flat, indicating QD nucleation on a wetting layer in the SK growth mode which occurs on GaAs (311)*B* for large lattice mismatched InAs. The dots on the (In,Ga)As template (three of them are marked by arrows in the lower left corner for clarity) are formed exclusively on top, in the center of the cells and are visible by the bright height contrast and the peaks in the line scan. Only a portion of the cells is occupied. The dot density can be enlarged from 2.4×10^{10} cm^{-2} to almost full cell coverage of 6×10^{10} cm^{-2} when the InAs layer thickness is increased to 0.38 nm, however, partial coverage is shown here to demonstrate the nucleation process (see below). A critical InAs thickness for dot formation of about 0.1 nm is measured from the distinct change of the reflection high-energy electron diffraction (RHEED) chevron pattern, which is much smaller than that on the GaAs surface of 0.42 nm due to In segregation during $In_{0.35}Ga_{0.65}As$ deposition. Hence, the presence of a wetting layer following the (In,Ga)As surface morphology (see AFM line scans between the peaks, i.e., InAs dots), abrupt QD height increase, evident from the line scan peaks, and increase of the QD density upon further growth are evidence of SK growth of InAs on the (In,Ga)As template. Most important, the (In,Ga)As template predetermines the nucleation sites of the InAs QDs and favors isolated QDs of higher density and size uniformity. This is underscored by the PL properties shown in Fig. 4. For the PL spectrum of InAs QDs on GaAs (dashed line), the narrow PL line at 869 nm is assigned to the wetting layer while the InAs QDs cause the broad line centered at 916 nm with linewidth of 42 meV. On the contrary, only one strong PL peak (solid line) is observed for the sample with InAs QDs formed on the 1.4 nm $In_{0.35}Ga_{0.65}As$ template with wavelength of 943 nm and reduced linewidth of 32 meV, indicating improvement of the QD size uniformity. Moreover, the PL peak intensity is more than 20 times higher than that from the InAs QDs on GaAs and no emission is observed from the (In,Ga)As template at 880 nm, indicating efficient carrier transfer from the template into the dots.

Control of the nucleation of QDs on the template is assigned to lateral strain modulation on the surface of the matrix of cells. Partial strain relief in the center of each cell is expected to generate local strain minima surrounded by a lateral strain field that increases towards the borders of each cell. These local strain minima are known to be preferential QD nucleation sites,[4,5,7,16] due to reduced lattice mismatch,

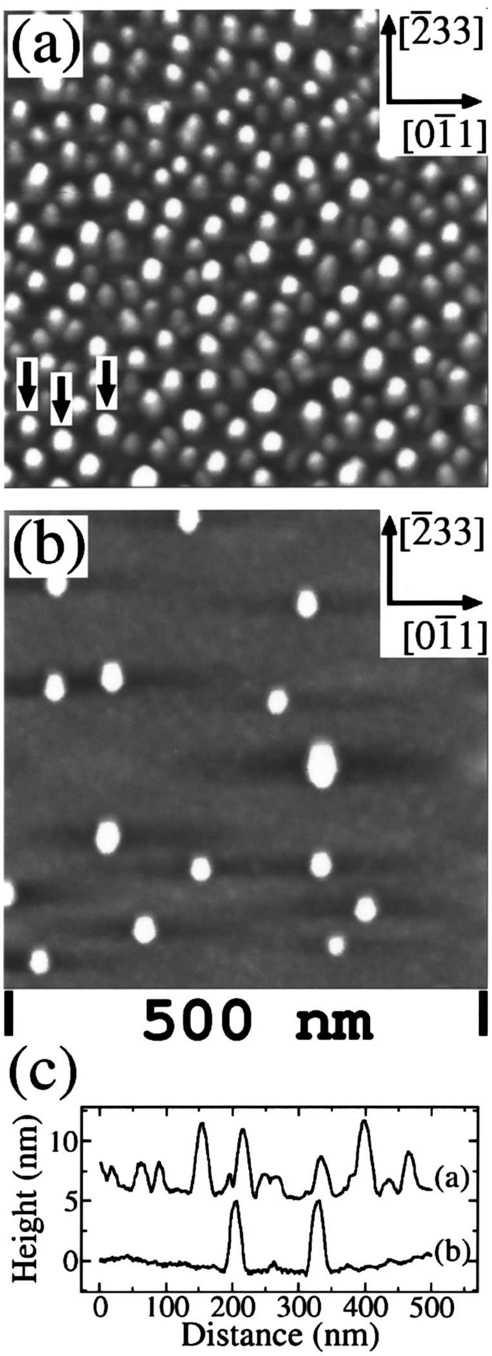

FIG. 3. AFM images of QDs formed at low growth rate by (a) 0.23 nm thick InAs on a 1.4 nm $In_{0.35}Ga_{0.65}As$ template and (b) by 0.46 nm InAs on GaAs. The height scale is 5 nm. AFM line scans along $[01\bar{1}]$ for (a) and (b) are shown in (c).

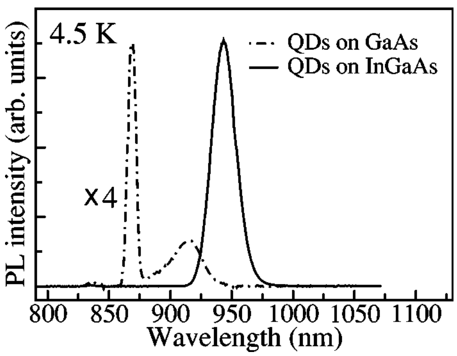

FIG. 4. PL spectrum (dashed line) measured at 4.5 K of the sample with QDs formed by 0.46 nm thick InAs directly on the GaAs surface. The solid line shows the spectrum of the QDs formed by 0.23 nm InAs on the 1.4 nm $In_{0.35}Ga_{0.65}As$ template.

and act as In adatom attractors due to strain-gradient-driven adatom migration. In fact, turning the argument around, the observed QD nucleation in the center of the cells can be taken to be a probe of the lateral strain field modulation and In distribution. This conforms with the lower amount of In required for island formation on the (In,Ga)As template due to In segregation and preferential In accumulation in the cell center. Moreover, the strain maxima at the borders of each cell provide barriers for In migration that limit the diameter of the effective collection area below the In adatom diffusion length, which relates the QD uniformity to that of the cell area. Thus, the template governs the nucleation site and collection area plus directed migration thereby suppressing the random nature of the nucleation process and controlling the QD growth and producing a uniform array of isolated InAs QDs. The QD density is directly determined by the InAs coverage to a maximum of the cell density which is much larger than that achieved on the GaAs surface. This is an important additional aspect of the template. The structural improvements of the QD array are reflected in the optical properties, most notably revealing efficient carrier transport from the template to the dots. Thus, the structure realized here can be viewed as a demonstration of self-organization of a two-dimensionally connected network of isolated QDs.

To summarize, we have studied the MBE growth of $In_{0.35}Ga_{0.65}As$ on GaAs (311)*B* to produce a matrix of closely packed cells due to strain-driven growth instability. The lateral strain distribution associated with these closely packed cells generates a unique template for the formation of InAs QDs. Control of the nucleation process strongly improves the size uniformity and increases the density of the InAs QDs which is reflected in superior optical properties.

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