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Fabrication of (In,Ga)As quantum-dot chains on GaAs(100)

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Nanostructure evolution during the growth of multilayers of $In_{0.5}Ga_{0.5}As$/GaAs (100) by molecular-beam epitaxy is investigated to control the formation of lines of quantum dots called quantum-dot chains. It is found that the dot chains can be substantially increased in length by the introduction of growth interruptions during the initial stages of growth of the GaAs spacer layer. Quantum-dot chains that are longer than 5 μm are obtained by adjusting the $In_{0.5}Ga_{0.5}As$ coverage and growth interruptions. The growth procedure is also used to create a template to form InAs dots into chains with a predictable dot density. The resulting dot chains offer the possibility to engineer carrier interaction among dots for novel physical phenomena and potential devices. © *2004 American Institute of Physics.* [DOI: 10.1063/1.1669064]

By reducing the three dimensions of semiconductor structures to the nanoscale, quantum dots (QDs) can be fabricated and shown to possess unique optical and electronic properties that can change the paradigm of electronic and optoelectronic devices. Among the approaches for the fabrication of compound semiconductor QDs, strain-driven self-assembly has been demonstrated to be one of the best for the generation of defect-free QDs. However, due to the stochastic nature of the nucleation of self-assembled growth, control of spatial ordering of the QDs has proved to be extremely challenging.

One approach to spatial ordering that produces vertical self-alignment occurs during QD stacking, and is based on a seeding effect resulting from the strain field transmitted from the QD layer just below.[1,2] In fact, the strain field from the underlying QDs, interacting through the spacer layer, not only causes vertical alignment but in some material systems also leads to lateral ordering, and even to three-dimensional QD "crystal-like" ordering.[3–6] However, for III–V semiconductors, such as (In,Ga)As QDs in a GaAs(100) matrix, the achievement of control of lateral ordering has dominantly relied on lithographic techniques,[7,8] which when compared to strain induced self-assembly, requires extra processing steps and can easily introduce defects into the QD structures.

A second approach to achieve lateral ordered (In,Ga)As QD arrays in a GaAs matrix has been realized using high index substrates, such (311) and (411).[9,10] In this case, the QD in-plane self-alignment is believed to originate from the anisotropy of the special crystallographic arrangement of high index surfaces. Even more recently, an unusual and perhaps unexpected approach to lateral ordering has been reported. This approach is based on the self-assembly of long lines of QDs, which are referred to as QD chains, and which form during the growth of multilayers of (In,Ga)As QDs separated by GaAs(100).[11–16] Compared to single layer (In-,Ga)As QDs, which are randomly distributed on the GaAs(100) surface, the long chains of QDs provide an exciting playing field to study carrier and optical interactions between the QD structures.

In this letter, the effect of GaAs capping and (In,Ga)As coverage on the formation of QD chains is investigated. It is found that by introducing growth interruptions during the initial stages of the growth of the GaAs spacer layer, the (In,Ga)As QD chains can be substantially lengthened. Long chains of QDs, over 5 μm along the $[01-1]$ direction, are achieved for particular (In,Ga)As coverage and growth interruptions.

In the experiment, epi-ready semi-insulting GaAs (100) substrates are first degassed for 20 min at 350 °C and then loaded directly into the growth chamber of a solid-source molecular beam epitaxy system. After the native oxide layer is desorbed by annealing the substrates at 610 °C for 10 min, a GaAs buffer layer of 0.5 μm is grown with a growth rate of 1.0 monolayer per second (ML/s) at 600 °C, under a constant As beam equivalent pressure of 1×10^{-5} Torr. The substrate temperature is then reduced to 540 °C for the growth of 16 $\times$(9.0ML $In_{0.5}Ga_{0.5}As$/60 ML GaAs) quantum dot multilayers. The growth rate of GaAs and $In_{0.5}Ga_{0.5}As$ is 0.4 and 0.8 ML/s, respectively. Finally, the last QD layer was typically deposited without a final GaAs capping layer so that the surface morphology can be characterized by atomic force microscopy (AFM).

Figure 1 shows AFM images of the surface morphologies after stacking 17 $In_{0.5}Ga_{0.5}As$ QD layers. For the structure imaged in Fig. 1(a), one growth interruption of 10 s is introduced after the first 3 ML GaAs spacer growth. For the structure imaged in Fig. 1(b), one growth interruption every 10 s per 3 ML GaAs growth is applied for the first 18 ML GaAs spacer growth. The QD density is 210/μm^2 for Fig. 1(a) and 205/μm^2 for Fig. 1(b). The difference is negligible when considering the statistical variation. The average QD size is also the same for both cases, 6 nm in height and 43 nm in diameter. Where there is a rather remarkable difference is in the length of the QD chains. The average length of the QD chains is about 1 μm for Fig. 1(a) but close to 2 μm for Fig. 1(b). The average chain length is about 0.7 μm without a growth interruption.[16] It is easy to see that the introduction of growth interruptions, during the initial stages of the growth of the GaAs spacer, significantly improves the QD alignment along $[01-1]$. Moreover, the QD alignment

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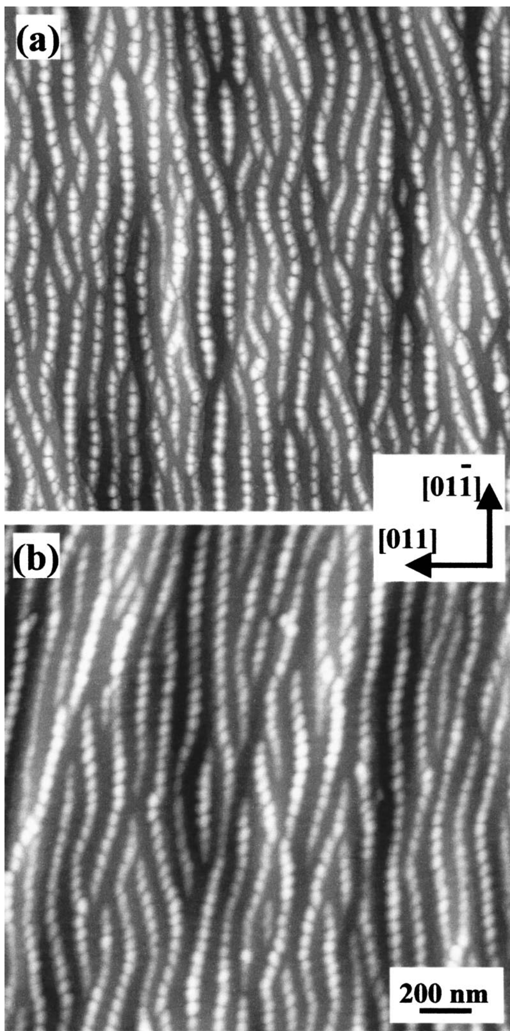

FIG. 1. 2 μm×2 μm AFM images of 9.0 ML $In_{0.5}Ga_{0.5}As$ QDs multilayers illustrating the effect of growth interruption during GaAs capping on the formation of QD chains: (a) one growth interruption after growth of the first 3 ML GaAs spacer layer; (b) one growth interruption per 3 ML for growth of the first 18 ML GaAs spacer layer.

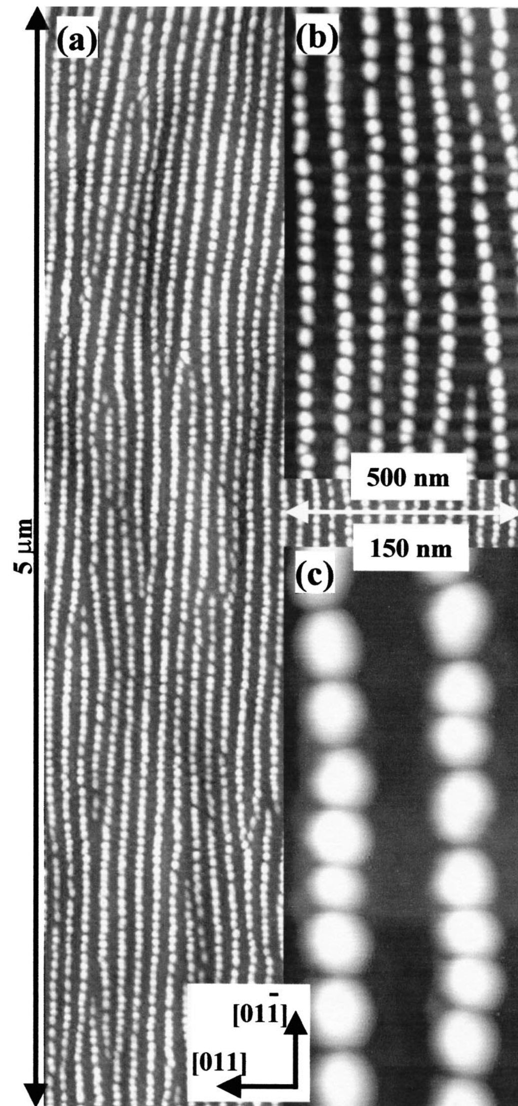

FIG. 2. AFM images of 6.0 ML $In_{0.5}Ga_{0.5}As$ QDs multilayers. (a), (b), and (c) are shown in different scales.

in the (In,Ga)As QD chains is also very sensitive to ML coverage.[17,18] Stacking 17 layers of 6.0ML $In_{0.5}Ga_{0.5}As$ QDs instead of 9.0 ML as in Fig. 1(b) results in remarkable long chains of uniform QDs. Figure 2 shows the AFM imaged surface in different scales. Most of QD chains are over 5 μm long along the $[01-1]$ direction. The average spacing of the QD chains is about 71 nm and the separation between QDs along the chains is about 36 nm. In this case, the QDs have a surface density of $350/\mu m^2$ with the average height of 7.5 nm.

Given the dramatic QD alignment observed in the chains, it is important to remember that the first layer consists of spatially random QD structures.[16] The chain-like lateral alignment of dots is gradually developed during the growth of (In,Ga)As multilayers. One simple model for lateral organization during a multilayer growth has been proposed in which dots are treated as spherical inclusions buried in a continuum elastic medium.[3,4] Under this model a minimum in the tensile strain field, with respect to the island material, at the spacer surface acts as a nucleation site for the next layer of islands. As a result, islands that are close together forming a close pair have merging stain fields that give rise to a single minimum centered between the two islands causing a single island to nucleate above the island pair. In this case the distance over which the strain field interacts is determined primarily by the spacer-layer thickness. Thus if the thickness is kept constant, island separations must eventually reach a stable distance beyond which they will no longer merge with one another. However, in our case, we observe chains of islands with a clear asymmetric separation. One possible explanation is that the strain field is not symmetric at all. The physical origin of the anisotropic strain field could result from the known asymmetry of the QD.[19,20] Since strain can drive material transport, resulting in strain relief, the anisotropic strain field triggers asymmetric material transport that could be enhanced by introducing growth interruptions, causing the asymmetric separation observed between adjacent three-dimensional islands.

As recently demonstrated, the density and size of the InAs QDs can be independently controlled in a much simpler bilayer system.[21–24] The QD density of the second layer is determined by the QD density in the first layer. However, the size of the QDs in the second layer is independently controlled by the amount of material deposited. We were also

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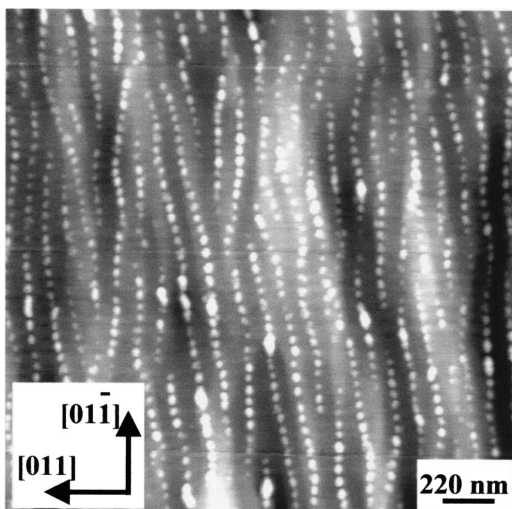

FIG. 3. 2.2 μm×2.2 μm AFM image of 2.0 ML InAs QDs grown on a template formed by 16×(9.0 ML $In_{0.5}Ga_{0.5}As$/60 ML GaAs) multilayers as same as the structure imaged in Fig. 1(b).

able to use the dot chain as a template with the capability to control the density and spatial ordering for InAs QD. After the growth of 16×($In_{0.5}Ga_{0.5}As$/GaAs) multilayers, as in the case of the structure shown in Fig. 1(b), the samples were cooled to 500 °C for InAs deposition. Figure 3 is the surface AFM image after 2.0 ML of InAs deposition. Many InAs QD chains run over 2 μm along $[01-1]$. The InAs QD chains have the same lateral periodicity as the $In_{0.5}Ga_{0.5}As$ QD chains in Fig. 1(b). However, since the InAs QDs are smaller than $In_{0.5}Ga_{0.5}As$ QDs, the InAs QDs are more separated in the chains. The average size of InAs QDs is 3.4 nm in height and 35 nm in diameter. While this is not a surprise considering the same template, this result demonstrates an approach to achieving (In,Ga)As QD chains with a wide range of In content. As expected, the InAs QD chains have the surface QD density of 200/μm^2, close to that in the $In_{0.5}Ga_{0.5}As$ QD chains in Fig. 1(b). Since the InAs QD density is determined by the template of the $In_{0.5}Ga_{0.5}As$/GaAs multilayers, the size of InAs QDs in the chains can again be independently controlled by the amount of material deposited, in the same fashion as in the bilayer system.[21–24] For further InAs deposition up to 2.5 and 3.0 ML, no obvious variation in QD density is observed but the average size of QDs is nearly doubled. This is in contrast to a report on a slightly different approach to produce (In,Ga)As chain-like structures that is based on engineering the high anisotropy of strain using (In-,Ga)As quantum wires.[14] While starting with quantum wires has its advantages, there is less control of the QD density.

In summary, a strain-driven self-assembly process to form one-dimensional QD arrays is demonstrated during the growth of (In,Ga)As/GaAs(100) multilayered stacks. The fully developed one-dimensional QD arrays together with the potential to use them as templates to achieve (In,Ga)As QD chains with controllable In content and QD size and density as demonstrated above could open a way to engineer the lateral carrier interaction among QDs. The demonstration may excite new experimental and theoretical efforts to explore the possibility of three-dimensional ordered quantum dot crystals.

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