Structural evolution in strained $\text{In}_{0.18}\text{Ga}_{0.82}\text{As}$ stacking multilayers on vicinal GaAs surfaces

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Abstract. Strain relaxation mechanisms for the growth of $m$-layers of $\text{In}_{0.18}\text{Ga}_{0.82}\text{As}/\text{GaAs}$ on a GaAs(100) substrate, tilted 2° towards the [0–11] direction, have been studied by molecular beam epitaxy (MBE) and atomic force microscopy (AFM). While dislocations alone provide a strain relaxation mechanism for nominal GaAs(100), additional strain relaxation mechanisms were observed for a vicinal GaAs(100) substrate. For $m \leq 8$, step bunching provided a mechanism for strain relaxation. For $m \geq 10$, in addition to the step bunching, bunched corners along two [051] and [0–1–5] directions provided the mechanism for strain relaxation. These surface patterns provide potential to act as a template for the growth of more uniform and organized nanostructures. For $m = 16$, the formation of dislocations provided an additional route for strain relaxation.

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1. Introduction

Achieving control of size, shape and position of nanostructures via growth by self-assembly has been a serious roadblock to the application of (InGa)As quantum dots (QDs) on a GaAs(100) substrate. As a result, lithographic techniques have been pursued to obtain the required control \([1]–[5]\). However, lithography has its own drawbacks, primarily introducing defects into the epitaxial growth. Recently, some new approaches have been employed to achieve control over size, shape and position of nanostructures by employing different substrate orientations or multilayer growth \([6]–[9]\). For example, by varying the surface orientation, surface diffusion and strain can be tuned to achieve organized and more uniform self-assembled QDs. In particular, vicinal GaAs(100) and high index surfaces have been used to demonstrate the ability to control diffusion of atoms and strain over the surface. Basically, the diffusion of atoms is limited by surface reconstruction of As dimers and step flow on the GaAs(100) surface. For example, anisotropy in surface diffusion can cause the formation of QD chains along \([0–11]\) due to the \((2 \times 4)\) surface reconstruction with dimer rows running along \([0–11]\) \([1]\). In principle, by fine-tuning the surface orientation to introduce steps on the surface, the surface diffusion length can be controlled in order to create isotropic surface diffusion and which is an excellent tool for growth of a more uniform nanostructure. Meanwhile, a multilayer of QDs can be used to tune strain to achieve vertical alignment as well as the uniform spacing, size and shape of nanostructures \([10, 11]\). Clear research results have also been complemented by theoretical models both of which demonstrated that vertical alignment of QDs in subsequent layers can lead to beautiful lateral ordering \([12, 13]\). Basically, when (InGa)As QDs are buried under a GaAs spacer layer, the transmitted strain distribution on the surface produces pronounced maxima and minima in the lateral plane. As a result, the next (InGa)As deposited layer recognizes a preferential nucleation of new QDs where the local strain is minima on the surface. Theoretical calculation shows that elastic energy distributions are determined by two key parameters, the elastic anisotropy of the matrix material and the lateral interaction of the strain field among QDs. Working together, it causes QD multilayer growth to become a successful approach to produce lateral ordered QD arrays. However, for (InGa)As with a low In composition less than 0.18, two-dimensional (2D) growth will proceed without formation of QDs. The build-up strain can only be released through formation of dislocation for the growth on singular GaAs(100) surfaces. For (InGa)As growth with a low In composition on vicinal GaAs(100) surfaces with orientation miscut, the build-up strain can be partly released through step-bunching \([14, 15]\). In this study, the uniformity and lateral ordering of the surface nanostructure, using a low In composition (InGa)As/GaAs multilayer system on vicinal GaAs (100) substrate, is investigated.

2. Experiments

The experiments were carried out in a molecular beam epitaxy (MBE) chamber equipped with reflection high-energy electron diffraction (RHEED). Double side polished, epitaxial ready \(n\)-type exact GaAs (100) and vicinal GaAs (100) substrates, tilted 2° towards the \([0–11]\) direction, are loaded into the MBE chamber. As a reference point, an exact GaAs(100) sample was loaded side-by-side next to each vicinal GaAs(100) sample, on a solid molybdenum block. The surface oxide layer was removed at 600 °C for 10 min, while exposed to a 6.4 \(\mu\)Torr As\(_4\) beam equivalent pressure (BEP) from a solid source valved controlled As cell. After oxide desorption,
the substrate temperature was lowered to 580 °C and a 0.4 µm thick GaAs buffer layer was grown with a growth rate of 1.0 monolayer per second (ML s\(^{-1}\)) and an As\(_4\) to Ga BEP ratio of ten. Subsequently, the substrate was cooled down to 540 °C, while keeping the As\(_4\) BEP the same for the deposition of a (InGaAs)/GaAs multilayer structure. The multilayer structure consists of \(m\) periods (with \(m = 1, 4, 8, 10, 12\) and 16) of 18 ML of In\(_{0.18}\)Ga\(_{0.82}\)As on top of 18 ML of a GaAs spacer with a 10 s interruption between each layer. The Ga growth rate during the growth of the In\(_{0.18}\)Ga\(_{0.82}\)As/GaAs multilayer structure was 0.37 ML s\(^{-1}\). The last layer of (InGa)As was left uncapped for topographic atomic force microscopy (AFM) imaging under ambient conditions. The growth was then followed by 10 s of annealing and the sample was then quickly quenched while lowering the As\(_4\) pressure.

After the growth, the structural quality of the epitaxial layers was characterized with a Phillips high-resolution x-ray diffraction system for (004) atomic planes. The mismatch between the average lattice constant of \([\text{InGaAs}]/\text{GaAs}\)_\(m\) multilayer system and the GaAs substrate was about 1.2%. The In composition, determined from the x-ray patterns, was 18% which was in good agreement with the In composition expected from the growth rate observed by RHEED oscillation.

3. Results and discussions

Figure 1 shows AFM images of the surface morphologies of (In\(_{0.18}\)Ga\(_{0.82}\)As/GaAs)_\(m\) after stacking of (a) \(m = 1\), (b) \(m = 4\), and (c) \(m = 8\) periods. In all samples, the 18 ML of In\(_{0.18}\)Ga\(_{0.82}\)As were deposited on top of 18 ML of a GaAs spacer. Similar to previous observations [10], the surface is characterized by a high density of steps running along \([0–1 1]\) with nonuniform edges (figure 1(a)). The average spacing between steps is about 80 nm which is ten times bigger than the average spacing between steps on a bare vicinal GaAs(100) surface. Since the average steps are about 8 nm wide on the bare GaAs(100) vicinal surface, there are roughly ten steps bunched together to form a big step on In\(_{0.18}\)Ga\(_{0.82}\)As. The height modulation corresponding to the ten bunched steps is around 2.8 nm. In figure 1(a), the root mean square (RMS) roughness is about 0.5 nm. The bunched steps are straightly extended over 500 nm. The formation of the step bunching can be explained by the elastic relaxation leading to a logarithmic attraction between steps regardless of the step density or the As flux [14]–[17].

As is seen in figure 1(b) and (c) when compared to figure 1(a), by adding more In\(_{0.18}\)Ga\(_{0.82}\)As/GaAs layers, the step bunches not only become much straighter but also exhibit a remarkably ordered distribution. The surface ordering of steps induced by multilayer stacking of In\(_{0.18}\)Ga\(_{0.82}\)As/GaAs overgrowth is shown by the fast Fourier transform (FFT) spectrum over the area of 5 µm x 5 µm on the inserts of figure 1. When compared to the insert of figure 1(a) and (b), the insert of figure 1(c) reveals a more ordered array of steps along the [0–1 1] direction. The steps on figure 1(b) and (c) are bunched with the same periodicity of 80 nm. The height of the steps and the RMS roughness of the surface for figure 1(b) and (c) are the same as in figure 1(a) which indicate that adding more layers on top of buried In\(_{0.18}\)Ga\(_{0.82}\)As has no significant effect on the number and height of bunched steps. However, additional layers do significantly improve the ordering of bunched steps. The bunched steps are straightly extended over more than 1 µm in figure 1(b) and (c). In figure 1(c) for \(m = 8\), the size and shape uniformity is more prominently noted. The lateral uniformity of size and shape in figure 1(c) is correlated with the fact that the multilayering leads to size, shape and spacing uniformity [13]. The ordered step bunching in
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Figure 1. AFM (5 μm x 5 μm) image of \((\text{In}_{0.18}\text{Ga}_{0.82}\text{As}/\text{GaAs})_m\) (a) \(m = 1\), (b) \(m = 4\), and (c) \(m = 8\) multilayer illustrating the effect of step bunching in nanostructure growth on vicinal GaAs (100) tilted 2° towards the [0–11] direction. Inserts are the FFT spectrum and line profile along the [0–11] direction across the line on the image. Orientations on all images are the same.

Figure 1(c) has potential as a template for an ordered array of self-assembled strained quantum wires. Meanwhile, the surface morphology of the reference GaAs(100) loaded with the vicinal samples shows a 2D layer-by-layer epitaxial growth with no significant dislocations.

In addition to the investigation of \(m = 8\) periods of \((\text{In}_{0.18}\text{Ga}_{0.82}\text{As}/\text{GaAs})_m\), we also grew \(m = 10\), \(m = 12\) and \(m = 16\) periods on both vicinal GaAs(100) side-by-side to a reference GaAs(100) substrate under the same growth conditions. After \(m = 8\) periods of \((\text{In}_{0.18}\text{Ga}_{0.82}\text{As}/\text{GaAs})_m\), adding more layers makes the surface rough which can be explained by the accumulation of strain on the surface. The AFM image from the reference GaAs (100) sample loaded with the vicinal sample, with \(m = 10\) periods, shows dislocations, indicating relaxation due to high strain accumulation. The strain accumulation in a multilayer system is similar to the accumulation of strain by increasing the In concentration in a single layer in that both result in rough surface [15]. Figure 2(a) shows an AFM image of the surface morphology of \((\text{In}_{0.18}\text{Ga}_{0.82}\text{As}/\text{GaAs})_m\) \(m = 10\) periods. In figure 2(a), strain relaxation rather amazingly, makes the step bunching irregular, with a larger average separation of about 104 nm and a higher modulation of about 3.6 nm. In figure 2(a), the RMS roughness is 5.1 nm which is almost ten times higher than the RMS roughness observed earlier in figure 1. Moreover, the step bunching is no longer continual along the [011] direction. Figure 2(a) shows a small density of corners in a background of steps running along the [0–11]. The corners can be characterized as short bunched steps, which will be called bunched corners, along the [0–1–5] and [051] directions.
Figure 2. (a) AFM (5 µm × 5 µm) image of (In$_{0.18}$Ga$_{0.82}$As/GaAs)$_{10}$ multilayer illustrating the effect of step bunching in nanostructure growth on vicinal GaAs (100), tilted 2° towards the [0–11] direction. Two [0–1–5] and [051] directions were marked with dashed line indicating the bunched corners on the image. The inserts is the line profile along the [0–11] direction across the line on the image. (b) A FFT spectrum of figure 2(a). The dashed lines indicate the 112° angle between two sides of arrow shape of the FFT spectrum.

The upper leg of bunched corners makes an angle of $-34°$ from the [0–1–1] direction, while the lower leg makes $+34°$ from the [011] direction. Both the [015] and [0–5–1] directions on the bunched corners are similar to each other by the fact that they make an angle of about $34°$ from the [0–11] plane. Figure 2(b) shows the FFT spectrum form 5 µm × 5 µm AFM image of (In$_{0.18}$Ga$_{0.82}$As/GaAs)$_{10}$ multilayer. The bunched corner angle of 112° can be confirmed by the angle between two ends of the arrow shape figure seen in the FFT spectrum, marked by the dashed line in figure 2(b). Comparing $m = 8$ periods in figure 1(c) to $m = 10$ periods in figure 2(a), the bunched corners start to break down the straight and long steps to irregular and smaller steps. Formation of bunched corners demonstrates that both step bunching and bunched corner participate in the surface relaxation. The formation of bunched corners in $m = 10$ periods also indicates that the strain accumulation on the samples with $m \leq 8$ periods is not strong enough to create bunched corners. Adding more layers confirms the high strain accumulation effect by observation of the surface morphology for the sample with $m = 10$ periods. Indeed, by increasing the thickness of spacer layer from 18 to 45 ML, the surface morphology of the sample with $m = 10$ periods changes from the surface with bunched corners to the step bunching morphology of figure 1(c) as a result of the reduction in strain transfer. The surface relaxation along the [0–1–5] and [051] directions seen in figure 2(a), indicates that further relaxation of accumulated strain is preferred along the [0–1–5] and [051] directions leading to the formation of bunched corners. The maximum length of a bunched corner in figure 2(b) does not exceed 370 nm, which is large enough to cover two bunched steps along the [0–11] direction in figure 1(c). The maximum length of a bunched corner can be correlated with the fact that the bunched corner is not the only strain relaxation mechanism on the surface and is limited by another mechanism.

Due to the existence of high density of ML steps on the GaAs vicinal surface, the accumulated strain during (InGa)As stacking is first relaxed through step bunching. However, in the relaxation mechanism through step bunching, the built strain can only relieve along one direction, which is not efficient enough at the later stage. The further build-up strain in the $m$-layers of (InGa)As/GaAs with $m = 10$ finds other directions to relieve, [0–1–5] and [051] in the...
this case. [0–1–5] and [051] are two directions with relatively low indexes, which often mean relatively simple crystal structures and less energy costs to create such surface modulations.

By adding more \((\text{In}_{0.18}\text{Ga}_{0.82}\text{As}/\text{GaAs})_m\) layers, the (InGa)As surface gets more and more rough and irregular as a result of the accumulation of strain. The average separation of steps are 110 and 130 nm for the sample with \(m = 12\) and \(m = 16\) periods of \((\text{In}_{0.18}\text{Ga}_{0.82}\text{As}/\text{GaAs})_m\), respectively. For the sample with \(m = 16\) periods, the corners are irregular and less uniform. In addition to the rough surface and step bunching, dislocations start to appear on the surface of \(m = 16\) periods of \((\text{In}_{0.18}\text{Ga}_{0.82}\text{As}/\text{GaAs})_m\) on the vicinal GaAs(100). The surface morphology of the reference GaAs(100) is still 2D growth, however, the surface has a many dislocations. Ten dislocations were observed on 5 \(\mu\text{m} \times 5 \mu\text{m}\) AFM image from the reference GaAs(100) sample with \(m = 16\) \((\text{In}_{0.18}\text{Ga}_{0.82}\text{As}/\text{GaAs})_m\). In comparison, while the strain relaxation is mainly carried out by dislocations on the reference GaAs(100) surface, for the vicinal GaAs(100), step bunching, bunched corners, rough surface, and in the extreme, dislocations, accounted for strain relaxation. The density of regular bunched corners reduced as the number of stacked periods increased from \(m = 10\) to \(m = 12\) and \(m = 16\) period of \((\text{In}_{0.18}\text{Ga}_{0.82}\text{As}/\text{GaAs})_m\) however, still corners with the same angle separation could be recognized on the surface with \(m = 12\) and \(m = 16\) periods. The presence of dislocation on \(m = 16\) periods is the result of the further strain relaxation that maintained 2D growth on a vicinal GaAs(100) substrate. As a result, we believe that strain relaxation by the four mechanisms of step bunching, bunched corners, rough surface, and dislocation, demonstrate that by tuning the In concentration, thickness of spacer, and growth condition of \((\text{In}_{0.18}\text{Ga}_{0.82}\text{As}/\text{GaAs})_m\) multilayer structure, a unique organized and uniform multilayer of self-assembled nanostructures can be fabricated on vicinal GaAs(100).

4. Conclusions

In summary, the evolution of surface morphology during the stacking of multiple layers of InGaAs with an In content of 0.18 is investigated by MBE and AFM. For such a low In content, the growth is 2D on singular GaAs(100), with the formation of dislocations as the only mechanism for strain relaxation. In contrast, on the substrate with a 2° tilted towards [01–1], the strain is partly released through step bunching. Through multilayering up to eight periods of InGaAs/GaAs, the bunched steps are increasingly equally spaced, ordered and highly anisotropic, providing potential as a template for the growth of quantum wires. With more layers of InGaAs stacking, the accumulated strain is relaxed through the formation of bunched corners along the [051] and [0–1–5] directions. These provide interesting additional strain relaxation routes for the growth of InGaAs/GaAs multiple layers with low In content on a vicinal GaAs(100) substrate. For such multiple heterostructures, the synchrotron x-ray measurement and analysis on the associated samples are under way to reveal the vertical evolution of strain and structure.

References

[1] Konkar A, Heitz R, Ramachandran T R, Chen P and Madhukar A 1998 J. Vac. Sci. Technol. B 16 1334
[2] Lee H, Johnson J A, He M Y, Speck J S and Petroff P M 2001 Appl. Phys. Lett. 78 105
[3] Ying B, Liu F and Lagally M G 2004 Phys. Rev. Lett. 92 025502
[4] Song H Z, Usuki T, Ohshima T, Sakuma, Kawabe, Okada Y, Takemoto K, Miyazawa T, Hirose S, Nakata Y, Takatsu M and Yokoyama N 2006 Nanoscale Res. Lett. (DOI: 10.1007/s11671-006-9012-x)
Kiravittaya S, Songmuang R, Rastelli A, Heidemeyer H and Schmidt O G 2006 Nanoscale Res. Lett. (DOI: 10.1007/s11671-006-9014-8)

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[5] Wang Zh M, Lee J H, Liang B L, Black W T, Kunets V P, Mazur Yu I and Salamo G J 2006 Appl. Phys. Lett. 88 233102
[6] Lan S, Akahane K, Song H Z, Okada Y and Kawabe M 1999 J. Vac. Sci. Technol. B 17 1105
[7] Meng X Q, Jin P, Xu B, Li C M, Zhang Z Y and Wang Z G 2002 J. Cryst. Growth 241 69
[8] Wang Z M, Holmes K, Mazur Yu I and Salamo G J 2004 Appl. Phys. Lett. 84 1931
   Li S S and Xia J B 1994 Phys. Rev. B 50 8602
[9] Shmidbauer M, Seydmohamadi Sh, Grigoriev D, Wang Zh M, Schafer P, Hanke M, Kohler R
   and Salamo G J 2005 Phys. Rev. Lett. 96 066108
[10] Wu W, Tucker J R, Solomon G S and Harris J S 1997 Appl. Phys. Lett. 71 1083
[11] Holy V, Springholz G, Pinczolits M and Bauer G 1999 Phys. Rev. Lett. 83 356
[12] Tersoff J, Teichert C and Lagally M G 1996 Phys. Rev. Lett. 76 1675
[13] Liu F, Davenport S E, Evans H M and Lagally M G 1999 Phys. Rev. Lett. 82 2528
[14] Phang Y H, Zhang Z and Lagally M G 1995 Phys. Rev. Lett. 75 2730
[15] Wang Z M, Shultz J L and Salamo G J 2003 Appl. Phys. Lett. 83 1749
[16] Liu F, Tersoff J and Lagally M G 1998 Phys. Rev. Lett. 80 1268
[17] Wen H, Wang Zh M, Shultz J L, Liang B L and Salamo G J 2004 Phys. Rev. B 70 205307