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The fabrication of self-aligned InAs nanostructures on GaAs(331)A substrates

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Abstract

Self-aligned InAs quantum wires (QWRs) or three-dimensional (3D) islands are fabricated on GaAs(331)A substrates by molecular beam epitaxy (MBE). InAs QWRs are selectively grown on the step edges formed by GaAs layers. The surface morphology of InAs nanostructures is carefully investigated by atomic force microscopy (AFM) measurements. Different growth conditions, such as substrate temperature, growth approaches, and InAs coverage, exert a great effect on the morphology of InAs islands. Low substrate temperatures favour the formation of wirelike nanostructures, while high substrate temperatures favour 3D islands. The shape transition is attributed to the trade-off between surface energy and strain energy. A qualitative agreement of our experimental data with the theoretical results derived from the model proposed by Tersoff and Tromp is achieved.

1. Introduction

During recent decades, the fabrication of defect-free quantum dots (QDs) based on the Stranski–Krastanov (SK) growth mode has attracted much attention due to their unique physical properties and potential device applications [1–5]. The stochastic nature of these islands leads, however, to undesirable nonuniformity in their size as well as their spatial distribution. It is almost impossible to control the nucleation of QDs in an organized way simply by using the SK growth mode. The size fluctuation also gives rise to large inhomogeneous spectral broadening. This seriously limits potential device applications of the QDs.

To solve these problems, many researchers have made considerable efforts to manipulate the size, the position, and the density of the QDs [6–8]. Most of the methods rely on conventional lithography [9–12] or electron beam techniques [13, 14]. The complicated procedure and the etched surface, however, are still the main drawbacks. Subsurface strain fields have also been introduced to control island nucleation, as seen in vertically stacked QDs [15, 16]. Despite these efforts, manipulation of the QD position is still a great challenge.

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In contrast, a feasible method of fabricating spontaneously ordered QDs or quantum wires (QWRs) is to use the periodic steps formed on the vicinal [17] or high-index substrates [18]. However, most of works with respect to QWRs focus on the GaAs/Al(Ga)As system [19]. Up to now, little work has been reported on self-organized InAs QWRs, since this material system always forms three-dimensional (3D) islands in SK growth mode. Additionally, the shape transition mechanisms of InAs nanostructures are still poorly documented.

In this paper, we report the fabrication of wirelike or 3D InAs nanostructures on GaAs(331)A substrates by MBE. We also discuss the shape transition of InAs nanostructures as well as the evolution mechanisms.

2. Experimental procedure

The samples were prepared by MBE on (331)A GaAs semi-insulating substrates. A 200 nm thick GaAs buffer layer was deposited at 580 °C, and then the substrate temperature was heated up to 610 °C for growth of a 150 nm GaAs layer. To form higher steps, the substrate temperature was maintained at 610 °C for annealing for 10 min. Then the substrate was cooled down to deposit various thicknesses of InAs layers. The InAs growth temperature varied between 450 and 540 °C. Two approaches were adopted to deposit the InAs layer: continuous deposition (without interruption) and intermittent deposition (i.e., growth of InAs layers under a repeated growth sequence of 0.1 ML InAs and a 5 s interruption). The In flux was calibrated to be 0.1 ML s^{-1} with reflection high-energy electron diffraction (RHEED) oscillations. In order to improve the homogeneity of the InAs growth, the substrate was rotated during layer deposition. As soon as the growth was over, the sample was immediately cooled down to room temperature. The surface morphology was studied with a Digital Instruments Nanoscope III AFM at room temperature in contact mode under air.

3. Experimental results

Figure 1 shows the AFM morphology of GaAs steps formed by a 150 nm GaAs layer. Quasi-periodic multi-atomic step arrays develop along the $[1\bar{1}0]$ direction, as shown in figure 1(a). RHEED patterns reveal that the GaAs(331)A surface is constituted of (110) terraces and (111)A steps, similar to previous work [20]. The bright and dark steps along $[1\bar{1}0]$ correspond to the (110) and (111)A surfaces, respectively. A line profile of the cross section measured along the dark line in figure 1(a) indicates that the lateral periodicity and the height of the steps was about 90 and 1.5 nm respectively, as shown in figure 1(b).

Figures 2(a)–(c) show the 3D surface AFM images of 2.5, 3.5 and 8 ML InAs grown on the corrugated GaAs surface, respectively. The InAs layer was grown at 510 °C in a continuous deposition mode. Obviously, the surface morphology of InAs strongly relies on the thickness of InAs layer. Flat cuboid-like InAs nanostructures elongated along the $[1\bar{1}0]$ direction, together with multi-faceted surfaces, can be observed after the deposition of 2.5 ML InAs (figure 2(a)). The sizes of cuboid-like InAs along $[1\bar{1}0]$ vary from 200 to 400 nm while the lateral periodicity remains almost constant (about 90 nm). As the lateral sizes of InAs nanostructures approach the step width, we can hardly distinguish the steps from the AFM images.

When the thickness of the InAs layer increases to 3.5 ML, the cuboid-like InAs nanostructures evolve into wirelike ones, as shown in figure 2(b). The faceted surfaces become blunt and rounded to a large extent. The length of InAs QWRs shows a bigger fluctuation along $[1\bar{1}0]$. In some cases, it amounts to 1100 nm, which results from the combination of InAs QWRs along the step lines. The width, however, is approximately the same. These facts indicate that the aspect ratio of the InAs islands increases with the thickness of InAs layer.

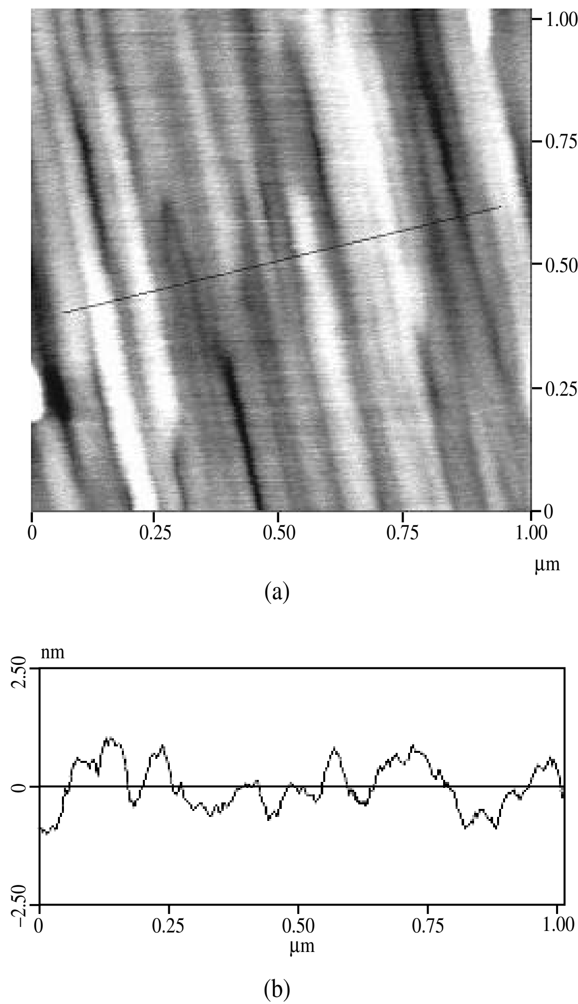

Figure 1. (a) The AFM morphology of the multiatomic steps formed on a GaAs epi-layer surface. (b) Line profile of the steps scanning along the dark line in (a).

On further increasing the InAs thickness to 8 ML, closely coupled InAs QWRs with a larger height undulation develop along $[1\bar{1}0]$. The aspect ratio of the InAs islands tends to become larger. Additionally, some irregular islands coexist with elongated InAs QWRs, resulting in a little instability of the periodicity and shape.

Interestingly, the lateral periodicity and height modulation amplitude of InAs QWRs can be doubled by growing the InAs layer in an intermittent deposition mode, as shown in figure 2(d). As a result, the area density of the QWRs decreases to some extent. This phenomenon should be assigned to the sufficient diffusion of In adatoms.

More surprisingly, the InAs growth mode can undergo an apparent conversion by changing the growth temperature, as shown in figure 3. At lower growth temperatures (450–510 °C), QWR growth dominates the growth process (see figures 2 and 3(a)). However, this turns to 3D island growth when the substrate temperature increases to 540 °C (see figure 3(b)).

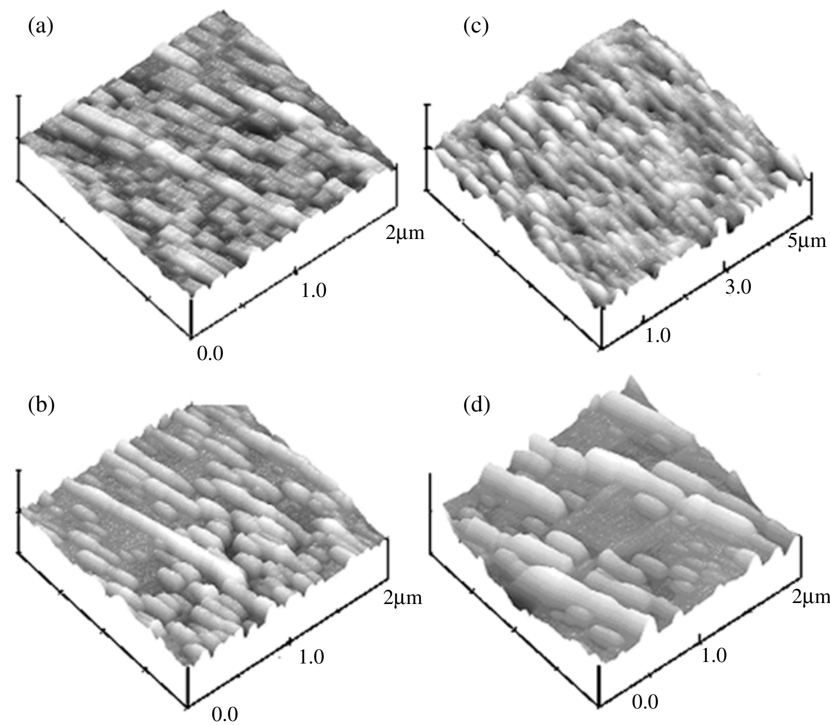

Figure 2. Three-dimensional AFM images of InAs nanostructures grown at 510 °C. (a)–(c) show the morphology of 2.5, 3.5 and 8 ML InAs grown in a continuous deposition mode, respectively. (d) presents the image of 3.5 ML InAs deposited in an intermittent mode.

4. Discussion and conclusion

As mentioned above, three key factors (InAs layer thickness, growth approach, and substrate temperature) account for the surface morphology evolution of InAs layer.

At a lower substrate temperature, though the surface morphology of InAs shows distinct changes with different InAs thickness or growth approach, wire-like growth dominates the growth process. This phenomenon can be partly assigned to the step bunching effect. First, a larger diffusion rate of In adatoms along the step lines is of benefit for the formation of elongated structures. The diffusion anisotropy of In adatoms along different crystallographic directions has already been reported [21]. Second, the step edges are energetically preferred by the In adatoms. To release the surface energy of the whole system, InAs nanostructures will preferentially develop along the step lines. Furthermore, InAs 3D island nucleation on GaAs(100) substrates under the same growth conditions indirectly prove that the wire formation bears some relation to step bunching.

However, the problem is complicated by the fact that the InAs layer can also form 3D islands at a higher temperature. If step bunching is the main driving force for the formation of wire structures, then how can we elucidate the transition of the InAs growth mode at the higher temperature? These facts indicate the following.

(1) Step bunching is not sufficient for the formation of wires but may serve to enhance this effect.

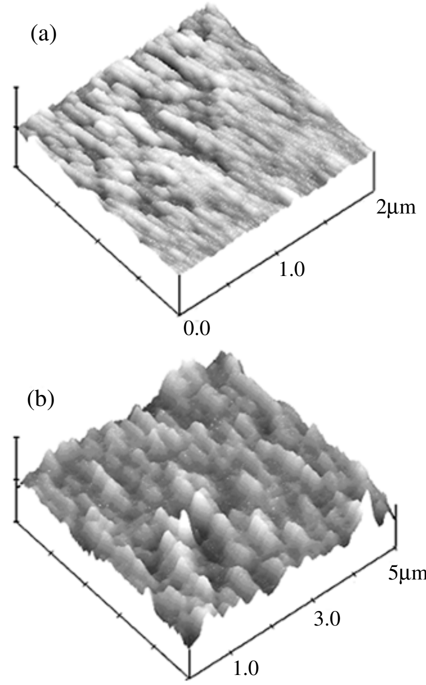

Figure 3. The surface morphology of InAs nanostructures grown at different temperatures: (a) 450 °C and (b) 540 °C. The InAs layer was grown in a continuous deposition mode.

(2) A comprehensive explanation of InAs growth should take into account both the thermodynamics and kinetic theories during the growth.

It is known that the evolution of the equilibrium crystal shape (ECS) in relation to surface faceting can be regarded as a thermodynamic process of minimizing the surface free energy. On the other hand, the formation of 3D islands is of benefit for the relaxation of the strain energy caused by the large lattice mismatch [22]. There must be a competition between the surface energy and the strain energy to reach a state with lower energy. In the following, we adopt the model proposed by Tersoff and Tromp [23] to present a qualitative description for this competition. According to their theory, the energy per unit volume (E/V) of a rectangular strained epitaxial island can be written as

$$\frac{E}{V} = 2\Gamma\left(\frac{1}{s} + \frac{1}{t}\right) - 2ch\left[\frac{1}{s}\ln\left(\frac{s}{\phi h}\right) + \frac{1}{t}\ln\left(\frac{t}{\phi h}\right)\right] \tag{1}$$

where s, t and h are the width, length and height of the island, respectively; $\phi = e^{-2/3}\cot\theta$, θ being the contact angle; Γ contains the surface and interface energies; c involves the bulk stress in the island and the Poisson ratio and shear modulus of the substrate. The first term describes the change in surface and interfacial energy when InAs elongated islands form on the surface in the absence of a wetting layer. The second term gives the bulk stress in the islands due to the substrate–island lattice mismatch. We can derive from expression (1) that the surface energy dependent term prefers to have a large area island for stability (elongated islands). On the other hand, the strain relaxation energy term favours islands of smaller area for greater stability (3D islands). The optimal trade-off between surface energy and strain energy is obtained through the minimization of E/V with respect to s and t under the limit of

constant island height (h). This gives $s = t = \alpha_0$, where

$$\alpha_0 = e\phi h \mathrm{e}^{\Gamma/ch}. \tag{2}$$

For island sizes $s, t < e\alpha_0$, the square island shape ($s = t$) is stable. Once the island grows beyond its optimal diameter α_0 by a factor of e, the square shape becomes unstable and a transition to rectangular shape takes place. As the island grows, the aspect ratio t/s becomes ever larger.

The conclusions derived from the model are in good agreement with our experimental results presented above. This suggests the model's partial validity and generality, though the model was initially proposed for the Ge/Si system. It also proves the theoretical assumption that the island height is kinetically limited is adequate. In fact, our experimental results indeed suggest that the island height grows much more slowly than the length.

We deduce that (if the model is true), different growth conditions, especially the substrate temperature, have a significant effect on the ratio of surface/interfacial energy to strain energy. Considering that the strain is mainly determined by the lattice mismatch between InAs and GaAs, the temperature should have a weaker effect on the strain energy in InAs islands with a fixed volume, i.e., the second term in expression (1). Therefore, we assume that the substrate temperature must have a larger impact on the surface/interfacial energy (the first term in expression (1)). In fact, it has been proved that the surface/interfacial energy does change with the temperature [22] and high temperature favours isotropic growth [24]. Tersoff and Tromp [23] thought thermal fluctuations at high temperature could lower the free energy of the island edges by increasing diffusion. Similarly, we conclude that high temperature may lead to the increase in the ratio of strain energy to surface energy. To more effectively relax the larger strain energy, InAs islands will adopt a compact shape (3D islands) at high temperature. As a result, the shape transition from elongated islands to square islands proceeds spontaneously, due to the larger contributions of strain energy over the surface energy. Similar elongated islands have been found in different heteroepitaxial systems such as Ge/Si [25], Au/Ag [26] and Au/Mo [27].

Finally, we emphasize that anisotropic stress along the two differently oriented step edges plays an additional role in the formation of the InAs QWRs. Theoretically, the island should align itself perpendicular to the direction of maximum stress [23]. Therefore, we infer that the direction of maximum stress is $[11\bar{6}]$.

In conclusion, different InAs nanostructures were fabricated based on various growth conditions. The surface morphology evolution of InAs nanostructures was investigated by AFM. Step bunching can enhance wirelike nanostructure formation. The shape transition of InAs layers was attributed to the trade-off between surface energy and strain energy in the islands.

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