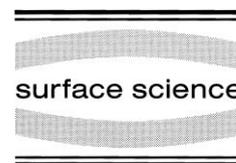




ELSEVIER

Surface Science 386 (1997) 169–181



# Self-organized nanoscale structures in Si/Ge films

Feng Liu, M.G. Lagally \*

*University of Wisconsin-Madison, Madison, WI 53706, USA*

Received 29 January 1997; accepted for publication 14 April 1997

---

## Abstract

Nanotechnologies of the future will demand the creation of large arrays of nanoscale structures and morphologies for electronic and optoelectronic devices. One approach to the creation of such arrays is to “let nature do it”. By depositing on each other semiconductor materials that differ slightly in lattice constant, one can use the resulting lattice strain to obtain arrays of three-dimensional crystallites or “quantum dots”. Repeated deposition of alternate layers of one material and the other into a multilayer film enhances the regularity in position and size of these quantum dots, giving the prospect of a three-dimensional array of tiny structures that can communicate electronically but without wires. The concept is illustrated with experiments using SiGe films deposited on Si(001). © 1997 Elsevier Science B.V.

*Keywords:* Atomic force microscopy; Germanium; Growth; Heterostructures; Scanning tunneling microscopy; Semiconducting interfaces; Semiconducting surfaces; Silicon; Surface stress

---

## 1. Introduction

Although the microelectronics industry has continued to amaze skeptics with its ability to respond to new technological issues as the dimensions of devices have shrunk, it is becoming clearer that at some scale present device fabrication technology will reach limits at which it becomes economically feasible to consider alternative approaches to computing and data storage. The miniaturization trend, were it to continue along the path articulated by Moore’s Law [1,2], would soon reach into the nanoscale, conventionally defined to be below several tens of nanometers, except for likely limits in the fabrication process [1]. Yet limits set by physical laws for present CMOS-based device strategies

may occur only at much smaller scales, suggesting a significant opportunity for the development of nanolithographies, nanoprocessing methods, and nanocharacterization technologies that will enable the present paradigm for silicon electronics to reach its ultimate limits. As illustrated in the article by Quate [3], in this volume, scanned-probe techniques, led by the scanning tunneling microscope and the atomic force microscope, are significant contenders as the silicon nanoprocessing and characterization tools of the future. Yet eventually physical limits (such as tunneling currents through gate oxides [1] or statistical effects in doping very small structures) in the present CMOS paradigm may require entirely new approaches to computing. One possible avenue is the use of heterojunctions (alloys of Si, Ge, and/or C in thin-film form in contact with alloys of a different composition or with elemental Si or Ge to provide internal poten-

---

\* Corresponding author. Fax: (+1) 608 265.4118;  
e-mail: lagally@engr.wisc.edu

tials through band offsets), paralleling the very successful approach used in III–V semiconductor technology. A unique capability of such Group IV heterojunction devices would be operation at the nanoscale level, i.e. operation as quantum confined devices or quantum tunneling structures, through structural miniaturization. One-dimensional miniaturization is easily achieved, by forming layered films using atomic beam epitaxy or chemical vapor epitaxy. However, unique quantum properties, some based on the actions of a single electron [2,4], can be achieved only with two- or three-dimensional confinement. Regular arrays of well-defined “quantum dot” structures would be particularly valuable in such applications. Such structures must be different enough in composition from the surrounding matrix so that significant band offsets occur, they must be small enough so that charge carriers are spatially confined, and they must be uniform in size, so that the energy levels of the confined charge carriers are well defined.

One could imagine a nanolithography, analogous to the scanned-probe approaches described by Quate [3], to create an array of quantum dots in a film or a pattern on which such dots might form [2,4]. Nature provides a possible alternative approach to creating such arrays that is certainly faster and easier and may produce devices in the not too distant future. Nature has a tendency to organize atoms into regular structures: snowflakes, crystal shapes of all kinds, biological entities. By the manipulation of proper parameters, one can frequently assist, control, or modify this self-organization. For selected systems, and in particular Si/Ge films, the control and manipulation of stress brought about by the different sizes of the component atoms allows one to use nature to grow arrays of 3D quantum dot-like crystallites in a matrix. This review summarizes briefly the present status of stress-mediated growth of quantum dot structures in Si/Ge films.

The growth of Ge and SiGe alloys on Si has attracted considerable interest in recent years as a classical model system for investigations of morphological instabilities of a strained thin film. Potential applications in future electronic and optoelectronic devices, already alluded to above,

have also driven this research. Using SiGe/Si heterojunctions, several types of devices [5] (e.g. heterojunction bipolar transistors, field-effect transistors, and long-wavelength infrared detectors) have already been fabricated, showing greatly enhanced performance over their conventional counterparts [5]. The building blocks of these new devices are quantum wells, in which carriers (electrons and/or holes) are confined in two dimensions. Misfit strain, arising from the 4% lattice mismatch between Ge and Si, is an important part of such quantum well devices, because it determines the maximum thickness of smooth films (wells) and controls band structures (band gap and band-edge offset), mobility of carriers, and quantum confinement potential. Because strain-induced self-organization of structural and morphological features provides an attractive route to nanostructure fabrication, as mentioned above, strain may play an even greater role in lower-dimensional quantum structures than it does in quantum wells [6].

In film growth, manipulation of the growth kinetics and/or surface thermodynamics allows manipulation of the size and shape of growing structures, be they 2D or 3D. A large body of knowledge exists by now on such control [7]. In lattice mismatched films in which the lattice mismatch is not too high (the classic Stranski–Krastanov systems [8], of which Si/Ge is a model example), the formation of 3D crystallites (hereafter referred to as islands) is thermodynamically required early in the growth of a film. Initially these 3D islands are coherent with the substrate lattice (i.e. free of dislocations, but strained). Very well-defined nanocrystals of Ge (or Ge-rich alloy) have been observed in Si/Ge growth, in particular on Si(001) [9–12]. Fig. 1 shows an example. Under proper growth conditions, these 3D islands have nanometer dimensions, thus in principle lending themselves to use as quantum dots. Yet, grown in a single film on a typical Si(001) surface, these 3D islands, despite their beauty and crystalline perfection [9–12], are not sufficiently uniform in size and position to consider their use in any practical applications. In III–V materials the situation is similar; in addition, the 3D islands do not appear to be as well defined (for a recent review emphasizing III–V materials, see Ref. [13]). But additional

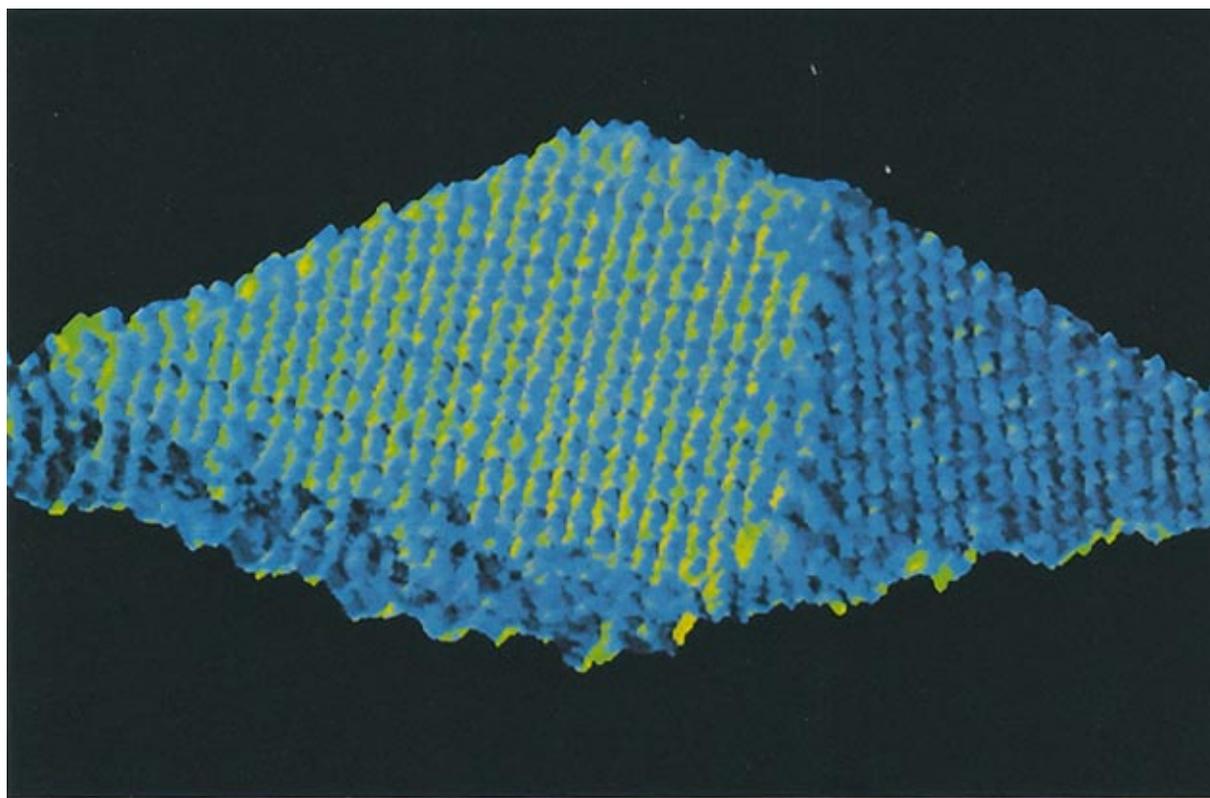


Fig. 1. Scanning tunneling microscope image of a single “hut” island formed during the deposition of pure Ge on Si(001). The height of the island is  $\sim 3$  nm, while the base dimensions are of the order of 20 and 40 nm. All faces are perfect  $\{105\}$  facets. The vertical scale is distorted: the islands actually have an  $11^\circ$  apex angle. After Ref. [10].

levels of control can be imposed. For example, vicinal surfaces, with monatomic-height steps, have been used as templates for the nucleation of 3D islands along rows [14,15], as well as for the growth of “quantum wires” [16]. Such templates significantly improve the positional uniformity of the array of nucleated clusters; however, monatomic-height steps tend to meander and their spacing is often irregular. The greatest improvement in narrowing the size and spatial distributions of the 3D islands comes through the use of multilayers of alternating SiGe and Si films, where the vertical repetition of the films acts as a selective filter for both size and spacing. The result is a three-dimensional array of coherent 3D islands in which the partial vertical correlation of the islands continues to improve the uniformity in size and lateral spacing as more layers get added. In this

review we briefly summarize the state of understanding of strain-induced 3D island formation in single films and multilayers of SiGe on Si(001), with the hope that this summary will give at least a hint of the vast potential of such structures for future nanoelectronic technologies.

## 2. Morphological evolution in strained films

Under given growth conditions (temperature, growth rate, sample orientation and miscut, mode of deposition, etc.), the mode of strain relaxation of a SiGe thin film during growth on Si depends critically on misfit strain, i.e. the thickness of the film and the concentration of Ge in the SiGe alloy. The activation barrier for different modes of strain relaxation scales differently with misfit strain  $\epsilon$ .

Theoretical analyses [17,18] show that the barrier for nucleating a dislocation decreases with increasing strain as  $\epsilon^{-1}$ , while the barrier for nucleating a coherent faceted 3D island decreases with increasing strain as  $\epsilon^{-4}$ . This competition in relaxation modes has been confirmed by growth of SiGe alloy on a nominally perfectly oriented Si(001) surface [18]: at low Ge concentration (below 1%, i.e. low misfit), dislocations form before noticeable surface roughening; at high Ge concentration (high misfit), coherent 3D islands appear before dislocations form. On a vicinal surface, however, the bunching of steps created by the misorientation, which has no nucleation barrier, is expected to be the dominant initial strain relaxation mechanism. Fig. 2 shows three AFM images of a single layer of 2.5 nm thick  $\text{Si}_{1-x}\text{Ge}_x$  alloy film grown on a vicinal Si(001) substrate miscut  $0.25^\circ$  from [001] for three Ge concentrations  $x$ , demonstrating the effect of alloy composition on surface morphology [15]. For the growth conditions used, films with a Ge concen-

tration of  $x=0.2$  (Fig. 2a) are smooth with an r.m.s. roughness  $\sigma=0.15$  nm, which corresponds to that of the substrate. The image exhibits no morphological features, although on a submicrometer-scale image equidistant monatomic steps are visible. For a Ge concentration of  $x=0.45$ , step bunch ripples cover the film surface (Fig. 2b). The r.m.s. value of surface roughness is 0.3 nm. The ripple formation is caused by step bunching [19,20], driven by misfit strain rather than by step flow kinetics [20]. At high Ge concentration ( $x=0.75$ , Fig. 2c), a “granular” structure is superimposed on the ripples, causing a considerable increase in the overall roughness ( $\sigma=0.8$  nm). High-magnification images show that this granular structure is caused by many interlocked 3D “hut” islands [10] (see Fig. 1), coherent with the substrate sitting on top of the ripples. Although the complete phase space for formation of such 3D islands has by no means been established, it is expected, from considerations described later in this review, that ripple and 3D island formation

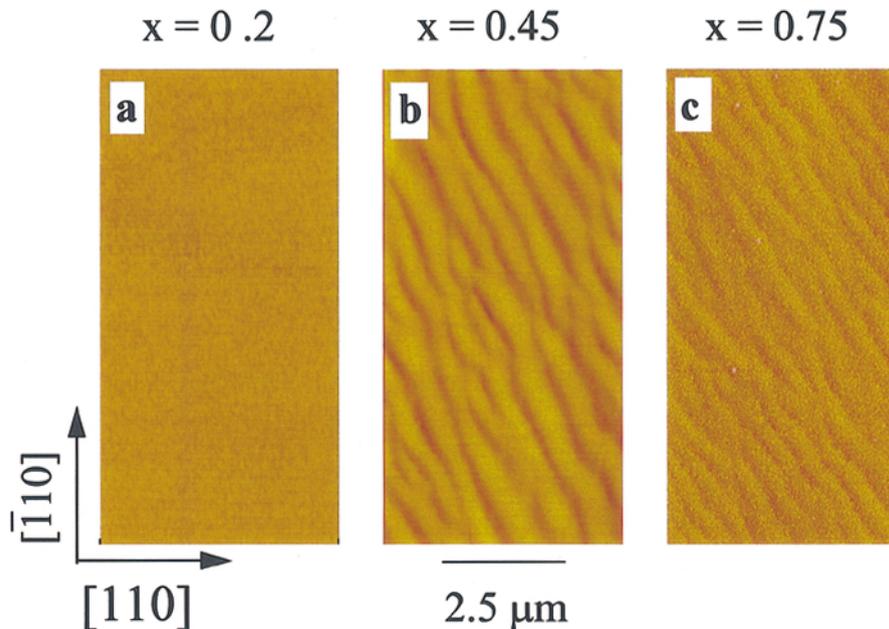


Fig. 2. Atomic force microscope image of three SiGe film growth fronts to illustrate the effect of alloy composition on morphology. All three films have a thickness of 2.5 nm. The scanning direction is [110]. (a) Ge concentration  $x=0.20$ . The film is smooth with an r.m.s. value of surface roughness  $\sigma=0.15$  nm, the same as that for the substrate; (b)  $x=0.45$ . The film displays a clear ripple structure, with  $\sigma=0.35$  nm. (c)  $x=0.75$ . The film displays roughness on top of the ripple structure,  $\sigma=0.8$  nm. The 3D hut islands (Fig. 1) produce this extra roughness. After Ref. [15].

move to lower Ge concentrations for thicker SiGe films and that there exist growth conditions for which the islands can be smaller and others for which the islands will be absent.

The feature of interest in the self-assembly of quantum dot arrays, namely the 3D coherent island formation at high Ge concentrations, is still poorly understood. We know that 3D islanding acts as the major mechanism for strain relaxation. As mentioned earlier, the growth of Ge or SiGe alloy on Si proceeds via the Stranski–Krastanov (SK) mode [8]. Ge initially grows layer by layer, forming a wetting layer several monolayers thick, followed eventually by the formation of dislocated islands that have the Ge lattice constant, with dislocations formed at the Ge/Si or SiGe/Si interface. Between these morphological stages occurs a transition from 2D to 3D growth that is characterized by the formation of 3D faceted islands coherent with the substrate [9–12]. It proceeds via kinetic pathways that are complex and not fully understood.

Because Ge has a lower surface energy than Si, Ge (or SiGe alloy) initially wets the Si substrate. The thickness of the wetting layer is extended beyond what would be expected just on energetic grounds by partial strain relaxation [21] through various surface roughening processes, such as formation of surface vacancies [22] and steps [23], surface reconstruction [24–26], step roughness modification [27], step bunching [19,20], and formation of 2D islands separated by one- to three-monolayer deep “canals” [10,28,29]. For a recent summary of the initial stages of Ge growth on Si(001), see Ref. [30]. In the growth of pure Ge on Si(001), 3D islands start to form when the Ge coverage exceeds 3 ML [10,28]. Initially all the 3D islands are coherent with the substrate (i.e. dislocation-free) [9–12], and have well-defined facets [10,12], giving a prism-like “hut” shape or a four-sided pyramid (see Fig. 1). All four faces are perfect {105} facet planes. The base axes of the huts are always aligned along two orthogonal  $\langle 100 \rangle$  directions.

The four facets in these 3D “hut” islands always appear in a perfect completion. This and the observation that the number density of the hut islands increases rapidly while their size grows

slowly as the Ge coverage is increased indicate that the formation of 3D islands is driven by the low surface free energy of {105} facet planes. Because hut islands form preferentially at lower growth temperatures ( $< 600^\circ\text{C}$ ), and because they transform completely to macroscopic islands after annealing at higher temperatures [10], they are believed to be a metastable intermediate phase facilitating the formation of stable and strain-relaxed macroscopic islands. The kinetics of hut island formation are still unknown. At low growth temperatures the concentration of huts is much higher than that of macroscopic islands, indicating that the huts have a lower formation barrier and are much easier to nucleate. Thus, hut islands define the onset of the transition from 2D to 3D growth and provide an easier kinetic path for formation of the equilibrium rough surface consisting of macroscopic islands. The small formation barrier for huts can result from the low surface energy of their {105} facets (see discussion below). Steps in  $\langle 100 \rangle$  directions, formed in the Ge layers during growth or deliberately introduced by initial substrate miscut, may lower the hut formation barrier by acting as nucleation sites [10,31].

Theories show that, under appropriate conditions, coherent hut islands can be not only energetically more stable than both the strained epitaxial film and dislocated islands, but also kinetically favored over the nucleation of dislocations [18,32,33]. The coherent hut islands allow the partial relaxation of strain by elastic deformation at the expense of introducing extra surface area. In general, the strain relaxation energy is proportional to the volume of the island  $V$ , while the increase in surface free energy scales with the surface area ( $V^{2/3}$ ). Considering an island of trapezoidal shape with a small facet angle, the total free energy of the island can be expressed [18] as  $E = a\Gamma V^{2/3} - b\epsilon^2 V$ , where  $a$  and  $b$  are coefficients related to the island facet angle,  $V$  is the volume of the island,  $\epsilon$  is the misfit, and  $\Gamma$  is the surface energy anisotropy between the normal orientation and the beveled edge. Clearly, for a sufficiently large island (or equivalently a pit), formation of the island (or pit) is energetically favored. Kinetically, the thermal activation barrier is proportional to  $\Gamma^3/\epsilon^{-4}$  for nucleating islands [18],

but scales as  $\epsilon^{-1}$  for nucleating dislocations [17]. Thus, for large misfit (large  $\epsilon$ ), islands are kinetically favored over dislocations, while for small misfit, dislocations are favored [18]. The formation barrier also depends sensitively on surface energy anisotropy  $\Gamma$ . The smaller the value of  $\Gamma$ , the lower the barrier. It is likely that the smaller difference in surface energy between the strained Ge(100) face and (105) faces makes the microscopic hut islands kinetically favored over macroscopic islands, although the macroscopic islands are energetically more stable by incorporating other faces with steeper facet angles to allow more strain relaxation. This picture is consistent with the observation that hut islands form preferentially at lower temperature and transform to large islands after annealing at higher temperature [10].  $\Gamma$  is also generally smaller at higher temperature, and the islands will have multiple facets with less well-defined overall shape and structure [9,10,12]. In order to grow thicker smooth films, it is possible to suppress hut island formation by decreasing  $\epsilon$  (e.g. grading the alloy concentration, as already pointed out), decreasing  $\Gamma$  (e.g. by using surfactants [34]), or growing at low temperature.

Most strained-film growth of Group IV materials, by low-pressure chemical vapor deposition (LPCVD) [35,36], by gas source MBE [37], and by solid source MBE [9–12,38], has focussed either on low concentrations of Ge in the SiGe film, in which the coherent 3D islands are absent or just beginning to develop, or on single films in which the distributions in size and spacing of the coherent 3D islands are broad. Fig. 3a, a higher-resolution AFM image of a region of Fig. 2c, is indicative of the best arrangement of islands that has been found in a single layer [15]. Each square or rectangle is a hut (Fig. 1) with four perfect {105} facets. All base axes are aligned along  $\langle 100 \rangle$ . The mean base dimension is  $\sim 30$  nm, but the distribution is broad. Because the bases align along  $\langle 100 \rangle$ , a substrate deliberately miscut by the proper amount toward  $\langle 100 \rangle$  can act as a template to restrict the distribution of base dimensions [10,15]. Fig. 3b shows an AFM image of hut islands grown on such a template: the long base dimension is now restricted to one  $\langle 100 \rangle$  direction and the other base dimension is narrowly fixed at the mean

separation of the step bunches that form in the initial layers of the SiGe film, giving the appearance of “chains” of 3D hut islands [15]. The  $\langle 100 \rangle$  steps may lower the nucleation barrier for the formation of {105} facets [10,28].

### 3. Self-organization in multilayer films

The discovery that interfacial morphology can replicate across many layers of a thin-film multilayer structure was initially made in the development of thin-film multilayer diffraction gratings for use as soft-X-ray optical elements [39,40]. Quantification of the degree of vertical correlation of a given interface morphology, as well as the correlation length both in the plane of the interfaces and vertically, became possible through the development of X-ray diffuse intensity measurements [40,41], which are specifically sensitive to such interfacial morphology. An application of this diffuse X-ray scattering method led to the discovery of stress-induced step bunching of vicinal Si(001) substrate steps during SiGe growth [19] described earlier and of the vertical replication of step bunches that form in one SiGe alloy layer through subsequent Si spacer layers and alloy layers [19]. Confirmation is provided through AFM measurements of the outer surface morphology of these multilayer films [15], which give a step bunch ripple height and spacing that corresponds to those provided by the X-ray scattering measurements. In addition, the diffuse X-ray scattering measurements allow one to determine that there is a slow lateral migration of the step bunches as they correlate vertically [19], something that is, of course, not possible with AFM.

A replication through multiple layers of the 3D island morphology formed in high Ge concentration SiGe alloy films has been demonstrated with AFM measurements taken at intermediate points in the build-up of the multilayer film (e.g. 2, 10, 20, 40 layer pairs) [42]. Although in general the replication of interface morphology in multilayer films is not well understood, in the model system represented by 3D SiGe islands sitting in or on a matrix of Si, a physically reasonable description of the replication process has been

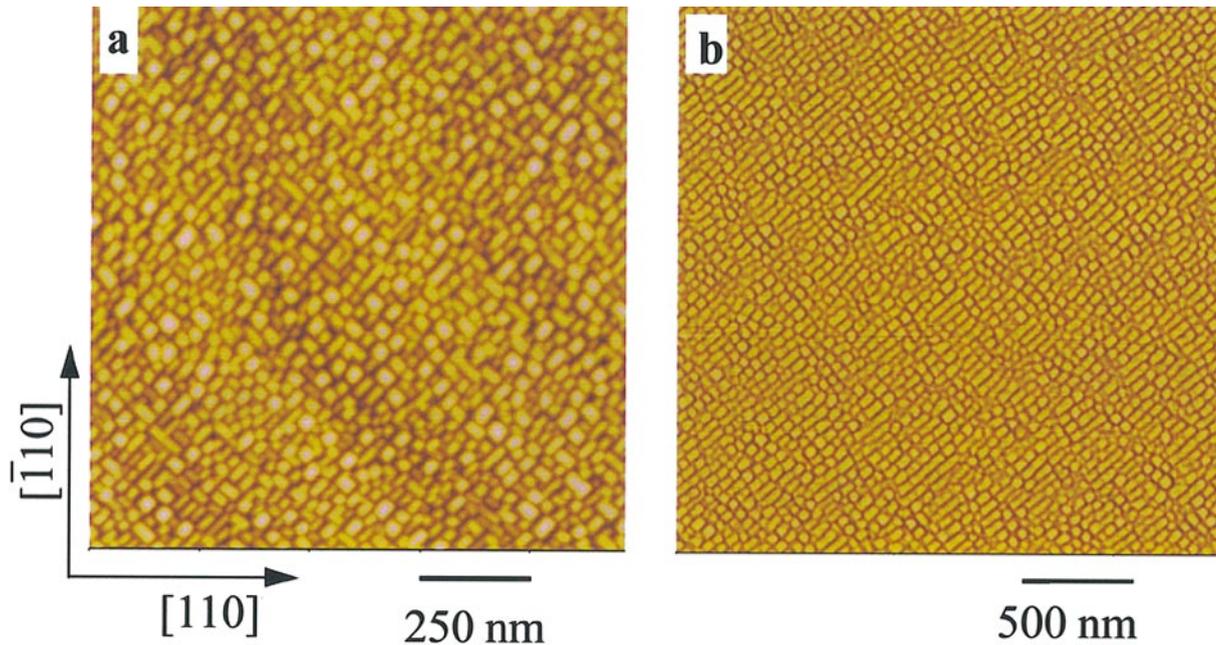


Fig. 3. Gray-scale AFM images of single 2.5 nm thick films of  $\text{Si}_{0.25}\text{Ge}_{0.75}$  deposited on Si(001). (a) Higher-resolution image of a section of Fig. 2c, showing the 3D island distribution. All bases align along  $\langle 100 \rangle$ . The ripples are faintly visible as broad light and dark stripes running from upper left to lower right. (b) Substrate deliberately miscut toward  $[100]$ . All steps and step bunches now have a  $\langle 100 \rangle$  orientation. The 3D islands align in chains along these directions, with their narrow dimension (perpendicular to the steps) controlled by the ripple spacing. After Ref. [15].

presented [43]. We will discuss this model in the context of the detailed observations on the SiGe system.

The growth by solid source MBE of multiple alternate layers of Si and SiGe with a high Ge content has led to the demonstration that stress-driven self-organization can produce a very regular array of crystallites coherent with the substrate and having a narrow size distribution [6,42,43]. The alternate formation of faceted SiGe 3D islands to relieve stress and relatively thin Si spacer layers to mediate the distribution of the strain leads to a progressively more uniform size and spacing of the islands, as well as a vertical organization [42]. The composition of the SiGe alloy and the thickness of the Si layer can be adjusted as controlling parameters. This finding has raised the possibility of increasing the uniformity in size and spacing of the nanoclusters in a controlled way, providing a possible mechanism for obtaining the quality required for electronic and optoelectronic “quantum dot” devices [6].

The self-organization process of the coherent SiGe islands in a SiGe/Si multilayer structure [6,42,43] grown on Si(001) is most remarkable. On a single layer of SiGe alloy, the islands, formed in the shape of prism-like “huts” with  $\{105\}$  facets and coherent with the substrate [42], exhibit a broad distribution in size and shape. Fig. 4a, a three-dimensional representation of the same sample shown in Fig. 3a, vividly illustrates these broad distributions. In a multilayer (consisting of alternating layers of pure Si and SiGe alloy) grown at the proper conditions, the 3D islands organize progressively into uniformly sized crystallites in a square pattern [42]. With increasing bilayer number, the 3D islands become larger, adopt more or less square bases, and arrange preferentially on chains along  $\langle 100 \rangle$  or into a square pattern. The self-organization has been quantified [15,42] by determining the power spectral density of the surface roughness [44]. For a particular set of growth parameters and fixed alloy composition [42], for example, the aspect ratio of island base

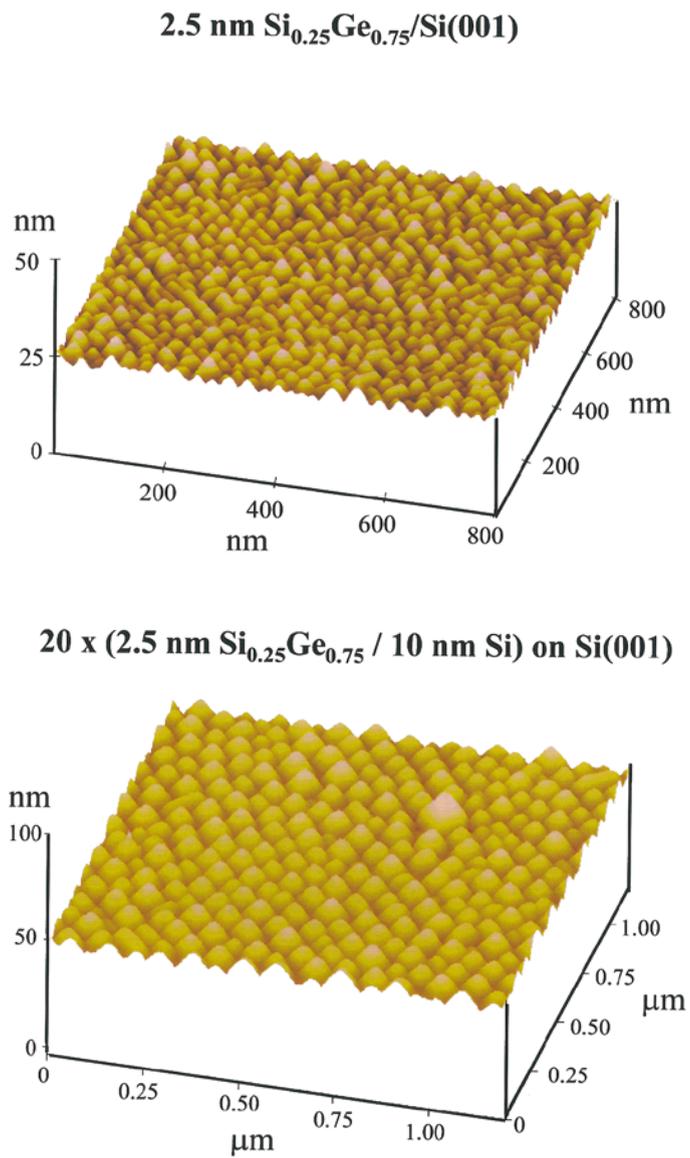


Fig. 4. Three-dimensional AFM images of hut island distributions illustrating the effect of multilayering on narrowing the size and spacing distributions. (a) Single layer, 3D view of the same sample as shown in Fig. 3a. (b) 20 bilayers of 2.5 nm SiGe and 10 nm Si, grown on Si(001). The islands have become square pyramids, very well arranged and uniform in size, and about twice as large as in (a). After Ref. [42].

lengths decreases from 1.5 for the first layer to  $\sim 1.1$  for the top layer of a 20 bilayer film, indicating that the islands are becoming predominantly square-based four-sided pyramids. The distribution in base sizes narrows by a factor of three. The mean length of the island bases approximately doubles to  $\sim 75$  nm. The {105} facets are maintained throughout. Fig. 4b shows an AFM image of the surface morphology of such a film, indicating a quite regular array of pyramids that are small enough to serve as quantum dots.

The self-organization can be controlled by changing the thickness of the Si spacer layers [42]. Pure Si deposited on top of the SiGe alloy layer smooths out the island morphology [42,45,46]. A 10 nm Si layer deposited on top of the island structure shown in Fig. 4b is sufficient to smooth the growth front to nearly what it was for the substrate. Increasing the Si spacer layer thickness to 300 nm while maintaining all other growth conditions leads to a much less pronounced self-organization. Growth without a Si spacer layer eliminates the self-organization [42].

The self-organization can be controlled further by using the polar and azimuthal miscut angles of the substrate to guide and constrain 3D island formation [10,15]. As we showed above, for substrates cut toward  $\langle 100 \rangle$ , the step bunches that form guide 3D island formation already in the first layer, because the islands have  $\langle 100 \rangle$  base directions. The coherent 3D SiGe islands are superimposed on the ripple structure. Because the organization is already better in the first layer, it is expected that a multilayer film will have an even better organization in size and spacing than what is shown in Fig. 4. These experiments have not yet been done, however.

Diffuse X-ray scattering measurements on such multilayer films reveal vertical correlations between 3D islands at different interfaces [19], in addition to the progressively improved uniformity in size and spacing in each consecutive layer determined with AFM. Other measurements [47–50], in both Group IV and Group III–V, show that 3D islands in adjacent alloy layers are located predominantly on top of each other. Apparently, the self-organization is a property of multi-

layer structure, i.e. it is mediated by the spacer layers.

A simple model based on continuum elasticity theory [43] captures the most salient features of the self-organization of coherent 3D islands in a strained multilayer film. The Si spacer layer mediates the strain due to the buried 3D islands, creating a surface strain distribution that is favorable for the formation of regularly spaced and sized islands. The islands are treated as spherical inclusions in an isotropic elastic matrix, and are assumed to be small relative to their lateral and vertical separation. As an island is buried to a depth  $L$ , it produces a surface strain (i.e. the traces of the 2D strain tensor) distribution with a deep minimum (negative) right above the buried island, a maximum (positive) at lateral offsets larger than  $L\sqrt{2}$ , and a smooth drop-off to zero at large distance [43]. Because formation of a 3D island on the surface is most favorable where the local surface strain (due to buried islands) reduces the mismatch between surface and island, it is assumed [43] that the only factor biasing island nucleation at the surface is the strain due to all buried islands, with nucleation at each surface strain minimum (i.e. local minimum in misfit), and nowhere else. (This assumption is appropriate if there is sufficient diffusion on the length scale of the lateral strain variation. Otherwise, reduced misfit of the Stranski–Krastonov wetting layer lowers the local chemical potential, decreasing the nucleation rate [18].) The volume of an island is assumed to be proportional to its Voronoi polygon, i.e. adatoms diffuse to the nearest island during growth. Thus, in general, a buried island causes a preferential nucleation of a new island above it. However, if two islands are very close together, they produce only one island when their surface strain fields overlap, combining two close minima into one larger minimum; if two islands are too far away, an extra new island nucleates between them at the shallow minimum of almost zero strain. In this manner the Si spacer layer acts as a bandpass filter for the morphology. The islands on the surface do more than just mimic, more or less faithfully, the arrangement in the layer below. The island spacings and sizes actually become more regular with each successive layer.

This simple theoretical model [43] omits many important complexities of real systems. For example, the islands are not well separated laterally [42], and they are far from spherical, having pyramidal or prismatic (“hut”) shapes consisting of  $\{105\}$  facets [10,42]. Nevertheless, this theory illustrates the qualitative behavior observed in experiments and captures a very real and simple effect: in a multilayer film, under appropriate growth conditions, 3D islands that form in a strained layer to relieve the strain will self-organize into arrays having progressively better spatial and size distributions, and in fact will form a vertical superlattice with correlation between the islands in the different layers. These growth processes provide a potential approach for fabricating practical quantum dot arrays.

#### 4. Quantum dot superlattices

The suggestion to fabricate quantum dot structures in semiconductor materials using strain-induced clustering was, to our knowledge, first made in 1985 [47]; the necessary structural coherence between substrate and 3D crystallite was confirmed in 1990 for both Group IV [9] and Group III–V [51] systems, and vividly illustrated the same year with scanning tunneling microscopy measurements of Ge “hut” structures grown on Si(001) [10]. Since then, a plethora of work, focussed primarily on III–V materials, has sought to prove, with some success, the possibility of fabricating arrays of regularly-sized 3D clusters that can serve as quantum dots by combining photo- or electroluminescence measurements with growth [13]. The introduction of multilayering not only improves the size and spatial distribution of the islands, making them more suitable as quantum dot arrays, but also introduces a third dimension. Fig. 5 shows schematically an ideal array of quantum dot islands: a uniform small size, a regular spacing laterally, a vertical correlation, and at all levels a structural coherence with the matrix. Such 3D coherent crystallites that are not only laterally but also vertically self-organized may play an important role in “quantum dot” lasers [52]. Experiments are underway at a number of places

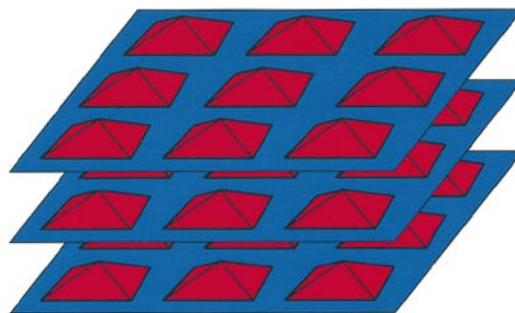


Fig. 5. Schematic diagram of an ideal three-dimensional array of 3D islands buried at regular intervals in an Si matrix and replicated vertically. Such arrays would have potential as quantum dot lattices.

to investigate the growth of such arrays, using both molecular beam epitaxy and chemical vapor deposition. One can expect rapid progress in narrowing the parameter space to provide the necessary control to create real structures like the one shown in Fig. 5.

A number of potential applications exist for such arrays of “quantum dot” structures. The difference in composition of 3D island and matrix provides a band offset that allows the possible trapping of charge. The small size of the 3D islands allows localization of the energy levels of these charge carriers. One can expect uses for such arrays in memories, in wireless “switches” as charge is transferred from one dot to an adjacent one with an electric field, in detector arrays, and in luminescent devices. As one example, consider the luminescent properties of SiGe dots. Neither Si nor Ge is thought of as a good luminescent material, although porous Si and Si/SiO<sub>2</sub> multilayers have shown luminescence [53,54]. We have been able to demonstrate luminescence from SiGe dot arrays that is clearly related to the existence of a regular array of 3D islands with a narrow size distribution [55]. Fig. 6 shows a photoluminescence spectrum. The luminescent line at 806 meV appears only if a highly organized array of  $\{105\}$  faceted pyramids as in Fig. 4b exists. A poorly organized array, as in Fig. 4a, does not produce this line, which is extremely sharp and persists to a temperature of 180 K, far above the temperatures usually associated with luminescence from Group IV materials.

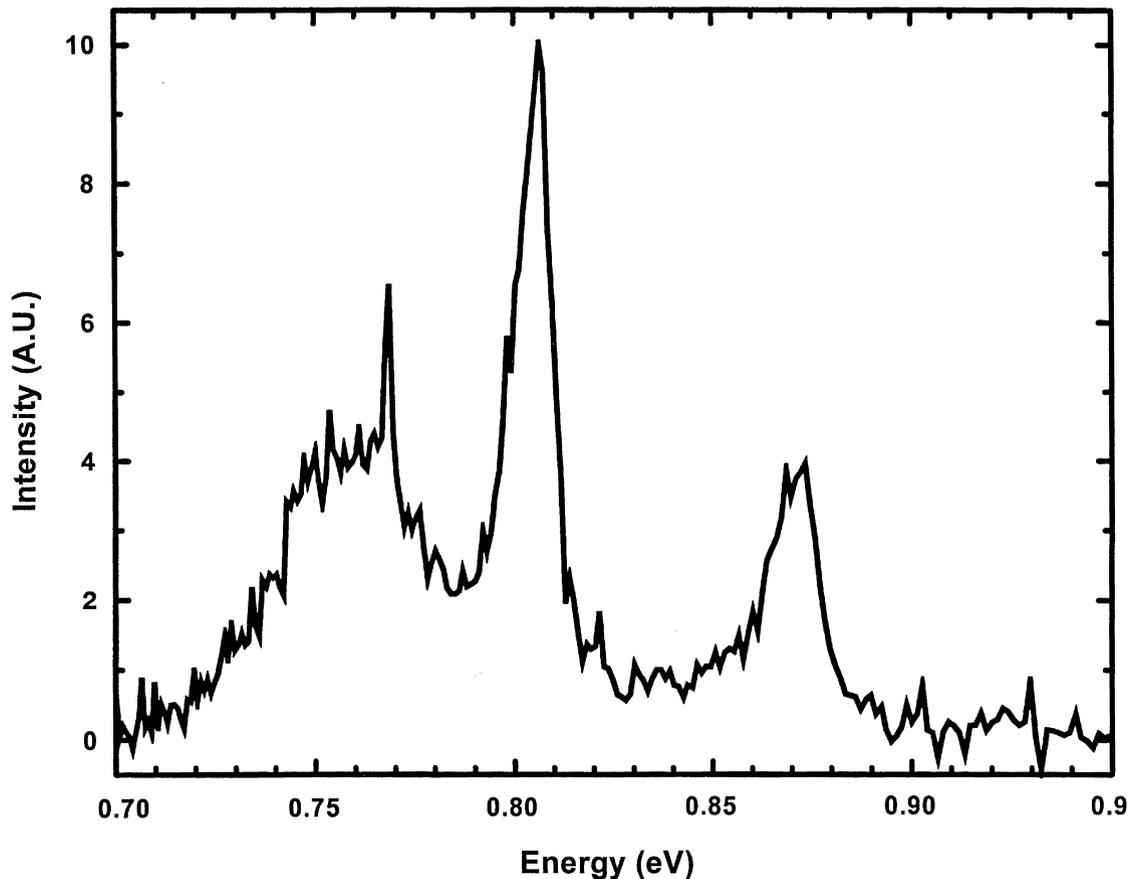


Fig. 6. Photoluminescence spectrum from the SiGe quantum dot array shown in Fig. 4b. The line at 806 meV exists only if a highly organized array of uniformly sized 3D pyramidal islands is present. Spectra from films such as those shown in Fig. 4a do not show this line. After Ref. [55].

## 5. Conclusions

This review has attempted to provide a brief overview of the use of lattice mismatch strain in the growth and self-organization of arrays of 3D crystallites (islands) coherent with the lattice of the matrix material. If such islands are small enough, uniform in size, and uniform in spacing, they are suitable as quantum dots, because of band offsets and charge confinement. The necessary uniformity has not yet been achieved in a single layer; however, the use of multilayering acts like a bandpass filter, producing much better defined size and positional distributions, and carries with it the extra benefit of vertical integration: the growth of

new islands on top of islands existing in layers below. Such structures may open the doors to a host of new electronic and optoelectronic applications. Of course, we still need to find ways to connect the dots with the outside world.

## Acknowledgements

Work described in the article that was performed at the University of Wisconsin-Madison was supported by NSF, ONR, AFOSR, and DAAD (Deutscher Akademischer Austausch Dienst). We acknowledge useful discussions with J. Tersoff and

thank C. Teichert and J. Sullivan for help with the figures.

## References

- [1] C.M. Hu, *Semicond. Int.* June (1994) 105.
- [2] M.S. Montemerlo, J.C. Love, G.J. Opitek, D. Goldhaber-Gordon, J.C. Ellenbogen, Technical Report No. 94W0000044, Mitre Corp., July 1996, in press.
- [3] C.F. Quate, this issue.
- [4] *Science* 275 (1997) 303.
- [5] T.E. Whall, E.H.C. Parker, *J. Mater. Sci.: Mater. Electron.* 6 (1995) 249.
- [6] *Physics Today* 49 (1996) 22.
- [7] Z. Zhang, M.G. Lagally, *Science* 276 (1997) 377; M.G. Lagally, *Jpn. J. Appl. Phys.* 32 (1993) 1493.
- [8] I.N. Stranski, L. von Krastanov, *Akad. Wiss. Lit. Mainz Math.-Natur K1 Iib* 146 (1939) 277.
- [9] D.J. Eaglesham, M. Cerullo, *Phys. Rev. Lett.* 64 (1990) 1943.
- [10] Y.W. Mo, D.E. Savage, B.S. Swartzentruber, M.G. Lagally, *Phys. Rev. Lett.* 65 (1990) 1020; Y.-W. Mo, M.G. Lagally, *J. Cryst. Growth* 111 (1991) 876.
- [11] F.K. LeGoues, M. Copel, R.M. Tromp, *Phys. Rev. B* 42 (1990) 11690.
- [12] F. Iwakaki, M. Tomitori, O. Nishikawa, *Surf. Sci. Lett.* 253 (1991) L411; A.J. Pidduck, D.J. Robbins, A.G. Gullis, W.Y. Leong, A.M. Pitt, *Thin Solid Films*, 222 (1992) 78.
- [13] R. Nötzel, *Semicond. Sci. Technol.* 11 (1996) 1365.
- [14] V. Bressler-Hill et al., *Phys. Rev. B* 50 (1994) 8479; M.S. Miller, H. Weman, C.E. Pryor, M. Krishnamurthy, P.M. Petroff, *Phys. Rev. Lett.* 68 (1992) 3464.
- [15] C. Teichert, Y.H. Phang, L.J. Peticolas, J.C. Bean, M.G. Lagally, in M.C. Tringides (Ed.), *Surfaces Diffusion: Atomistic and Collective Processes*, Proc. NATO Conf., Plenum Press, 1997, in press.
- [16] K. Pond, A.C. Gossard, A. Lorke, P.M. Petroff, *Mater. Sci. Eng. B* 30 (1995) 121.
- [17] F.K. LeGoues, P.M. Mooney, J. Tersoff, *Phys. Rev. Lett.* 71 (1993) 396.
- [18] J. Tersoff, F.K. LeGoues, *Phys. Rev. Lett.* 72 (1994) 3570.
- [19] Y.H. Phang, C. Teichert, M.G. Lagally, T.J. Peticolas, J.C. Bean, E. Kasper, *Phys. Rev. B* 50 (1994) 14435.
- [20] J. Tersoff, Y.H. Phang, Z.Y. Zhang, M.G. Lagally, *Phys. Rev. Lett.* 75 (1995) 2730.
- [21] J. Tersoff, *Phys. Rev. B* 43 (1991) 9377.
- [22] K.C. Pandey, in D.J. Chadi, W.A. Harrison (Eds.), *Proc. Int. Conf. on the Physics of Semiconductors*, Springer, Berlin, 1985.
- [23] O.L. Alerhand, D. Vanderbilt, R.D. Meade, J.D. Joannopoulos, *Phys. Rev. Lett.* 61 (1988) 1973; V.I. Marchenko, A.Ya. Parshin, *Sov. Phys. JETP* 52 (1980) 129.
- [24] J. Tersoff, *Phys. Rev. B* 45 (1992) 8833.
- [25] X. Chen, F. Wu, Z.Y. Zhang, M.G. Lagally, *Phys. Rev. Lett.* 73 (1994) 850.
- [26] F. Liu, M.G. Lagally, *Phys. Rev. Lett.* 76 (1996) 3156.
- [27] F. Wu, X. Chen, Z. Zhang, M.G. Lagally, *Phys. Rev. Lett.* 74 (1995) 574.
- [28] Y.M. Mo, M.G. Lagally, *J. Cryst. Growth* 111 (1991) 876.
- [29] F. Wu, M.G. Lagally, *Phys. Rev. Lett.* 75 (1995) 2534.
- [30] F. Liu, M.G. Lagally, *Chem. Rev.* 97 (1997) 1045.
- [31] K.M. Chen, D.E. Jesson, S.J. Pennycook, M. Mostoller, T. Kaplan, T. Thundat, R.J. Warmack, *Phys. Rev. Lett.* 75 (1995) 1582.
- [32] J. Tersoff, R.M. Tromp, *Phys. Rev. Lett.* 70 (1993) 2782.
- [33] D. Vanderbilt, L.K. Wickham, in C.V. Thompson, J.Y. Tsao, D.J. Srolovitz (Eds.), *Evolution of Thin-Film and Surface Microstructure*, MRS Proc. vol. 202, Materials Research Society, Pittsburgh, PA, 1991, p. 555.
- [34] M. Copel, M.C. Reuter, E. Kaxiras, R.M. Tromp, *Phys. Rev. Lett.* 63 (1989) 632; F.K. LeGoues, M. Copel, R.M. Tromp, *Phys. Rev. Lett.* 63 (1989) 1826.
- [35] R. Apetz, L. Vescan, A. Hartmann, C. Dieker, R. Carius, H. Lüth, *Appl. Phys. Lett.* 66 (1995) 2207.
- [36] E. Palange, G. Capellini, L. Di Gaspare, F. Evangelisti, *Appl. Phys. Lett.* 68 (1996) 2982.
- [37] P. Schittenhelm, M. Gail, J. Brunner, J.F. Nützel, G. Abstreiter, *Appl. Phys. Lett.* 67 (1995) 1292; J. Brunner, J. Nützel, M. Gail, U. Menczgar, G. Abstreiter, *J. Vac. Sci. Technol. B* 11 (1993) 1097.
- [38] H. Sunamura, Y. Shiraki, S. Fukatsu, *Appl. Phys. Lett.* 66 (1995) 953; H. Sunamura, N. Usami, Y. Shiraki, S. Fukatsu, *Appl. Phys. Lett.* 66 (1995) 3024.
- [39] T. Oshino, D. Shindo, M. Hirabayashi, E. Aoyagi, H. Nikaïdo, *Jpn. J. Appl. Phys.* 28 (1989) 1909.
- [40] D.E. Savage, N. Schimke, Y.H. Phang, M.G. Lagally, *J. Appl. Phys.* 71 (1991) 3283 and references cited therein.
- [41] Y.H. Phang, R. Kariotis, D.E. Savage, M.G. Lagally, *J. Appl. Phys.* 72 (1992) 4627; Y.H. Phang, D.E. Savage, R. Kariotis, M.G. Lagally, *J. Appl. Phys.* 74 (1993) 3181; D.E. Savage, Y.H. Phang, J.J. Rownd, J.F. MacKay, M.G. Lagally, *J. Appl. Phys.* 74 (1993) 6158.
- [42] C. Teichert, M.G. Lagally, L.J. Peticolas, J.C. Bean, J. Tersoff, *Phys. Rev. B* 53 (1996) 16334.
- [43] J. Tersoff, C. Teichert, M.G. Lagally, *Phys. Rev. Lett.* 76 (1996) 1675.
- [44] J.M. Elson, H.E. Bennett, J.M. Bennett, *Appl. Opt. Opt. Eng.* 69 (1979) 191; H.N. Yang, G.C. Wang, T.M. Lu, *Diffraction from Rough Surfaces and Dynamic Growth Fronts*, World Scientific, Singapore, 1993, p. 64; W.M. Tong, R.S. Williams, *Ann. Rev. Phys. Chem.* 45 (1994) 401.
- [45] T.S. Kuan, S.S. Iyer, *Appl. Phys. Lett.* 59 (1991) 2242.
- [46] H. Sunamura, Y. Shiraki, S. Fukatsu, *Appl. Phys. Lett.* 66 (1995) 953.
- [47] L. Goldstein, F. Glas, J.Y. Martin, M.N. Charasse, G. LeRoux, *Appl. Phys. Lett.* 47 (1985) 1099.
- [48] J.Y. Yao, T.G. Andersson, G.L. Dunlop, *J. Appl. Phys.* 69 (1991) 2224.
- [49] L. Vescan, W. Jäger, C. Dieker, K. Schmidt, A. Hartman, H. Lüth, in M.F. Chisholm et al. (Eds.), *Mechanisms of*

- Heteroepitaxial Growth, MRS Symp. Proc. No. 263, Materials Research Society, Pittsburgh, PA, 1992, p. 23.
- [50] Q. Xie, A. Madhukar, P. Chen, N.P. Kobayashi, *Phys. Rev. Lett.* 75 (1995) 2542.
- [51] S. Guha, A. Madhukar, K.C. Rajkumar, *Appl. Phys. Lett.* 57 (1990) 2110.
- [52] S. Fafard, K. Kinzer, S. Raymond, M. Dion, J. McCaffrey, Y. Feng, S. Charbonneau, *Science* 274 (1996) 1350.
- [53] R.T. Collins, P.M. Fauchet, M.A. Tischler, *Physics Today* 50 (1997) 24.
- [54] Z.H. Lu, D.J. Lockwood, J.-M. Baribeau, *Nature* 378 (1995) 258.
- [55] J.-L. Lin, C. Teichert, J.C. Bean, L. McCaughan, M.G. Lagally, *Phys. Rev. Rapid Comm.*, submitted.