Self-Organized Replication of 3D Coherent Island Size and Shape in Multilayer Heteroepitaxial Films

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A model is proposed to elucidate the evolution of the morphology of strained 3D islands in multilayer heteroepitaxial films. The model explains the experimental observation that islands grown in successive layers not only replicate, forming individual island columns, but self-organize to reach a common size and shape, independent of their initial density. [S0031-9007(99)08750-5]

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Advances in modern science and technology continue to make electronic and optoelectronic devices faster and smaller. As its size approaches the nanometer scale, a crystal exhibits electronic and optoelectronic properties different from those of the bulk, leading to potential new applications, such as "quantum dot" (QD) structures in which nanometer-size clusters of one material are embedded into the matrix of a second one.

The growth of three-dimensional (3D) islands in strained films via the Stranski-Krastanov (SK) growth mode provides a possible route to the fabrication of QDs. After forming a thin wetting layer, strained layers may spontaneously form 3D islands to relieve the misfit strain. Under appropriate growth conditions, these islands achieve nanometer size and are coherent with the substrate lattice (i.e., free of dislocations) [1] but, in general, are not uniform in size, shape, or spacing. However, uniformity in size and shape is a prerequisite for their potential use in QD electronic or optoelectronic devices, for which precise knowledge of the electronic energy levels is essential. It has been discovered that multilayering improves island uniformity: With increasing number of bilayers [the spacer layer (same as the substrate) plus the SK layer], the 3D islands become more uniform in size, shape, and spacing [2–11]. For sparse initial island arrays, typical for III-V systems, individual vertical columns of islands are formed with islands in each column converging to a stable size and shape [2–7]. For dense initial island arrays, typical for the SiGe system, multilayering leads both to uniform island size and shape and to uniform spacing [8–11].

Theoretical models, focusing on island nucleation, have been proposed to explain the vertical self-organization [2,8]. The 3D islands buried by the spacer layer produce a tensile region above themselves (assuming the original SK growth occurs for a compressed layer, as in all systems so far investigated). When the next SK layer is deposited, this tensile region induces a preferential nucleation of new 3D islands above the buried ones by a strain-directed diffusion [2] and/or by lowering the overall lattice misfit [2,8]. The repetition of layers creates columns of 3D islands. A model [8] also shows that the interaction of the localized strain fields induced by the buried islands at the spacer layer surface can in effect improve the island size uniformity, but requires that uniform spacing be a mandatory condition for achieving uniform size. Uniform spacing is achieved in subsequently deposited SK layers through the annihilation of small islands in initially dense arrays and the creation of new islands in initially sparse arrays. Experiments, on the other hand, show that islands can self-organize in individual columns with improved uniformity in size and shape without a need for uniform spacing. Such an effect is most noticeable in initially sparse arrays, e.g., the III-V systems, for which size and shape uniformity is achieved without a need for the creation of new islands to reach uniform spacing [2–7]. There is so far no explanation for this behavior.

In this Letter, we demonstrate a new model of the vertical self-organization of coherently strained 3D islands in a multilayer heteroepitaxial film. The model elucidates the effect of the strain field induced by buried islands at the surface of the embedding-matrix layer not only on the nucleation of newly formed islands but also on their growth (size and shape). Finite island size and anisotropic island base shape are introduced, rather than a "point" [8]. As we shall show, size and shape of the "inclusion" change the strain distribution at the embedding-layer surface enough so that dramatically different self-organization effects occur.

The model shows that, for initially sparse island arrays, replication forms individual columns of islands as more bilayers are added; the evolving strain field causes the islands in successive layers in each column either to grow or to shrink in size, depending on whether the initial island was larger or smaller than some characteristic size, and gradually to converge toward that size. Island base shape transformations from anisotropic to isotropic are also produced by the model. For initially dense island arrays, lateral coalescence of close-lying islands leads to uniform island spacing.

Compressively strained 3D islands buried in the embedding layer produce a nonuniform surface strain field: The regions right above the islands become expanded (tensile strain) while the region between islands becomes compressed. The nature of this tensile region depends on

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the size and shape of the buried island. Figure 1 shows schematically in 2D the surface strain (i.e., the trace of the strain tensor) caused by a rectangular island (corresponds to a disk in 3D) [12] buried at a depth L and centered at the lateral position $x = 0$, in an isotropic elastic medium. For a rectangular island whose height H is much smaller than the embedding layer thickness L, the strain at the surface of the embedding layer is expressed as

$$\epsilon(x) = -\epsilon(L)[\zeta(1 + \zeta^2)^{-3/2}(2 + \zeta^2) - \eta(1 + \eta^2)^{-3/2}(2 + \eta^2)], \quad (1)$$

where $\epsilon(L) = CH/L^2$, $\zeta = (x + W)/L$, and $\eta = (x - W)/L$; C is a coefficient related to misfit and elastic constants and W equals one-half of the island width. A sign convention is chosen such that ϵ, like the associated interaction energy, is negative in the tensile region [8]. Numerical solutions show that the extent of tensile strain $\{2x_0$, defined by $[\epsilon(\pm x_0) = 0]\}$ increases with the increasing size of the buried island $(2W)$ as a power law with an exponent of $\frac{1}{2}$, and the ratio of x_0/W decreases with increasing island size with an asymptotic value of one as W approaches infinity.

We simulate the growth of a multilayer of islands using a deterministic model. We start with the worst-case scenario, an ensemble of islands in the first layer with a random distribution of positions and sizes. (Other mechanisms may exist to improve the island uniformity in the first layer [13].) In all subsequent layers, we assume that islands preferentially nucleate in the tensile regions only and they will grow larger when they are sitting inside a larger tensile region, because doing so will lower the strain energy by minimizing the overall misfit between the island and the spacer layer. The surface-strain-dependent island size distribution in a given layer can be

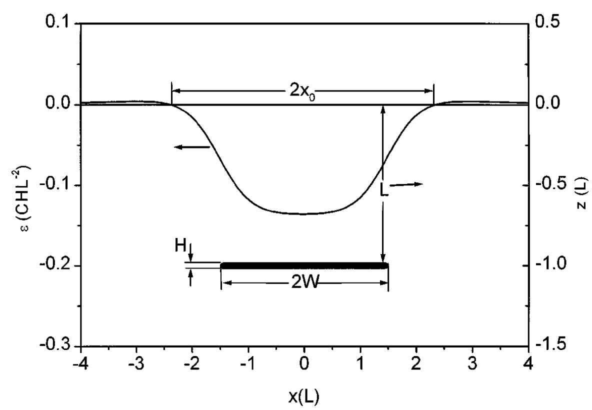

FIG. 1. Strain distribution (ϵ, left y axis, in units of CHL^{-2}) at the surface of an embedding layer of thickness L, induced by a compressively strained coherent rectangular island of height H buried at a depth $z = L$ (right y axis). C measures misfit and elastic constants. The area of negative tensile strain above the island $(2x_0)$ increases with increasing island base size $(2W)$ as a power law with an exponent of $\frac{1}{2}$. Regions of weak compressive strain lie outside $|x_0|$.

defined as

$$B_i = (B_t/x_{0t})x_{0i}, \quad (2)$$

where $(2B_i)$ is the base size of the ith nucleated island and $2B_t = \Sigma(2B_i)$ is the sum of base sizes of all nucleated islands, $2x_{0i}$ is the extent of the ith tensile region, and $2x_{0t} = \Sigma(2x_{0i})$ is the total tensile "area" produced by all of the buried islands. In the simulation, an island first forms in a triangle shape (a 2D pyramid) with a base size of $(2B_i)$ and a fixed angle. As it is buried, it is transformed into a rectangular shape with conserved volume and a fixed height-to-base ratio [12]. The latter is used for the calculation of the strain field seen by the next islanding layer.

The value of the ratio of the size of a nucleated island and the area of the tensile region in which the island is sitting, B_i/x_{0i}, defines two different regimes of growth, with respect to the convergence of island size. For $(B_i/x_{0i}) > 1$, the newly formed island is always larger than the buried island below, because the tensile region (x_{0i}) is always larger than the size of the buried island (W_i). Consequently, in this regime of growth, the size of the islands will increase indefinitely in a multilayer film. In real growth, however, an island cannot grow forever, because its size is limited at least by the nominal thickness of the film; neither can an island grow beyond the point at which it meets neighboring islands. For the other regime, $(B_i/x_{0i}) < 1$, the newly formed island may be either larger or smaller than the buried island below, leading to more complex behavior, including vertical self-organization. In the following, we will focus on this growth regime.

In a single-layer film, in general, the average island size is larger and the island spacing is smaller in a low-misfit system than in a high-misfit system. For example, the size of SiGe islands grown on Si(001) decreases with increasing Ge concentration (maximum 4% lattice mismatch) and they are generally larger than InAs islands grown on GaAs(001) (7% lattice mismatch). Also, the SiGe islands form a dense array, with their bases in close contact [9], while the InAs islands form a sparse array. To account for all situations, we apply our model to two extreme cases: a sparse island array, with all islands far away from each other and very little overlap between their strain fields, and a dense array, with all islands having their bases in close contact and their strain fields strongly overlapping.

Figure 2(a) shows typical examples of the growth of a multilayer film for an initially sparse island array. The spacer layer thickness is chosen to be $L = 12$ and base-area-to-tensile-strain-area ratio is kept constant [14] at $(B_i/x_{0i}) = 0.75$. The three islands in the first layer have a starting base size of $4.5L$, $10L$, and $2.3L$, respectively. For such a sparse island array, the strain distribution produced at the embedding-layer surface consists of a deep negative minimum at each island position, so that a new island nucleates on top of each buried island in the next layer [15], leading to the formation of vertical columns of islands as

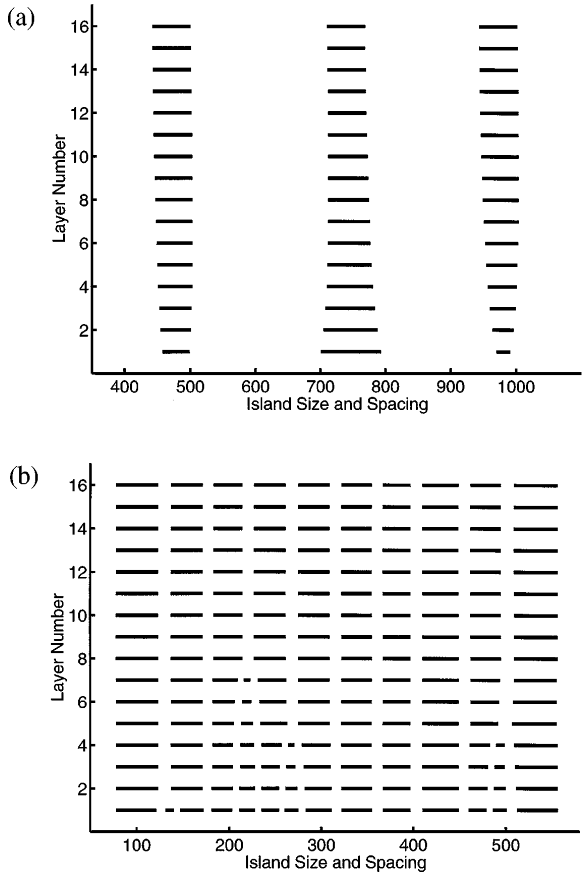

FIG. 2. Evolution of island size in a multilayer film of 16 bilayers simulated for island arrays for embedding layer thickness $L = 12$ and island-size-to-tensile-region ratio $(B_i/x_{0i}) = 0.75$. The x axis marks the island position and size, and the y axis marks the bilayer number of the film. Lengths of horizontal lines represent the size of island bases; island heights (not shown) scale with the base sizes. (a) A sparse island array. Three typical cases are selected for illustration. Islands all reach the same size. (b) A dense island array. Islands achieve uniform spacing, in addition to converging in size.

more bilayers are added. Most remarkably, the island sizes in each column either increase or decrease and gradually converge toward the same final size and then stay fixed. The stable island size is defined by spacer layer thickness and the base-area-to-tensile-strain-area ratio; it is $6.4L$ with the parameters used in Fig. 2.

The formation of island columns and the convergence of island sizes in different columns have been observed in both III-V [2–7] and group-IV multilayer films [8–11] and can be explained as follows. Islands nucleate and grow with a random size distribution on the initial strain-free surface. New islands in subsequent layers adopt a size distribution in accordance with the relative lateral extents of the tensile regions they are sitting on. A small buried island produces a tensile region above itself, much larger in lateral extent than its own size; this region collects more atoms, and the new islands above it grow rapidly in the first few subsequent layers [right column in Fig. 2(a)]. An initially larger island produces a tensile region whose extent is more like its own size; islands above it in subsequent layers grow more slow or may even shrink [middle column in Fig. 2(a)] when $(B_i/x_{0i}) < 1$. As the distribution of lateral extents of tensile regions becomes more and more uniform in each successive layer, islands evolve to a common stable size.

For an initially dense island array, lateral coalescence of close-lying islands within the same layer provides a second mechanism for improving island size uniformity (in addition to vertical replication of islands in successive layers). (Island coalescence can, of course, also occur for initially sparse islands if two islands nucleate closely together. An island that will, as a consequence, be too large will reduce its size in subsequent layers [4,10].) Coalescence and vertical size convergence lead to both uniform island size and uniform spacing. Figure 2(b) shows an example of the evolution of a dense island array in a multilayer film, for $L = 12$ and $(B_i/x_{0i}) = 0.75$. The starting islands are created randomly with a mean size of $3.0L$ and a standard deviation of $1.1L$, with their bases in contact. When two islands are sufficiently close, an x_0 point (where strain equals zero) may not exist between them. The tensile region created by each island is then defined by the local maximum in the negative strain curve (i.e., the point of least strain) between them as the boundary. The evolution of larger islands is dominated by vertical replication: These islands form columns and gradually converge in size, as for sparse islands. Smaller islands are eliminated and/or combined into larger islands by coalescence. The coalescence causes a decrease of areal island density and an increase of average island volume, because islands maintain their shape (i.e., facet angle) upon coalescence. For example, the average island size in Fig. 2(b) increases from $3.0L$ to $4.3L$.

After coalescence events cease, islands continue to change their sizes slightly and soon become stabilized in individual columns. However, the converged island size distribution for an initially dense island array [Fig. 2(b)] is not as uniform as that for an initially sparse island array [Fig. 2(a)]. When islands form in close proximity, their strain fields overlap strongly with each other. Consequently, islands in neighboring columns keep each other from changing, locking into different sizes.

Simulations for 3D islands demonstrate that these mechanisms also produce a shape transformation of vertically replicated islands in a multilayer film, independent of the initial density of islands. As an island with anisotropic base shape is buried, it produces a tensile region that is anisotropic. However, the aspect ratio of the tensile area, and hence the aspect ratio of the nucleated-island base, is smaller than the original aspect ratio of the buried-island base, leading to a more isotropic shape in the next SK layer. Figure 3 shows an example of the evolution of 3D island shape in a single-island column. The initial island, a rectangular disk, has a base aspect ratio of 4:1, which

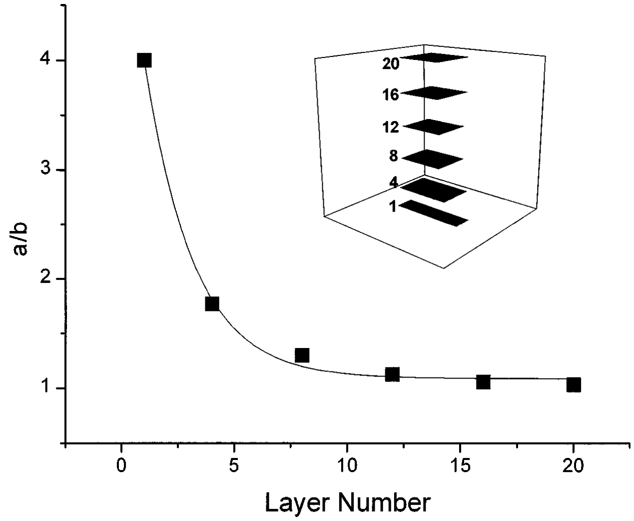

FIG. 3. Evolution of a 3D island shape in a multilayer film of 20 bilayers simulated for a single island column for embedding layer thickness $L = 12$ and island-size-to-tensile-region ratio $(B_i/x_{0i}) = 0.75$. The initial island has a rectangular base with an aspect ratio a:b of 4:1. The aspect ratio of the island decreases with an increasing number of bilayers; the squares are simulated values and the solid line is a spline fit. The inset shows a schematic view of the island shape evolution.

decreases rapidly toward 1:1 with an increasing number of bilayers. The improvement of island shape uniformity in a multilayer film has been observed in both III-V [7] and group-IV systems [9].

In summary, we have demonstrated a mechanism for the self-organized replication of the size and shape of strained islands in a multilayer film consisting of alternating strained-3D-island layers and embedding matrix layers. As islands of the SK layer material nucleate and grow on a strain-modulated surface of the embedding matrix layer, their size distribution and shape uniformity are controlled by the strain distribution, which in turn depends on the size and shape of the buried islands. The nature of these interdependent relations leads to a self-organized evolution of islands and their strain fields. Simulations show that, for an initially sparse island array, self-organization, dominated by vertical replication, leads to the formation of island columns with both island size and shape converging toward common stable ones in each column. For an initially dense island array, self-organization, now driven by both replication and coalescence, creates arrays of islands with uniform spacing as well as uniform size and shape.

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[1] D. J. Eaglesham and M. Cerullo, Phys. Rev. Lett. **64**, 1943 (1990); Y. W. Mo, D. E. Savage, B. S. Swartzentruber, and M. G. Lagally, Phys. Rev. Lett. **65**, 1020 (1990).
[2] Q. Xie, A. Madhukar, P. Chen, and N. P. Kobayashi, Phys. Rev. Lett. **75**, 2542 (1995).
[3] M. S. Miller, J. Malm, M. Pistol, S. Jeppesen, B. Kowalski, K. Georgsson, and L. Samuelson, J. Appl. Phys. **80**, 3360 (1996).
[4] G. S. Solomon, J. A. Trezza, A. F. Marshall, and J. S. Harris, Phys. Rev. Lett. **76**, 952 (1996); G. S. Solomon, S. Komarov, J. S. Harris, Jr., and Y. Yamamoto, J. Cryst. Growth **175-176**, 707 (1997).
[5] Y. Nakata, Y. Sugiyama, T. Futatsugi, and N. Yokoyama, J. Cryst. Growth **175-176**, 713 (1997).
[6] M. K. Zundel, P. Specht, K. Eberl, N. Y. Jin-Phillipp, and F. Phillipp, Appl. Phys. Lett. **71**, 2972 (1997).
[7] R. Heitz, A. Kalburg, Q. Xie, M. Grundmann, P. Chen, A. Hoffmann, A. Madhukar, and D. Bimberg, Phys. Rev. B **57**, 9050 (1998).
[8] J. Tersoff, C. Teichert, and M. G. Lagally, Phys. Rev. Lett. **76**, 1675 (1996).
[9] C. Teichert, M. G. Lagally, L. J. Peticolas, J. C. Bean, and J. Tersoff, Phys. Rev. B **53**, 16 334 (1996).
[10] E. Mateeva, P. Sutter, J. C. Bean, and M. G. Lagally, Appl. Phys. Lett. **71**, 3233 (1997).
[11] P. Schittenhelm, G. Abstreiter, A. Darhuber, G. Bauer, P. Werner, and A. Kosogov, Thin Solid Films **294**, 291 (1997).
[12] Different choices of island shape (e.g., a triangle or a pyramid in 3D) lead to qualitatively the same results, as long as the influence of the finite size and base shape anisotropy of islands on the strain field are included.
[13] V. A. Schukin, N. N. Ledentsov, P. S. Kop'ev, and D. Bimberg, Phys. Rev. Lett. **75**, 2968 (1995); I. Daruka and A. Barabasi, Phys. Rev. Lett. **79**, 3708 (1997).
[14] The assumption of a constant island-size-to-tensile-area ratio B_i/x_{0i} implies that the total island volume will change with an increasing number of bilayers. If the total island volume x_{0t} were kept constant, then B_i/x_{0i} would vary at each bilayer and a similar self-organization process could occur. However, experiments [4,8,9] indicate a changing total island volume with an increasing number of bilayers.
[15] If the initial island density is very low, additional islands may nucleate at random between buried islands. Our model then applies to the stage of growth after nucleation events saturate.