Self-Organization in Growth of Quantum Dot Superlattices

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We investigate the growth of multilayer arrays of coherently strained islands, which may serve as "quantum dots" in electronic devices. A simple model reproduces the observed vertical correlation between islands in successive layers. However, the arrangement of islands is not simply repeated from layer to layer. Instead, the island size and spacing grow progressively more uniform. In effect, the structure "self-organizes" into a more regular three-dimensional arrangement, providing a possible route to obtain the size uniformity needed for electronic applications of quantum dot arrays.

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"Quantum well" structures, in which electrons are confined on a nanometer scale in one direction, are already used in semiconductor devices. Much effort has been devoted to creating analogous structures in which electrons are confined in two directions ("quantum wires"), or in all three directions ("quantum dots" or "quantum boxes"). However, the controlled fabrication of such nanostructures is a daunting challenge. There has therefore been intense interest in nanostructures that could, in effect, fabricate themselves. In particular, strained islands form spontaneously ("self-assemble") in growth of InAs on GaAs or SiGe on Si, because islanding allows some elastic relaxation of the strain [1]. These islands are three dimensional, epitaxial, and coherently strained (i.e., no dislocations); throughout this paper, the term "island" refers only to these. Self-assembly offers an attractive route to the fabrication of quantum dot arrays, especially since the islands can be unexpectedly uniform in size [2–5]. Still, they are not sufficiently uniform for most practical applications, and no systematic method for improving uniformity is known.

Another requirement for technological applications is a high spatial density of dots. Toward this end, several groups have grown quantum-dot superlattices. For example, alternating growth of GaAs and strained InAs yields layers of InAs dots embedded in GaAs. A fascinating feature of such structures is that the dots (i.e., buried islands) in successive layers are spatially correlated [6–10]. At the surface, new islands tend to nucleate directly above buried islands. This spatial bias arises from the strain at the surface due to buried strained islands [6–9].

Here, we examine the spatial arrangement of quantum dots in such multilayer structures, with a simple and relatively generic model. We find that the dots can exhibit dramatic self-organization. Islands on the surface do more than just mimic, more or less faithfully, the arrangement in the layers below. The island sizes and spacings actually become more regular with each successive layer. This phenomenon may have great practical importance, providing a mechanism for obtaining the uniform island sizes required in electronic devices. Moreover, the size and spacing of islands can be directly controlled via the thickness of the layers. We show for comparison the experimental data that motivated this analysis. They provide clear evidence that such self-organization may be achieved in practice.

The actual process of island nucleation is complex, especially for the strained islands considered here [11]. Formation of an island on the surface is most favorable where the strain at the surface (due to buried strained islands) reduces the mismatch between surface and island. Because the nucleation rate is an exponential function of the nucleation barrier, which in turn depends sensitively upon the strain [11], we assume that islands nucleate wherever this strain gives a local minimum in the misfit. (This assumption is appropriate if there is significant diffusion on the length scale of the lateral strain variation. Otherwise, reduced misfit of the Stranski-Krastonow wetting layer lowers the local chemical potential, decreasing the nucleation rate [11].)

Other effects may also contribute to the behavior, leading to qualitatively similar conclusions. For example, Xie *et al.* [9] treated the island distribution by considering the effect of strain on diffusion, rather than focusing on the nucleation event. Also, while the Si or GaAs surface tends to planarize again over each layer of islands [6,8,12], small surface undulations due to the buried islands could also bias subsequent island formation at the surface. A detailed analysis of the possible contributing mechanisms would require specifying parameters that depend strongly on the system, temperature, and flux, and that are in most cases poorly known. For example, the nucleation rate is extremely sensitive to the island shape and to the surface energy anisotropy [11].

Therefore, to understand the growth in the most general manner possible, rather than focus on a specific single

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system or mechanism, we consider the following generic model. The quantum dots are treated as spherical inclusions in an isotropic elastic matrix, and are assumed to be small relative to their lateral and vertical separation. The only factor biasing nucleation at the surface is the strain due to these buried islands, with nucleation occurring at each local minimum in misfit, and nowhere else.

For brevity, we will refer henceforth to Ge islands on Si. This should be understood as referring equally to SiGe alloys on Si, or InAs (or InGaAs) on GaAs, or any similar system. Consider a system with small coherently strained Ge islands on a flat Si surface. These islands are then buried under a thickness L of additional Si. The new surface will also be nearly flat, but with some local strain due to the buried islands. (The system is assumed to be entirely free of dislocations.) The surface strain (i.e., the trace of the two-dimensional strain tensor) may be calculated using continuum elasticity theory, following Maradudin and Wallis [13]. An island buried at a depth L and lateral position $x = 0$ leads to a surface strain ϵ at lateral position x, where

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

The resulting strain is shown in Fig. 1 for an illustrative arrangement of buried islands. For a single buried island, the favored nucleation position is directly above, and we choose a sign convention where ϵ, like the associated interaction energy, is negative there. Nucleation is disfavored at lateral offsets than $L\sqrt{2}$, with the strain dropping smoothly to zero at large distance.

The coefficient C in Eq. (1) is proportional to the volume of the buried island and to the misfit, and also involves the elastic constants [13]. We assume that, during growth, adatoms diffuse to the nearest island, so the island volume is proportional to the area of its Voronoi polygon (the region closer to that island than to any other).

Within this highly simplified model, growth is a deterministic process. We first specify a distribution of Ge islands on the Si substrate. Then these are buried under a depth L of Si. The strain ϵ at the surface of this Si spacer layer is calculated by summing Eq. (1) over all the buried islands. Upon deposition of more Ge, new islands are assumed to nucleate at each minimum of ϵ, with volumes proportional to the areas of the corresponding Voronoi polygons. This sequence is then repeated for each successive layer.

An important point to note, and a central reason for choosing such a simplified model, is that the distribution of islands then depends only on the initial arrangement (sizes and spacings), scaled by the layer thickness L. Moreover, as seen below, after sufficiently many layers the arrangement becomes insensitive even to the initial conditions. To facilitate visual inspection of our results, we reduce the problem to two dimensions, confining islands to a line of the surface. We have also carried out some simulations in three dimensions [14], discussed briefly below, to check that the behavior is similar.

The results of this deterministic model are shown in Figs. 2 and 3. We have used periodic boundary conditions, with a cell of $300L$. Figure 2 shows the island distribution in successive layers, for two sets of initial conditions. In both cases the initial positions of islands are random (a worst-case assumption), with volumes determined as for later layers. But in Fig. 2(a) the initial average spacing is much smaller than the layer thickness, while it is much larger in Fig. 2(b). The vertical correlations discussed by several authors [6–10] are clearly seen in all layers whenever the islands are not so close together that their strain fields strongly overlap. (These vertical correlations

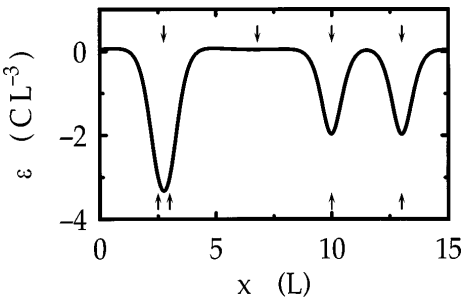

FIG. 1. Surface strain ϵ vs lateral displacement x (in units of spacer thickness L), for four islands buried at depth L, from Eq. (1). Arrows at bottom indicate lateral positions of buried islands. Arrows at top indicate minima in ϵ, i.e., favored positions for subsequent nucleation. (The very shallow minimum in the middle is discussed in the text.)

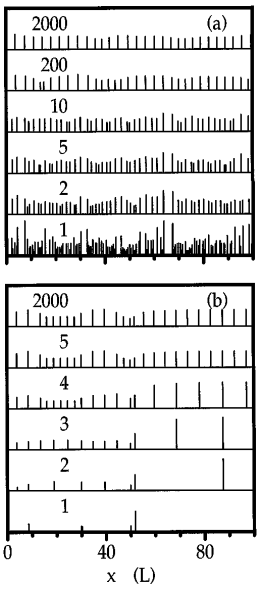

FIG. 2. Calculated island positions and sizes in selected successive layers. Layer numbers are indicated. Heights of vertical lines represent island volumes, relative to average for that layer. Sequences begin with (a) closely spaced islands and (b) widely spaced islands. The periodic cell size is $300L$; for clarity, only a third of the cell is shown.

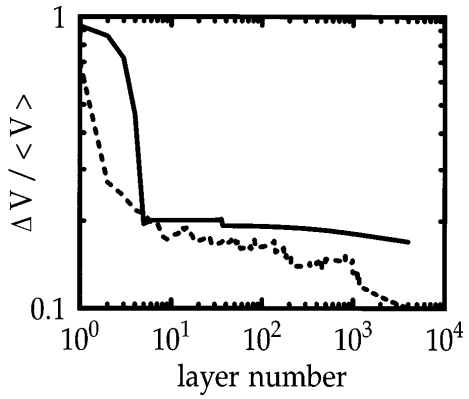

FIG. 3. Root-mean-square variation ΔV of island size within the layer, relative to average volume $\langle V \rangle$ for that layer, vs layer number for same systems as in Fig. 2. Dotted and solid lines correspond, respectively, to dense and sparse initial island densities.

are best viewed by tilting the figure to foreshorten the vertical axis.)

An even more striking and important point is that, for successive layers, the island size and spacing becomes progressively more uniform. Regardless of the initial condition, after many layers the lateral island spacing is about $3.5L$. We have also verified that an intermediate initial spacing also leads to a similar final configuration. Thus, by controlling the Si layer thickness, one can directly control the final island spacing. The total volume of material in the islands can be controlled independently by varying the nominal thickness of each Ge layer, so the nominal thicknesses of the alternating layers in the superlattice provide control over all relevant parameters within this model.

By referring to Fig. 1, it is easy to understand the origin of this behavior. For initial island spacing much greater than $3.5L$, new islands will nucleate not only above existing islands, but also in the space in between. Conversely, for islands that are closely spaced, there will not be a distinct minimum in ϵ above each island. Two or more islands that are very close will be replaced by a single island in the next layer. Thus the nucleation positions in Fig. 1 are much more uniformly spaced than the initial islands. In general, there is a "thinning" of closely spaced islands, and a filling in of gaps, with the characteristic length scale determined by the Si layer thickness L. The spacer, in effect, acts like a band pass filter for the spatial frequency, with successive applications of this filter leading to a progressively more uniform island spacing and size distribution.

In general, the complexities of real systems may introduce competing length scales, e.g., the surface diffusion length. Moreover, it can happen that islands of different sizes have different shapes, or that the largest islands incorporate dislocations. Thus the present model may correctly describe some regimes of growth, while becoming inapplicable under other possible conditions. In particular, the behavior for initially sparse islands is doubtless less generic (i.e., more sensitive to assumptions of the model) than for initially dense islands. The strain minimum between distant islands is weak, so nucleation in this region will be more sensitive to other effects. Moreover, in three dimensions, these weak minima may disappear altogether, so that a more detailed model is required to describe the filling in of gaps by random nucleation. Fortunately, the diffusion length which controls such nucleation may be manipulated experimentally via the temperature and flux to find the optimal growth regime. In any case, for densely spaced initial islands, the results of three-dimensional simulations [14] appear similar to those reported here. The initial island density can be controlled independently of subsequent layers, by growing the first layer under different conditions, to explore either the dense or sparse regime.

Figure 3 quantifies the increasing uniformity that is visually evident in Fig. 2. There may be no limit to the uniformity achievable in principle within this model. However, note the log scale—there is a dramatic slowing down of the process with increasing layer number. This slowing down may ultimately limit the degree of size uniformity achievable in practice.

It is also interesting to notice in Fig. 2 that the size variations among islands are not random. Closely spaced islands tend to have especially similar sizes, with a characteristic length over which such correlations are lost. We suspect that the slow improvement at large layer number is intimately tied to the disappearance of short-range variations. The band-pass filter analogy implies that short-range variations (which introduce high harmonics) are easier to filter out than long-period modulations.

Finally, we show some initial experimental evidence that such self-organization can be realized in practice. We have grown coherent (dislocation-free) islands of $Si_{0.25}Ge_{0.75}$ on Si(001), with a Si spacer thickness of $L = 100$ Å, and average $Si_{0.25}Ge_{0.75}$ coverage of 25 Å on each successive Si surface. Detailed experimental results will be published elsewhere [12]. Here we wish only to illustrate the qualitative behavior. We emphasize that the experimental conditions do not correspond closely to the simple assumptions of the model. For example, the islands are not well separated laterally; and they are far from spherical, having pyramidal or prismatic ("hut") shapes consisting of (105) facets [15]. This wide flat shape will result in a weaker lateral variation of the strain field, somewhat as a thicker Si layer would. Changing the island shape also changes the directionality of the strain field equation (1) somewhat [13].

Nevertheless, the qualitative similarities to Fig. 2(a) are striking. Figure 4 shows the surface after the first $Si_{0.25}Ge_{0.75}$ deposition and for a superlattice after the twentieth consecutive layer. The islands are larger in

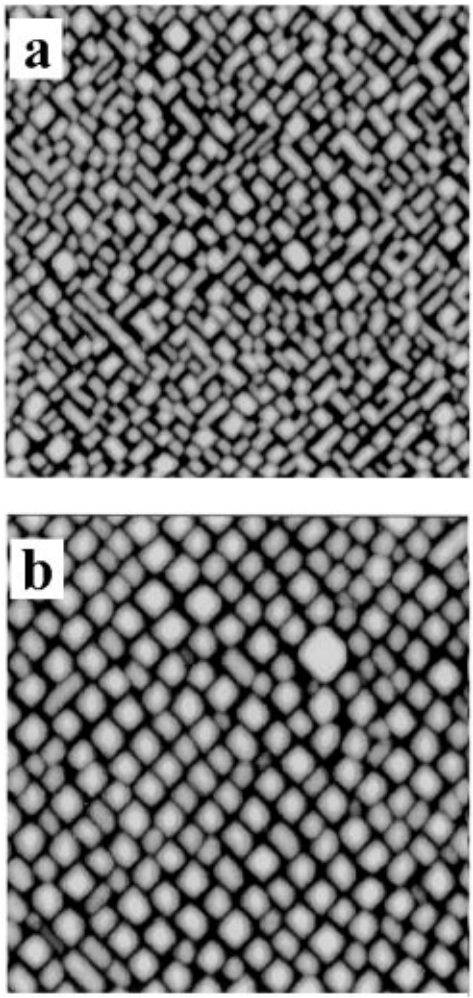

FIG. 4. AFM images of surface of Si-$Si_{0.25}Ge_{0.75}$ superlattices. Scanning direction is [110]. (a) 0.8 μm $\times$ 0.8 μm image after deposition of the first alloy layer; (b) 1.25 μm $\times$ 1.25 μm image after deposition of the 20th alloy layer. Gray scale range is 5 nm in (a) and 10 nm in (b), with light meaning high.

the latter case, and the increased regularity is apparent, as in Fig. 2(a). Cuts along the [100] direction (the direction of the island edges) through a two-dimensional power spectrum of this surface roughness show that the characteristic variation as a fraction of average size is reduced from 1.1 to 0.3, in qualitative agreement with the model.

The model here omits many important complexities of real systems. Nevertheless, it apparently captures a very real and simple effect. For multilayer arrays of strained island, such as several groups are already growing, under appropriate growth conditions the system will self-organize. With growth of successive layers, the size and spacing of the quantum dots becomes more uniform, increasing the likelihood that these structures could be used in practical electronic devices.

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