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Replication and alignment of quantum dots in multilayer heteroepitaxial growth

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Abstract

We propose a model to elucidate the self-organization process of islands in multilayer heteroepitaxial growth. The model is based on the preferential nucleation and growth of islands in regions of tensile strain in the total strain field on a spacer-layer surface induced both by embedded islands and by growing surface islands. Surface islands nucleate at the regions of tensile strain induced by embedded islands. The islands grow in size and induce increasingly large compressive strain on the spacer-layer surface. A surface island reaches a stable size after the total strain field around it becomes compressive. The model predicts that islands in successive layers not only form ordered columns but also show uniform distributions of island size and spacing, in agreement with experimental observations.

Keywords: Growth; Self-assembly; Surface stress; Molecular beam epitaxy

Strain-driven formation of nanoscale coherent islands on lattice-mismatched layers in heteroepitaxy offers a very attractive way for fabrication of arrays of quantum dots of many potential technological applications. Normally, these islands are not well organized in space and show large dispersion in size distribution. Recent experiments showed that size uniformity and spatial ordering of islands in heteroepitaxial growth can be greatly improved by growing multilayers of islands separated with spacer layers [1–7]. Such a self-organization phenomenon was found in semiconductor heteroepitaxial systems including InAs/GaAs [1,4], Ge/Si [2,3], CdZnSe/ZnSe [5] and PbSe/PbEuTe [6]. It was also found in magnetic systems such as Co/Au [7].

Several models have been proposed to investigate the morphological evolution of the stacked islands in multilayer heteroepitaxial growth, emphasizing contributions of the strain fields on spacer surfaces induced by embedded islands [1,2,8,9]. In these studies [1,2,8], an embedded island was simply considered as a force dipole of zero dimension within the continuum theory of elasticity. Vertical ordering of islands was qualitatively demonstrated by analyzing the preferential nucleation centers at local maxima of the tensile

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strain fields on spacer-layer surfaces [2]. In reality, islands have finite spatial extent with a linear size often comparable to the spacer-layer thickness. Finite size of islands must be considered in order to achieve a comprehensive understanding of the self-organization process. Considering the small height-to-base aspect ratio of embedded islands, Liu et al. [9] modeled an embedded three-dimensional (3D) island as a thin rectangular inclusion of a constant height. We note that a surface island induces compressive strain on the spacer-layer surface due to the lattice mismatch. The strength of such a compressive strain is often comparable to that of the tensile strain field induced by embedded islands. However, all existing models [1,2,8,9] completely neglect the influence of the strain fields induced by surface islands in multilayer heteroepitaxial growth.

In this Letter, we propose a novel model to elucidate the self-organization process of islands in multilayer heteroepitaxy. The model is based on the preferential nucleation and growth of islands in regions of tensile strain in the total strain field on a spacer-layer surface induced both by embedded islands and by growing surface islands. In our model, the strain field induced by embedded islands controls the location of the surface islands while the compressive strain field induced by surface islands plays a crucial role in controlling the size of islands. Specifically, surface islands first nucleate at regions of tensile strain induced by embedded islands. The surface islands grow in size and induce increasingly large compressive strain field on the spacer-layer surface. Eventually, a surface island stops growing after it reaches a critical size which makes the total strain field around it become compressive. In this case, adatoms on the spacer-layer surface prefer to nucleate at other places of tensile strain. Our model predicts that in the case of a sparse distribution of islands, islands of different sizes successively reach a common size, forming vertically stacked columns. In the case of a dense distribution of islands, both size distribution and island spacing are significantly improved after growth of successive layers. In this case, small islands adjacent to large islands approach to the large ones and closely distributed small islands coalescence. On the other hand, two closely distributed larger islands repel one another. The theoretical predictions are in excellent agreement with experimental observations.

We take Ge/Si multilayer heteroepitaxy as a specific example to introduce our model. Because of the small height-to-base aspect ratio of embedded islands, we consider a 3D Ge island as a thin rectangular inclusion as used in [9]. The volume of the island is $V = l \times w \times h$, where l, w, and h are the length, width, and height of the island, respectively. Ge islands are embedded in Si with a spacer-layer thickness d. We calculate the strain field using the continuum theory of elasticity, which has been shown to provide accurate results for embedded quantum dot systems [10]. For $l \gg h$ and $w \gg h$, we derive that the strain on the spacer-layer surface induced by a Ge island embedded at $(0, 0, -d)$ is given by

$$\varepsilon_e = \frac{\varepsilon_0(1+v)(1-2v)}{\pi(1-v)} \sum_{i=1}^{2} \sum_{j=1}^{2} (-1)^{i+j} \times \frac{X_i Y_j (X_i^2 + Y_j^2 + 2d^2)}{(X_i^2 + d^2)(Y_j^2 + d^2)(X_i^2 + Y_j^2 + d^2)^{1/2}}, \tag{1}$$

where $X_i = x + (-1)^i w/2$ and $Y_j = y + (-1)^j l/2$. Parameters v and ε_0 are Poisson's ratio of the spacer material and the lattice misfit with $\varepsilon_0 = (a_e - a_s)/a_s$, respectively. For Ge/Si, $a_s = 0.5656$ nm, $a_e = 0.5431$ nm, and $v = 0.218$. We note that Ref. [9] only gives an analytical expression of ε_e for a stripe-shaped embedded island with $w \gg l$ and $w \gg h$. The formula we derived in Eq. (1) is general. It applies to a rectangular island with arbitrary width and length, which significantly facilitates the calculation of the strain fields.

Exact information about the strain field induced by a surface island can be obtained either by atomistic molecular dynamics simulations or by finite element calculations based on continuum theory of elasticity. In our study, we are interested in the strain field on the spacer-layer surface around the surface island, $\varepsilon_s(x, y)$. Lacking of existing analytical expression for ε_s, we approximate it by taking $d = 0$ in Eq. (1). It is evident that atoms on the base layer of the surface island are forced to follow the lattice spacing of the spacer layer. As a consequence, the strained base layer of the island induces a compressive strain field on the spacer-layer surface. Within continuum theory of elasticity, the base layer of the surface island can be regarded as an embedded layer with $d = 0$. Considering the contribution of the upper layers of atoms of the surface island to the relief of the strain on its base layer, the strain on the spacer-layer surface outside of the surface island can be approximated by $\varepsilon_s(x, y) = \lambda\varepsilon_e(x, y)$ with $d = 0$, $w = w_s$, $l = l_s$ and $h = 1$ ML, where w_s and l_s are the width and length of the surface island, and $\lambda \leqslant 1$ is a fitting parameter. We compared this approximation with existing atomistic simulations [11] and found good agreement. The total strain field on the spacer-layer surface in the region $(|x| > w_s/2, |y| > l_s/2)$ induced by an embedded island and a surface island on top of it is given by $\varepsilon(x, y) = \varepsilon_e(x, y) + \varepsilon_s(x, y)$. In the following, we present our model based on the analysis of the total strain field. Because the value of λ does not change the qualitative picture, we take $\lambda = 1$ hereafter. For the sake of convenience of presentation, we consider islands of square bases, namely, $l \equiv w$, and $l_s \equiv w_s$.

In Fig. 1, we illustrate how the total strain field changes as a surface island grows in size. In Fig. 1, the embedded island has $w = 40a_{\mathrm{Si}}$, $h = 20$ ML. The spacer-layer thickness is $d = 80$ ML. Fig. 1(a) shows the strain fields induced independently by the embedded island and by surface islands of three different sizes $w_s = 5a_{\mathrm{Si}}$, $20a_{\mathrm{Si}}$, and $46a_{\mathrm{Si}}$. We can see from Fig. 1(a) that ε_e is tensile in the region directly above the embedded island. However, the strain field ε_s induced by a surface island is always compressive. Moreover, a larger surface island results in a larger surface strain field. Fig. 1(b) shows the total strain field $\varepsilon(x, y = 0)$ obtained by superposition of ε_e and ε_s. It is clear that the total strain field significantly changes as the size of the surface island increases. For a small surface island, the strain field around it is still tensile. The area of the tensile strain decreases as the size of the surface island increases. There exists a critical size $w_c = 46a_{\mathrm{Si}}$, above which there is no tensile strain around the surface island.

The strain fields on the spacer-layer surface control the growth of surface islands. Let us first consider the case of islands with large spacing

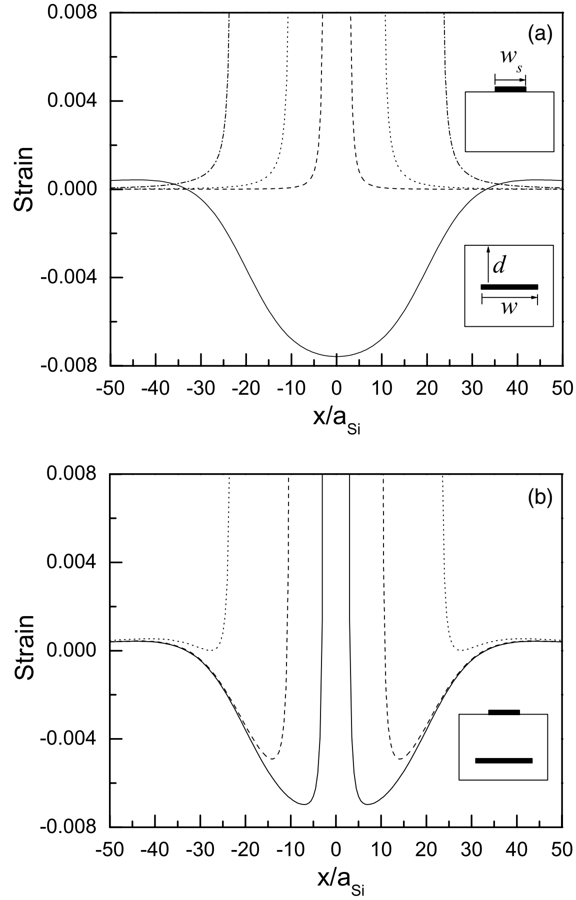

Fig. 1. Strain fields at the spacer-layer surface. (a) Solid line corresponds to $-\varepsilon_e(x, y = 0)$ induced by an embedded island with $w = 40a_{\mathrm{Si}}$ and $d = 80$ ML. Dashed lines, dotted lines, and dot-dashed lines correspond to $-\varepsilon_s(x, y = 0)$ induced by a surface island of size $w_s = 5a_{\mathrm{Si}}$, $20a_{\mathrm{Si}}$, and $46a_{\mathrm{Si}}$, respectively. (b) Total strain field $-\varepsilon = -(\varepsilon_e + \varepsilon_s)$ for $w_s = 5a_{\mathrm{Si}}$ (—), $20a_{\mathrm{Si}}$ (- - -), and $46a_{\mathrm{Si}}$ ($\cdots$). Insects are schematic illustration of the surface island and the embedded island.

(sparse distribution), where overlapping of strain fields induced by different embedded islands is negligible. Because of the higher nucleation rate, a surface island nucleates at the place on the spacer-layer surface with local maxima of the tensile strain induced by the embedded island [2]. The growing surface island modifies the strain field around it. Nevertheless, it continues growing provided that the total strain field around it is tensile. As the surface island reaches the critical size w_c, the surface island stops growing because of the absence of the tensile strain around it. In this case, adatoms on the spacer-layer surface prefer to nucleate at other places of tensile strain. Surface islands larger than w_c are found only if new surface islands start to form at places without tensile strain, which increases the island density. Because the island density remains constant after achieving self-organization, the size of a surface island on top of an embedded island is w_c.

We find that w_c is a function of the size of the embedded island. A small embedded island becomes larger and larger at successive layers and eventually reach a constant size w^*. On the other hand, a large embedded island becomes smaller and smaller to reach w^*. This leads to a common size of surface islands, w^*, after growth of a number of layers of islands, independent of their initial sizes. The convergence of the island size in successive layers can be clearly seen in Fig. 2(a), where three islands have initial sizes $w = 10a_{Si}$, $40a_{Si}$, and $120a_{Si}$ with $d = 80$ ML and $h = 20$ ML. From Fig. 2(a), one can see that the two small islands starting from $w = 10a_{Si}$ and $40a_{Si}$ grow in size successively to reach a constant size $w = w^* = 75a_{Si}$. The islands on successive layers starting from $w = 120a_{Si}$ shrink to w^*. Our detailed numerical analysis show that w^* only depends on the spacer-layer thickness d, with a smaller d corresponding to a smaller w^*. We emphasize that the observed convergence of island size in Fig. 2(a) is due to the contribution of the surface strain induced by the growing islands. One can see from Fig. 1 that the compressive strain given by a growing island reduces the area of the tensile strain induced by an embedded island. Owing to the larger the growing islands the smaller the area of tensile strain, growing islands limit growth of their size successively, resulting in a uniform island size distribution. In order to clearly show the crucial role of the growing islands in control of island size, we show in Fig. 2(b) island evolution without considering the contribution of the surface strain induced by growing islands, namely, $\lambda = 0$. From Fig. 2(b), we can see that islands with initial sizes as in Fig. 2(a) continuously increase their sizes until they merge into a very large island, showing no significant improvement of the size distribution quite different from the observations in Fig. 2(a) and in experiments.

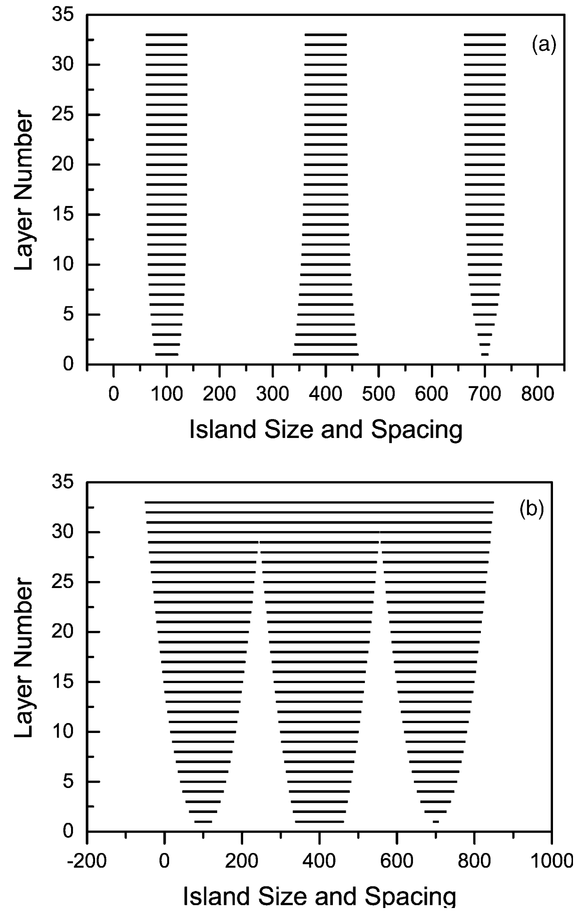

Fig. 2. Evolution of island size in growth of multilayers of sparsely distributed islands with a spacer-layer thickness $d = 80$ ML. Islands at starting layer have sizes of $w = 10a_{Si}$, $40a_{Si}$, and $120a_{Si}$. (a) $\lambda = 1$: Islands successively reach the same size $w = 75a_{Si}$ after growth of successive layers. (b) $\lambda = 0$: Islands continuously increase their sizes until they merge into a very large island, showing no significant improvement of the size distribution.

In the case of dense distribution of islands, our model predicts that both island size and island spacing are significantly improved after growing successive layers. Fig. 3 shows an example of the evolution of islands with small spacing. In Fig. 3, initial position, spacing, and size of an island are randomly chosen for the first layer. New islands on the next layer nucleate in the regions of local maximums of the tensile strain induced by islands on previous layers. The size of a surface island, w_c,

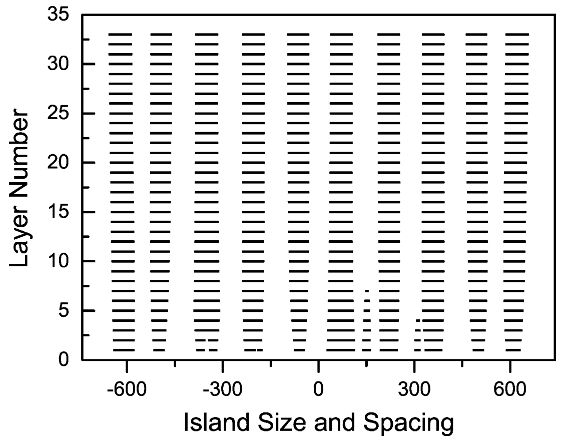

Fig. 3. Evolution of island sizes and positions in growth of multilayers of densely distributed islands with a spacer-layer thickness $d = 80$ ML. Islands at starting layer are randomly distributed at different positions with different island width from $w = 12a_{\mathrm{Si}}$ to $w = 84a_{\mathrm{Si}}$. Island spacing becomes uniform and island size distribution converges to a narrow range from $w = 61a_{\mathrm{Si}}$ to $67a_{\mathrm{Si}}$ after growth of successive layers.

is obtained by calculating the total strain field induced both by the islands on previous layers and by the surface islands with $\lambda = 1$. Islands on the next layer are considered as a single island once they have spatial overlap. From Fig. 3, we can see that both island size and island spacing are significantly improved after growing a number of layers. Island size distribution converges to a much narrower range from $61a_{\mathrm{Si}}$ to $67a_{\mathrm{Si}}$ compared with the initial distribution from $12a_{\mathrm{Si}}$ to $84a_{\mathrm{Si}}$. Island spacing is improved through the following three processes: First, small islands adjacent to large islands gradually merge to the large island. Second, small islands close to each other coalescence. Third, closely distributed larger islands never approach to each other. In order to further examine the important role of the surface strain induced by growing islands in the case of dense distribution of islands, we have also studied the island evolution without considering the contribution of growing islands by taking $\lambda = 0$ (with the same initial distribution of islands as in Fig. 3). In this case, we find that islands merge into large islands successively as in Fig. 2(b), showing no improvement of spatial ordering of islands. In addition, we find that islands start to merge much earlier compared with the case of sparse distribution of islands. This is because densely distributed embedded islands lead to severe overlap of the tensile strain fields, resulting in broadening of the size of tensile strain. Therefore, without the contribution of the compressive strain induced by growing islands, surface islands rapidly merge into large islands.

From the above discussions, we have seen that the repulsion of larger islands is one of the essential processes for achieving spatial ordering. In order to clearly understand the detail of this process, we show in Fig. 4 the evolution of two columns of islands with the same initial size $w = 30a_{\mathrm{Si}}$. We can see from Fig. 4 that islands in different columns grow in size and reach a common size successively, as observed in Fig. 3. However, different from the observation in Fig. 3, the two columns in Fig. 4 clearly deviate from their vertical alignment. The oblique alignment of islands shown in Fig. 4 is in good agreement with recent experiments [12]. We note that the occurrence of the oblique alignment in Fig. 4 is due to the absence of neighboring islands around the two columns of islands. In this case, the compressive strain field contributed by the surface island in one column forces the center of the tensile strain field of another column to move away from its original

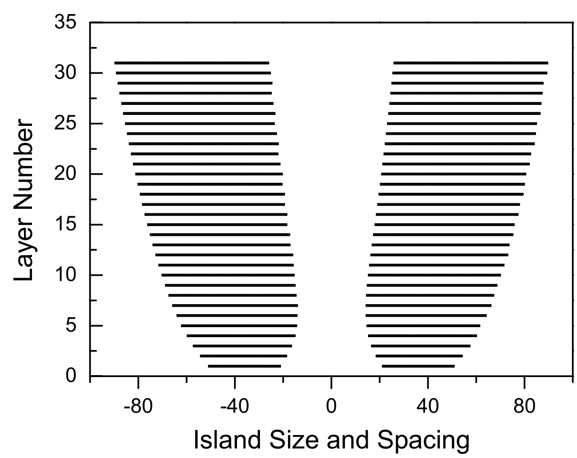

Fig. 4. Evolution of island size and position in growth of multilayers of islands with a spacer-layer thickness $d = 80$ ML. Two islands at starting layer of width $w = 30a_{\mathrm{Si}}$ successively reach the same size $w = 64a_{\mathrm{Si}}$. Island centers move away from their initial positions, forming oblique alignment of islands.

position. In the case of densely distributed islands in Fig. 3, neighboring islands almost induces the same repulsion effect in different directions. As a consequence, island centers remain their positions after achieving uniform distribution of island size and spacing, forming vertical alignment of islands as observed in Fig. 3.

In summary, we have proposed a model to elucidate the self-organization process of islands in multilayer heteroepitaxial growth. Different from existing models, our model features preferential nucleation and growth of islands in tensile regions of the total strain field on the spacer-layer surface induced both by embedded islands and by growing surface islands. The model predicts that in the case of sparse distribution of islands, islands with arbitrary sizes successively reach a common size, forming vertical columns of islands. For densely distributed islands, both island size and spacing are significantly improved after growth of successive layers. We have also studied islands of rectangular bases in addition to islands of square bases as reported in this paper. We find that a rectangular base with different length and width gradually become a square after growing a number of layers, resulting in the same shape for all islands [9]. It is interesting to note that most of our results (formation of vertical columns of islands, improvement of the uniformity both in the island size and spacing) agree well with the predictions obtained in the model proposed by Liu et al. [9] which does not take into account the surface strain induced by the growing island. However, we should note that in their model one has to assume island size to be smaller than the size of the tensile strain induced by embedded islands in order to keep successive confinement of island size. In our model, we believe that it is the contribution of the surface strain induced by the growing island that makes the island size smaller, resulting in the confinement of island size and a uniform island size distribution. Finally, we note that the growth of self-organized arrays of strained islands is a complex process which involves a number of conditions such as growth temperature, deposition rate, as wells as dislocations and defects. In our model, we examine the importance of the surface strain induced by growing islands to the self-control of island size and spacing. Our model focuses on the growth regime with modest growth temperatures and deposition rates where coherent islands preferentially form at the regions of tensile strain, as discussed by other theoretical and experimental studies [1–3,9]. At low/high growth temperatures or at large deposition rate, the strain fields induced both by the embedded islands and by the growing islands have less pronounced influence on the island nucleation and growth. Our model does not apply to these regimes. We should also note that in our model we assume that an island stops growing when the surface strain is not tensile, as suggested in [2], in order to facilitate discussions of the important role of the surface strain induced by growing islands. This assumption is mainly based on the consideration of the influence of surface strain fields on island stability. The assumption is not valid for growth systems where island stability strongly depends on island chemical potentials which are often functions of island size, curvature, strain, and density even under conditions of modest growth rates and temperatures.

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