Simulation model for self-ordering of strained islands in molecular-beam epitaxy

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The growth of quasiordered ultrasmall InAs islands by molecular-beam epitaxy utilizing the intrinsic elastic strain between the islands and the substrate is a promising approach to fabricate regular arrays of defect-free quantum dots. In this paper, a simulation of the island growth kinetics based on a phenomenological approach that introduces an effective exclusion zone that mimics the strain effects surrounding a growing island is described. The simulation results, in qualitative agreement with experiments, show that the growth kinetics can induce a quasiordering in the island position if either the exclusion zone or the nuclei density is sufficiently large.

I. INTRODUCTION

The thin-film island growth mode via the Stranski-Krastanow mechanism has recently been the subject of several studies. This growth regime offers the distinct advantage of producing defect-free quantum-dot structures that exhibit zero-dimensional quantum confinement properties. Recent experiments have demonstrated the successful growth of high-quality InAs ultrasmall islands by molecular-beam epitaxy[1] and metal-organic chemical-vapor deposition,[2] which have produced quantum dots that show discrete energy levels.[3–5] It has been suggested[6] that growth of islands by surface diffusion of adatoms can lead to a system with uniform island sizes. One can thus have some hope of controlling the size distribution of the islands by controlling the kinetics of surface diffusion.

Aside from the issue of size uniformity of the quantum dots, practical applications for devices utilizing these quantum dots may require the control of the nucleation positioning of the dots. The natural formation of self-aligned[7] and self-ordered[2] regular arrays of dots on GaAs(100) mesa structures[7] and on GaAs(311)*B* (Ref. 2) structures shows a short-range ordering in the positioning of the dots. Understanding the self-ordering mechanism may provide a way to grow highly ordered and prepositioned dots without any special preprocessing.

In this paper we will show a nucleation mechanism that permits some degree of ordering in the positions of the islands, despite the fact that the islands can nucleate in a ''random'' fashion. It has been suggested[8] that once a cluster nucleates at a particular location (a defect site), it acts as a sink for the surrounding adatoms. There is therefore an effective zone surrounding the cluster within which the ad-

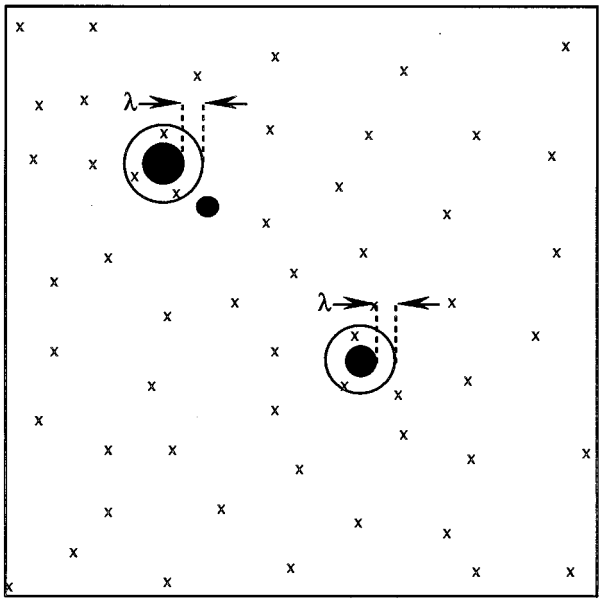

FIG. 1. Schematic diagram illustrating the exclusion zone concept. The symbols × represent the potential nucleation sites, which are distributed randomly on the surface. Dark circles represent growing clusters that have nucleated at defect sites. Any sites × that are within the rings (width λ) will cease to function as nucleation sites. In this model, λ is independent of the clusters' radii.

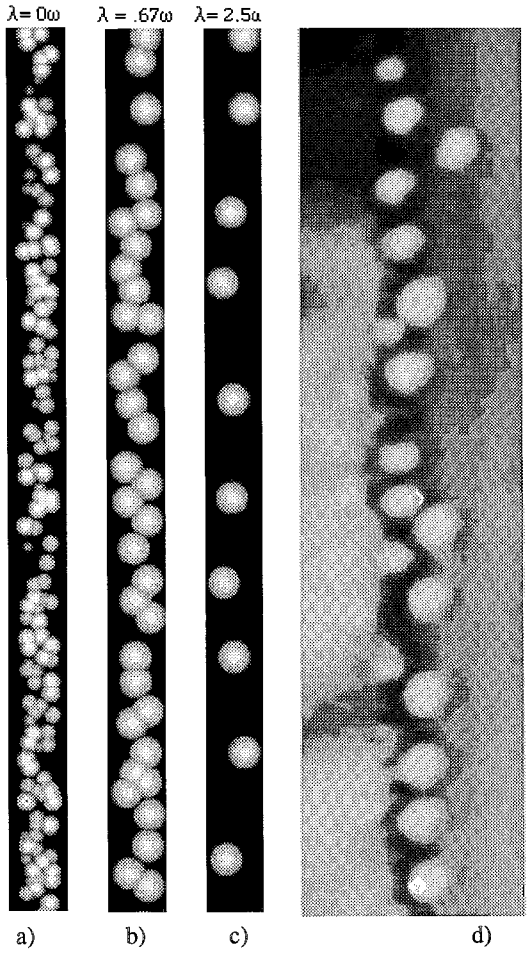

FIG. 2. (a)–(c) Initial potential nucleation density for these simulated images is set at 0.04. The exclusion zone widths for images (a), (b), and (c) are set at 0, 0.67w, and 2.5w, respectively. As seen from the images, the effective island density is reduced as the zone width λ increases. (d) Atomic force micrograph of a self-aligned string of InAs islands. The average island size is about 30 nm (see Ref. 7).

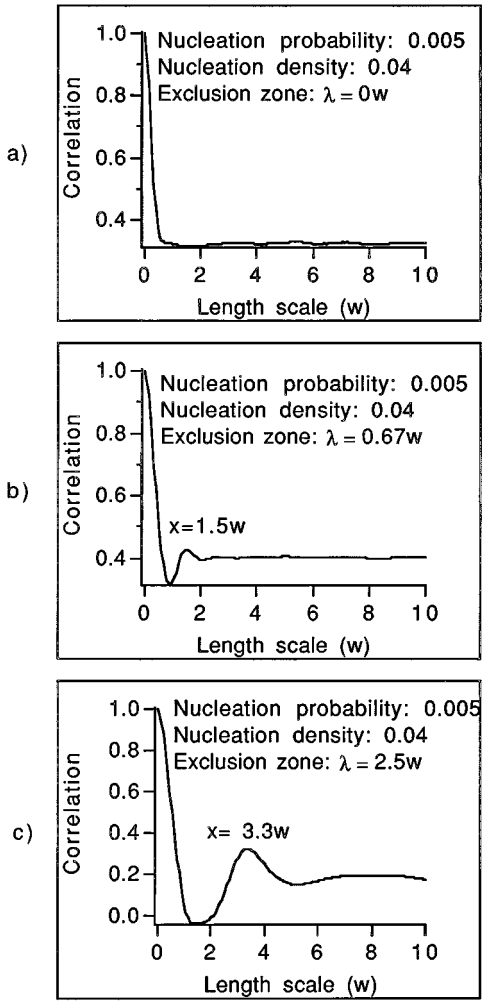

FIG. 3. Plots of the height-height correlation plotted as a function of length scale for the images in Figs. 2(a)–2(c). The peaks appearing in (b) and (c) show that there is a repeating feature at length scales indicated in the plots.

atoms are ''drawn'' to the cluster instead of forming a new cluster. This zone is termed the ''exclusion zone'' in which no new nucleation can occur. Outside this zone, islands can still nucleate at random positions. The positions of the islands are, to some degree, mediated by this exclusion zone.

To model the growth process, we adopt a nucleation model that prescribes a scheme for the nucleation kinetics. Suppose that cluster nucleation preferentially takes place at defect sites whose total population N_∞ (per unit area) is constant. These potential nucleation sites have a certain probability to be occupied by a nucleated cluster. Assume that this nucleation probability α (per unit time), which is determined by the nucleation condition (e.g., vapor pressure and energy barrier of nucleation) at each site, is constant at any given time. The rate of nucleation is thus proportional to the number of unoccupied sites and is given by[8]

$$\frac{dN}{dt} = \alpha N_\infty \exp(-\alpha t), \tag{1}$$

where N is the number of nucleated islands at time t. The rate of nucleation dN/dt or the rate of particle formation is an exponential decay with time. As more defect sites are occupied, fewer sites are available for new nucleation.

We have attempted to simulate the experimental results of Mui *et al.*[7] in which the InAs islands self-align quasiperiodically along a ⟨110⟩ direction parallel to the mesa structures defined by {311} and (100) facets. In these experiments faster surface diffusion kinetics on the {311} facets induces a local increase in the InAs film thickness along a narrow strip of the (100) surface adjacent to the {311} facet. The critical thickness for the island nucleation is reached at an earlier stage of the growth than for the remainder of the surface thus inducing a local island formation.[1(b)]

We start our simulation with a narrow strip having width w and length L. Initially, a fixed number of potential nucleation sites (defect sites) are randomly distributed within the strip. As time progresses, each site is ''turned on'' (or nucleated) randomly with a fixed probability a. Once a cluster germinates at one of the sites, its shape is constructed as a hemisphere with radius r that follows the equation[6,9] $r=K(t-\tau)^{1/3}$, $\tau<t$, where K is a constant and τ is the birth time of the cluster. As the island grows, any potential nucle-

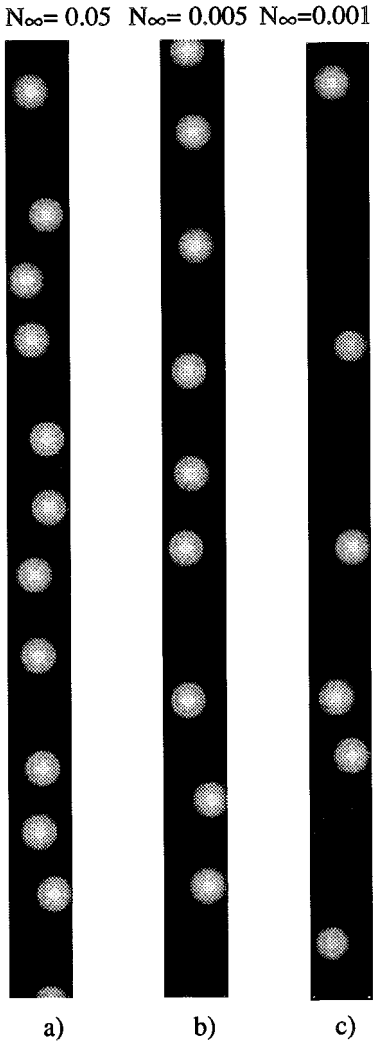

FIG. 4. Exclusion zone in this set is fixed at $\lambda = 2w$. The varying parameter in this case is the initial potential nucleation density N_∞. The values of N_∞ in (a), (b), and (c) are 0.05, 0.005, and 0.001, respectively.

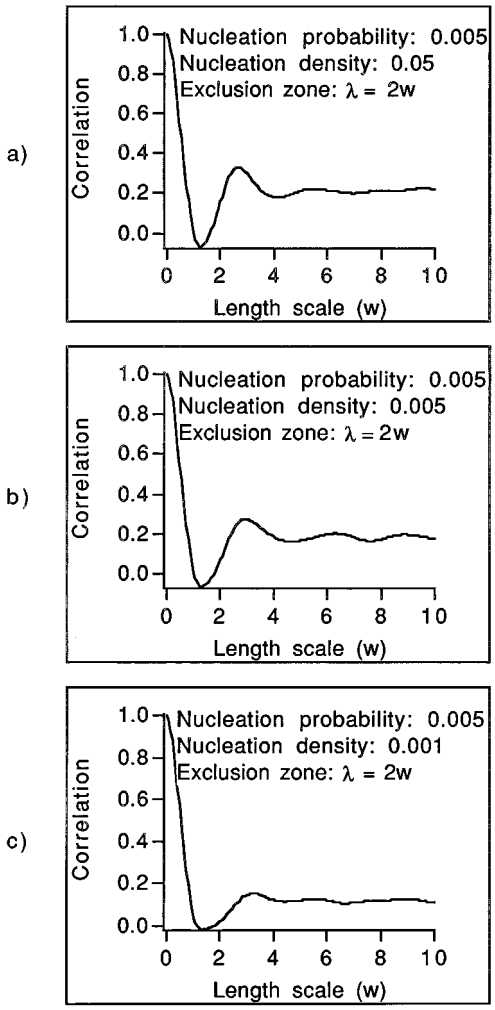

FIG. 5. Height-height correlation plots of images in Figs. 4(a), 4(b), and 4(c) are shown in (a), (b), and (c), respectively.

ation site within the zone of width λ measured from the perimeter of the island ceases to exist as a potential nucleation site (see Fig. 1). Within this exclusion zone, the preferential diffusion of the adatoms towards the existing cluster is driven by the lower surface atom concentration and local strain in the immediate vicinity of the cluster. In this simulation, λ is set as a constant, although in general it will be a function of the local strain and therefore of the island size.

The simulation is stopped when the islands occupy about 30% of the strip area. This value corresponds to the observed coverage in the experiment. The islands are taken as perfect hemispherical caps, and to measure the degree of ordering of the islands positions, the height-height correlation function of the surface profile of the strip is calculated. Specifically, if $h(\mathbf{r})$ is the height profile of the strip at position $\mathbf{r}$, the height-height correlation function $G(l)$ at length scale l along the length of the strip is defined as

$$G(l) = \langle h(y+l)h(y) \rangle, \tag{2}$$

where y is the longitudinal coordinate along the strip, and $\langle\ \rangle$ denotes the averaging of all height pairs separated by a distance l along the strip's length.

Equation (2) is very sensitive to any periodic features on the surface, and a peak in the plot of Eq. (2) will indicate the length scale at which these features repeat. The degree of positioning ordering of the islands is measured by the intensity (the absolute value) of $G(l)$. The two varying parameters being investigated in this simulation are the width of the exclusion zone λ and the nucleation density N_∞.

II. RESULTS

In all of the following results, the nuclei density is in units of the total surface lattice points used in the computation (35×2048) and the exclusion zone is expressed in units of the strip's width w. Figures 2(a)–2(c) are the images of the islands that nucleate and grow with exclusion zone values set at 0, $0.67w$, and $2.5w$, respectively. Figures 3(a)–3(c) are the corresponding plots of the correlation function G. As one might have expected, if there is no exclusion zone ($\lambda = 0$) [Fig. 2(a)] the islands can nucleate at random positions

within the strip, in which case there is no positioning ordering [see Fig. 3(a)]. However, as the exclusion zone increases to larger values [see Figs. 2(b) and 2(c)], the islands self-align in a quasiperiodic fashion, as indicated by the peaks in the correlation plots [see Figs. 3(b) and 3(c)]. The positions of the peaks correspond to roughly $R+\lambda$, where R is the average radius of the ensemble. For a given nucleation density, starting from $\lambda=0$, the degree of ordering increases as λ increases [compare Fig. 3(b) with Fig. 3(c)]. However, the simulations indicate that there is a value λ, between $0.67w$ and $2.5w$ in this case, beyond which the ordering of the islands' positions cannot be improved further.

We show in Fig. 2(d) an atomic force micrograph of such a region. Note that over this long distance, a 1000-nm-long surface strip, there is evidence of short-range ordering of the islands.

A different set of simulation results is shown in Fig. 4. For this simulation, a fixed value of the exclusion zone $\lambda=2w$ and a varying nucleation density N_∞ were chosen. The corresponding autocorrelation plots are shown in Fig. 5. Starting from a relatively high nucleation density [Fig. 4(a)], the degree of ordering is reduced as the density of nucleation sites decreases [see Figs. 5(a)–5(c)]. As the nucleation site density is decreased, and when the islands are sufficiently far apart from each other (so that the average distance between them is greater than $R+\lambda$), the correlation is very weak [Fig. 5(c)]. Since the simulation is stopped for a coverage of 30%, we find that the island radii are similar for the three nucleation fractions used in this case.

It is clear from these two sets of simulation that the exclusion zone can play a significant role in ordering the island positions even though these islands nucleate at defect sites that are distributed randomly in a narrow surface trip.

III. CONCLUSION

This simulation demonstrates that the presence of an exclusion zone surrounding each cluster can effectively mediate the position ordering of the islands, at least in one dimension. This exclusion zone may be associated with the minimization of the strain energy during growth of the islands. In effect we have simulated the quasiordering of the self-aligned islands observed by Mui *et al.*[7] Although we have relied on the declining nucleation model to determine the rate at which islands are generated, it is expected that the quasiordering can occur independently of any nucleation scheme.

ACKNOWLEDGMENTS

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