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# Quantum dot self-assembly in growth of strained-layer thin films: A kinetic Monte Carlo study

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We use Monte Carlo (MC) simulations to study island formation in the growth of thin semiconducting films deposited on lattice-mismatched substrates. It is known that islands nucleate with critical nuclei of about one atom and grow two dimensionally until they reach a critical size $s_c$, when it is favorable for the islands to become three dimensional. We investigate the mechanism for this transition from two-dimensional (2D) to three-dimensional (3D) growth. Atoms at the edge of 2D islands with the critical size $s_c$ become mobile as a result of strain and are promoted to the next level. Edge atoms of the resulting island remain highly strained and are promoted to the higher layers in quick succession. This process of depletion is rapid and occurs at a sharply defined island size. We discuss why this leads to the uniformity seen in self-assembled quantum dots in highly mismatched heteroepitaxy. The results of the MC simulations, although done in $(1+1)$ dimensions, should be applicable to $(2+1)$ dimensions.

## I. INTRODUCTION

There has been considerable attention in recent years on the nature of the formation of three dimensional (3D) islands called quantum dots (QD's) (Refs. 1–4) during the growth of strained-layer superlattices. For Ge grown on Si(001), for example, the nature of islands seen have been characterized by Mo *et al.*[5] ''Hut'' clusters are the first type of islands to appear with well defined (105) facets, tilted at 11.3° to the surface, then there is a transition to larger islands with $(11n)$-like faces,[6] and finally to even larger but dislocated islands. It is possible to bypass the hut cluster stage by growing at slightly higher temperatures.[5,7] Another much studied system is the growth of InAs on GaAs(001) (mismatch $\sim 7\%$); here the particular interest is in the uniformity in the size of islands formed.[1,2] This uniformity, with dispersions of 10% in height and 7% in diameter of the islands at the initial stages of formation, decreases[1] with coverage $\theta$. There seems to be a distinct coverage $\theta_c$ [=1.5 monolayers (ML's),[1] 1.75 ML's,[2] 1.7 ML's (Ref. 3)] at which the transition from two dimensional (2D) to three dimensional (3D) growth occurs for the InAs/GaAs system. This critical thickness transition is slightly dependent on growth conditions; the work of Gerard *et al.*[3] shows that by substantially increasing the deposition rate, for example, it is possible to shift it from 1.7 to 1.95 ML's. There is evidence that the material to build an island comes mainly by depleting its immediate environment: the thickness of the InAs layer before islanding occurs, which is between one and two ML's, is reduced to one ML in the immediate region surrounding the island.[1–3] This suggests that the critical layer thickness for InAs/GaAs(001), beyond which it is energetically favorable to form islands, is actually one ML and that the extra thickness before islanding may be due to the presence of a barrier at the 2D to 3D transition. There are signs that some depletion is also present in the growth of Ge on Si.[8,9] It is known that island shapes and sizes can depend on growth conditions,[10–12] so that kinetic effects are important. Under much higher deposition rates and lower growth temperatures than those used by Leonard *et al.*[1] and Moisson *et al.*,[2] Ruvimov *et al.*[4,12] found that islands also exhibit size uniformity ($<20\%$); however, island size increases with coverage.[4,12] While Moisson *et al.* observed (104) and (110) facets on the islands, Grundmann *et al.*[4] and Ruvimov *et al.*[12] saw only (110) facets; Moll *et al.*[11] showed that the equilibrium shape of an InAs island involves (111), $(\overline{111})$, and (110) facets in proportions which change with the size of the island.

Kinetic effects clearly change the shape and size of islands, and may even affect the critical thickness for the 2D-3D transition; however, the uniformity of islands seems to be robust for the highly strained InAs/GaAs system. We first look at the experimental results of islanding in InAs/GaAs systems because of the availability of data at small increments of coverage $\theta$.[1–3] There are a number of observations that need to be discussed. The first is the narrow distribution in width and height of the 3D islands. The second is the existence of a sharp (possibly first order) 2D-3D transition at a critical coverage $\theta_c$.[1] There is also the phenomenon of fast depletion (of the order of seconds[3]), where a 3D island is created quickly [compared to a deposition rate of 0.01 ML/sec (Ref. 1)] largely out of the atoms from the its 2D environment. Finally, it is also seen that under conditions of slow deposition 3D islands remain essentially constant in size over a coverage interval of $\Delta\theta \sim 0.4$. Note that these results are affected by growth conditions. Under high deposition rates (compared to diffusion rates) which is possible at low temperatures, the sharp island size distribution may disappear, see, for example, growth of Ge on Si(001),[9,10,13] where the lattice mismatch is smaller ($\sim 4\%$). For the Ge/Si system, where the strain is much less than that in the InAs/GaAs system, depletion seems to take a time of the order of minutes at 550 °C,[8,9,13] and recent growth experiments were carried out at typical deposition rates of a few ML's/sec.[9] Under these growth conditions, even the sharp 2D-3D transition may disappear.[14]

In this study then we focus on the early stages of growth for thin films which grow in the Stranski-Krastanov (SK) mode. We study growth under conditions where diffusion is fast compared to deposition, so that effects due to the process of depletion can be distinguished from those due to deposition. In a previous work,[15] we investigated the energetics of the 2D to 3D transition in detail by means of molecular-dynamics simulation, using an empirical potential that has been appropriately tuned.[15] We argued that the 2D to 3D transition occurred when 2D islands had grown much larger than the size $s_o$, when 3D islands *first* become energetically favorable; this effectively is a barrier, which once scaled by a 2D island, allows it to reorganize itself into a 3D shape, with an *immediate* gain of energy. This gain, which is more pronounced for the highly mismatched InAs/GaAs system than for the Ge/Si, can be quite substantial, about 5–10 meV/atom for the former. We feel that this is the underlying factor for the uniformity of sizes of islands seen in this system. Priester *et al.*[16] have attempted to provide an explanation for the uniformity of the 3D islands, but have not taken into account the factor of the barrier, which should affect their considerations.

It is known for the growth of Si/Si(001) (Refs. 17–19) that islands nucleate with a critical size of one to three atoms and then grow two dimensionally. This picture of nucleation is also supported by the results of Chen and Washburn,[20] who used a critical nucleus of $i=1$ in the scaling function $\Phi(N/\overline{N})$ (Ref. 22) in fitting the island density results of Leonard *et al.*[1] Island nucleation of Ge on Si(001) should be similar (in both cases the dimer is the stable nucleus). For our picture below, the critical nucleus only needs to be small, so that its further growth will be two dimensional. Although Theis and Tromp[21] have reported, for high-temperature ($\sim 650\,^\circ\mathrm{C}$) growth, critical nuclei of $\sim 650$ dimers, this clearly, is still small when compared to smallest 3D island sizes of 20 000–50 000 atoms for the growth of InAs on GaAs.[16] We suggest that the 2D–3D transition picture is the following: 2D islands nucleate with critical nuclei of about one atom and grow two dimensionally until a critical size $s_c$ when strain makes it favorable for there to be a transition to 3D growth. This size $s_c$ is quite large, roughly a few hundred angstroms. There is direct experimental evidence for this picture of growth. Mo and Lagally observe,[23] after growth of about 3 ML's of Ge on Si at 500 °C, a growth front roughness of three layers over an area of 60×60 nm. Gerard *et al.*[3] observe one layer roughness over extensive 2D areas (~2000 Å) for the growth of InAs on GaAs(001) at 520 °C. We stress that $s_c \gg s_o$, the size at which 3D clusters have just become energetically favorable. Indeed the 2D island must reach a size comparable to that of the two-layer island when the latter becomes energetically favorable. Once this size barrier is reached, the transition to islands of two or more layers in height is possible since taller islands are already favorable at smaller sizes. There is a rapid rearrangement of its atoms in order to achieve the shape of the optimally energetic (105) facetted clusters. There is an immediate gain in energy of 1–2 meV/atom for the Ge/Si system; for InAs/GaAs, we estimate this gain, assuming that the elastic energy scales with the square of misfit, to be 5–10 meV/atom.[24] This latter amount is substantial and is probably the reason for the phenomenon of depletion seen in the highly mismatched systems.

The above picture obtained from an energetics study is complemented by our work here on kinetics. In this study, we approach island growth on strained-layer superlattices, by using finite temperature nonequilibrium Monte Carlo (MC) simulations, where diffusion rates of adatoms depend on strain as well as the usual local bonding. Computational time and size constraints force us to carry out our kinetic MC simulations in 1+1 dimensions, i.e., in our MC simulations the substrate is one dimensional and the growth is two dimensional. We do not believe that our 1+1-dimensional simulations introduce any qualitative complications, although it will be necessary in the future to verify our proposed picture using the full three-dimensional MC simulations. Our results show that under growth conditions of fast diffusion relative to deposition, i.e., not very low growth temperature, the picture obtained from energetics is largely correct. There is a sharp 2D–3D transition which occurs at an island size $s_c$ which is well beyond the critical size $s_o$ at which the 3D islands first become energetically favorable. Depletion is observed and narrow 3D island distributions are obtained. The average size of 3D islands does not change with coverage. In this work, we attempt to understand the microscopic dynamics and mechanisms underlying these results. In the following section we describe the simulation method and the parameters chosen. Then we present detailed results of the simulation in Sec. III and discuss the results in Sec. IV. We conclude in Sec. V.

## II. SIMULATION MODEL

In our MC growth simulations (which is done in 1+1 dimensions), an adatom moves [under solid-on-solid restrictions (SOS)] by hopping randomly to neighboring sites at a rate that depends on its bonding. (We obey detailed balance in our kinetic MC simulation.) The hopping activation energy depends on the bonding environment and the elastic energy associated with strain. The hopping rate is given by the expression

$$R_n = R_o \exp^{-E/k_bT}, \tag{1}$$

where $R_o = d'kT/h$ is a characteristic vibrational frequency and $d'=1$ is the substrate dimension. The activation energy $E = E_{bond} - E_{strain}$, with $E_{bond}$ being determined by the number of nearest neighbors (nn) and next-nearest neighbors (nnn). The elastic energy is given by harmonic interactions between an atom and its nn and nnn neighbors, using spring constants $k$. Following Orr *et al.*,[25] we obtain $E_{strain}$ for a particular site by taking the difference in elastic energies of the system when the site is unoccupied and when the site is occupied. This energy is calculated by allowing atoms in a 5(height)×7(width) cell centered at the site first to equilibrate under molecular-dynamics simulation and then to relax to its minimum energy configuration by means of the method of steepest descent. Every 100 time steps or so the entire system is allowed to relax globally to avoid any local strain accumulation. $E_{bond}$ is chosen in the following way:

$$E_{bond} = \begin{cases} E_o = (0.7\ \mathrm{NN} + 0.2\ \mathrm{NNN})\ \mathrm{eV}, & \text{if NN} \leqslant 2 \\ E_1 = 4.0\ \mathrm{eV}, & \text{if NN} = 3 \\ E_2 = 1.45\ \mathrm{eV}, & \text{steps of height} \geqslant 2. \end{cases} \tag{2}$$

TABLE I. Growth roughness of the unstrained system as a function of island-end diffusion barrier height.

| $T$ | 750 K | | | 800 K | |
|---|---|---|---|---|---|
| $E_{strain}$ (eV) | $E_{end}$ (eV) | $\theta$ | RI | $\theta$ | RI |
| 0.5 | 1.3 | 0.56 | 29.3 | 0.56 | 41.0 |
| 0.4 | 1.4 | 0.63 | 7.4 | 0.65 | 17.6 |
| 0.3 | 1.5 | 0.61 | 1.3 | 0.63 | 3.2 |
| 0.2 | 1.6 | 0.64 | 1.1 | 0.62 | 1.3 |

where NN is the number of nn's and NNN is the number of nnn's. $E_o$ applies to single adatoms or atoms at step edges, except when step heights are two layers or greater. Then $E_2$, a reduced barrier height, is applied to the surface atoms on top of these steps, so that inclined (11) island facets are favored over vertical ones. $E_1$ is the barrier for the rest of the surface atoms which have three nn's. It is chosen a little higher than that given by bond counting to eliminate intrasubstrate breakaway (especially at the foot of islands) and therefore to avoid substrate roughening, which is not seen experimentally.[3,5] For simplicity we have also used the same barrier for midisland surface atoms; results are not different from those using bond counting for these atoms. The parameters have been chosen so that diffusion will dominate over deposition, for example, a single adatom will diffuse a distance of approximately 600 unit cells for each deposition event at 750 K. This is about 50–100 times the width of the islands that form. Using diffusion rates from Mo *et al.* and others[5,26] and experimental deposition rates and island sizes,[2,1] we get comparable results of the ratio diffusion distance/island size$\sim$100. We choose the spring constant $k = 200\ \text{eV}/a^2 \sim 200$ times the magnitude of the diffusion barrier for a single adatom,[25] and a deposition rate $R$ of 0.01–0.2 ML's/sec, where a is the unit-cell size. We carried out simulations for strained-layer lattices with misfits $\delta a/a$ of 0–7%, at temperatures $T$ of 700–800 K. The natural lengths are $(a+\delta a)$ for the strained material and $a$ for the substrate. We start with systems at thicknesses of 11 ML's, with the three top layers at the larger lattice constant. System sizes vary from 500 to 8000 cells. At zero strain, growth was layer by layer as would be expected under the above conditions of fast diffusion—there is no kinetic roughening at this ''high-temperature'' growth in the absence of strain.

## III. RESULTS

We report on two preliminary studies that will help in understanding the final results. First we carry out simulations for the unstrained system where we set $E_{strain}=0$ everywhere except for atoms at the ends of islands; at these sites $E_{strain}$ is varied from 0.2 to 0.7 eV, corresponding to diffusion barriers of $E=E_{end}$ of 1.3 to 1.8 eV when NN$=$2. Simulations are done for temperatures $T$, from 700 to 800 K and deposition rates 0.1–0.2 ML's/sec and over coverages of $\theta$ from .5 to 0.8. In Table I, we display the results for two growth temperatures, 750 and 800 K, with a deposition rate of 0.2 ML's/sec, a system size of $10^4$ cells and a coverage $\theta\sim 0.6$. We calculate a roughness index (RI) as the percentage of sites in islands, which have heights $>1$, i.e., RI is a rough measure of the deviation from ''two dimensionality'' (one-dimensionality in our simulations) in the islands. For $E_{end}\geqslant 1.5$ eV growth is smooth, islands are flat (very small RI), but growth is distinctly rough for $E_{end}\leqslant 1.4$ eV, there being much larger proportions of islands with two or more layers in height. Clearly the transition from smooth to rough growth is sharp.

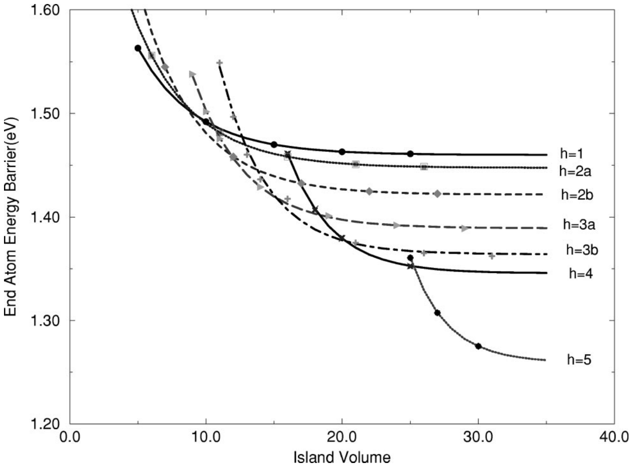

FIG. 1. Energy barriers $E_{end}$ of atoms at ends of islands plotted against island volume. The island configurations are: (a) one-level islands (h$=$1), (b) two-level islands with 1 atom (h$=$2a) and two atoms (h$=$2b) on the second level, (c) three-level islands with one (h$=$3a) and 2 atoms (h$=$3b) on the third level and three and four atoms, respectively, on the second level, (d) four- and five-level atoms with one atom on the top level (h$=$4, 5 respectively) and the same shape as in (c).

Next we look at island-end energy barriers $E_{end}$ for some island configurations when elastic interactions are included. Specifically, we carry out calculations for misfits of 5%. In Fig. 1, we plot $E_{end}$ against island volume (number of atoms), for seven island configurations, comprising (a) one-level islands (h$=$1), (b) two-level islands (with one atom (h$=$2a) and two-atoms (h$=$2b) on the second level, (c) three-level islands with one (h$=$3a) and two atoms (h$=$3b) on the third level and three and four atoms, respectively, on the second and (d) four- and five-level islands each with one atom on the top level (h$=$4 and 5, respectively) and the same shape as islands in (c). Island volumes are varied by changing the length of level 1 of the islands, while keeping upper configurations fixed. If we take $E_{end}<1.5$ eV as the condition for rough growth, then islands with volumes $>15$ will have end atoms with diffusion barriers $<1.5$ eV for all the consequtive configurations 1,2,3 and higher levels. The following picture of 3D islanding is suggested: 2D islands grow two dimensionally until a certain size when end atoms are promoted to the second level; this process becomes more rapid as it proceeds because $E_{end}$ decreases with the number of atoms on the second level (while island volume is kept constant). This process then continues in the same fashion with the subsequent promotion of atoms to the third and higher levels. This, we believe, is the mechanism for the phenomenon of depletion seen experimentally.[1–3]

We now present results of our full kinetic MC simulation done on systems of substrate sizes $L=2000$, 4000, and 8000 cells. The observations we report below are true of all these sizes and so are not affected by finite-size effects. For these simulations, we also consider the effect of a strain enhancing

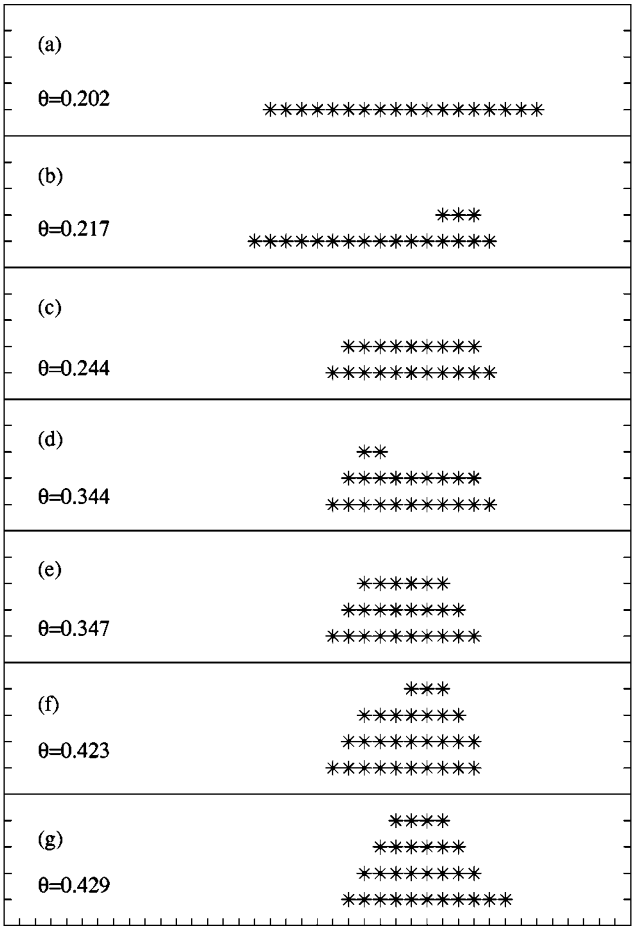

FIG. 2. The process of depletion: A 2D island undergoing a transition to the 3D shape. (a) shows the 2D island just before it is rapidly folded up into a two-level island in (b) and (c). (d)–(g) show rapid promotion to the upper layers.

factor $F_{end}=1.0$, 1.2, and 1.5 on the first level end atoms of islands (i.e., $E_{end}$ is replaced by $E_{end}\times F_{end}$). It is known that there is tremendous strain at the foot of islands.[27,28] Our results are not particularly sensitive to variations in this strain, aside from making islands a little smaller as $F_{end}$ is increased. In the figures and tables below, the results are for $T=750$ K, $\delta a/a=0.05$, and $R=0.08$ ML/s. In Fig. 2, we follow the development of a single island over a growth period of about 0.2 ML (<3 sec). Figures 2(a)–(c) show a one-level (2D) island of volume 19 atoms being folded up into a two-level island in 0.5 sec. The material for this two-level island (volume = 20) comes almost completely from the original one-level island. In the rest of Figs. 2(d)–(g), we see similarly rapid buildups of the third and fourth levels after a brief waiting period. The whole process starting from Figs. 2(a)–(g) takes less than 3 sec. The bulk of the material (~80% for the three-level island and ~65% for the four-level island) for the formation of the 3D island comes from the original 2D island (compare with the experimental results of the three groups above[1–3]). Figure 2 shows a typical 2D-3D transition sequence for islands in our simulation. It clearly illustrates the process of depletion seen experimentally. (Note that in our simulations what we refer to above as 2D and 3D are really 1D and 2D, respectively, since we are using 1+1-dimensional simulation).

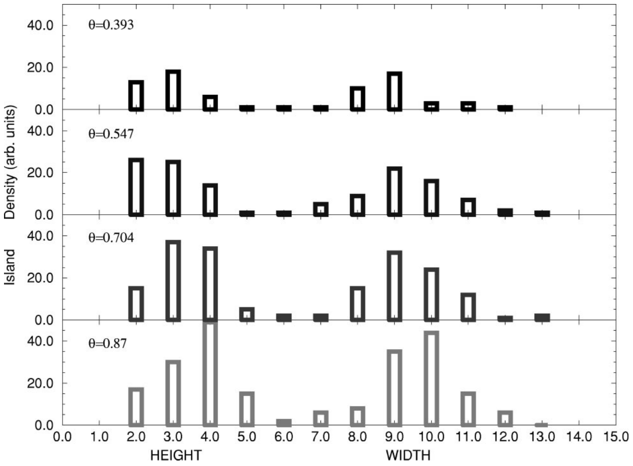

FIG. 3. Height and width distributions of islands at coverages $\theta=0.393$, 0.547, 0.704, and 0.87 ML.

In Fig. 3 we display width and height distributions of islands for a range of coverages $\theta=0.393-0.87$. There is uniformity in the island size distributions which are sharply clustered around the mean width or height, each with a half-width of ~1 cell. Furthermore, while island density increases with coverage, the average island size remains essentially constant. In Table II, we show the average volumes at which islands undergo transitions from the first to the second levels, from the second to the third, and from the third to the fourth level. The root mean square deviation is two atoms in each case, showing that transitions occur at sharp distinct sizes. Table II indicates that on average, at the point of formation of the three-level island, almost 80% of the material comes from the 2D island and that 73% of the four-level island comes from that island.

We plot in Fig. 4 the total number of islands with three or more levels as a function of coverage $\theta$ for systems of size $L=4000$. The results are the same for systems of other sizes ($L=2000$ and 8000) when appropriately normalized. We see that island density is zero until a certain coverage $\theta_c$ is reached, when the density increases rapidly. Leonard *et al.*[1] observed this experimentally and fitted the island density $\rho_{isl}$ with the function $\rho_{isl}=\rho_o(\theta-\theta_c)^{\alpha}$. They obtained a value of $\alpha=1.76$ while we get $\alpha=1.34$. The difference in the value of $\alpha$ could be due to our using a 1+1-dimensional simulation. We arrive at similar conclusions if we look at islands with two or more levels instead of the $\geqslant$ three levels we have chosen above. Since we consider only elastic effects in our simulations, we do not include a possible wetting layer $\theta_w$(=1 ML for the growth of InAs on GaAs,[1–3] and 3 ML's for Ge on Si) caused by absorbate-substrate interactions; our $\theta_c$ should be compared with experimental $\theta_c-\theta_w$ (=0.5–0.7 ML for InAs on GaAs).

In Table III, we show the energies of islands of various configurations comprising two, three, and four levels, rela-

TABLE II. Mean volumes at which islands undergo transitions from levels 1–2, 2–3, and 3–4.

| Transitions from levels | 1–2 | 2–3 | 3–4 |
|---|---|---|---|
| Island volumes | 19.0 ±2 | 21.4 ±2 | 26.3 ±2 |

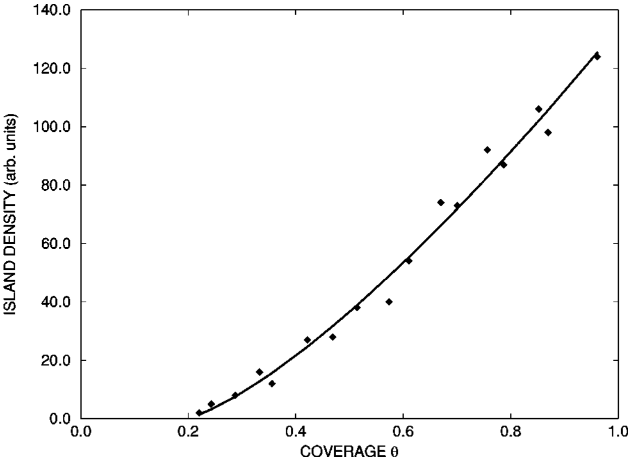

FIG. 4. Plot of 3D island density versus coverage $\theta$ for system sizes $L=2000$, 4000, and 8000. Island density is normalized to that for $L=4000$.

TABLE III. Energetics of islands of various configurations relative to the energy of the corresponding one level island at the same volume.

| | Island configurations | | | |
|---|---|---|---|---|
| Volume | two-level | three-level | four-level | $\Delta E$ |
| 4 | 1,3 | | | 0.58 eV |
| 6 | 2,4 | | | 0.18 eV |
| 8 | 3,5 | | | −0.14 eV |
| 9 | | 1,3,5 | | 0.24 eV |
| 12 | 5,7 | | | −0.72 eV |
| 12 | | 2,4,6 | | −0.42 eV |
| 14 | 6,8 | | | −0.97 eV |
| 15 | | 3,5,7 | | −1.06 eV |
| 24 | 11,13 | | | −1.79 eV |
| 24 | | 6,8,10 | | −2.72 eV |
| 24 | | | 3,5,7,9 | −2.6 eV |
| 28 | 13,15 | | | −1.28 eV |
| 28 | | | 4,6,8,10 | −3.48 eV |

tive to the energies of their corresponding one-level configuration at the same volume. We see that the first energetically favorable two-level island is the one whose volume is eight atoms with a configuration of three atoms on the second level and five on the first. Three-level islands become favorable at a volume of about 12 atoms but for this volume the two-level configuration has the best energetics. Three and four-level islands are energetically optimal at volumes of 15 atoms and 24–28 atoms, respectively. These figures can be compared to the transition volumes of Table II. There clearly is a correspondence between energetics and kinetics. However, one interesting point emerges, although a two-level island becomes energetically favorable at a volume of eight atoms, kinetically the transition occurs at a volume well beyond that (around 19 atoms). Energetics sets the lower size limit for the beginning of depletion, but it is kinetics that determines the actual point. This is the $s_c \gg s_o$ kinetically driven scenario we discussed before. Note also that because of the small size of the islands we encounter here, it is the two-level island that first becomes energetically favorable before the taller islands; we expect the situation to be reversed when mean island sizes are larger as surface energies become less significant—this would be the case with the sizes actually seen experimentally. This aspect of physics is not appropriately captured in our small system $1+1$-dimensional simulations.

## IV. DISCUSSION

The experimental results of islanding in InAs/GaAs systems of a number of groups are shown in Table IV. The first four groups observed uniformity in the size distributions of the islands, in particular, Leonard *et al.* reported dispersions of 10% in height and 7% in diameter at the first appearance of the islands, at $\theta \sim \theta_c$; with further deposition, this uniformity is reduced, island density increases but sizes remain essentially the same.[1,29] The first three groups concluded that there is depletionlike behavior. Leonard *et al.* show that more than 80% of the atoms to form an island comes from its environment, rather than from additional deposition. Gerard *et al.* display an atomic force micrograph (Fig. 3 in Ref. 3) of the depletion zone around an island, whose size is ~1000 Å. They also show that the time scale of this mass movement to form an island is from 2 to 10 sec. This phenomenon of depletion is clearly consistent with the results of our simulation. It takes a few seconds in a highly mismatched system, but is much longer, ~ minutes, in the Ge/Si system; so in this system it is probably masked by the deposition rates used and only two groups have reported seeing it in this system.[8,9] From an energetics perspective, beyond the critical coverage $\theta_c$, as deposition continues, there is much more energy to be gained for the new material to create new 3D islands than to grow existing ones. So there is an increase of island density but little size gain. Our simulation shows that the process of depletion is driven by two factors. First 2D islands are grown well past the size $s_o$ at which 3D islands become energetically favorable. At a distinct critical size $s_c$ determined by kinetics, atoms at the 2D island ends are, as a result of strain, easily promoted up to the next level. Second, strain continues to be adequate to keep island-end atoms mobile even with little further island growth, so that the next higher levels are formed quickly, also at distinct sizes. The process of depletion lasting seconds only, then, is largely the pulling in of existing material to form 3D islands. It is completed when the tallest island is formed. Subsequent growth of these islands is mainly by the formation of new facets. Facet formation is generally much harder than adding atoms to the ends of a 2D island. As can be seen from Fig. 1, the diffusion barrier for an atom at the end of a 2D island of size 10–15 atoms is ~1.5 eV while it is ~1.38–1.4 eV at the bottom edge of a three- to four-level island. This difference translates into a substantial difference in mobility, as we have seen above, so that as long as 2D islands are present, their growth is strongly favored over that of 3D islands. The uniformity of islands at coverage of $\theta_c$ is due to depletion occurring at distinct sizes. The continuing size uniformity coupled with constant mean 3D island size while 3D island density increases, as deposition proceeds, especially for $\theta - \theta_c \leqslant 0.5$,[1] is due to the preferred growth of 2D over that of 3D islands. Chen and Washburn[20] obtained results of continuous increase in the size of 3D islands (see their Fig. 5)

TABLE IV. Growth regimes of various experimental groups.

|  | Leonard[a] | Moison[b] | Gerard[c] | Ruvimov[d] | Polimeni[e] | Solomon[f] |
|---|---|---|---|---|---|---|
| Growth temperature | 530 °C | 500 °C | 520 °C | 480 °C | 420 °C | 500 °C |
| Deposition rate ML/sec | 0.01 | 0.06 | 0.06 | 0.6 | 0.1 | 0.18–0.39 |
| Island uniformity | 24% | 40% |  | $<20\%$ |  |  |
| $\theta_c$ | 1.5 | 1.75 | 1.7 |  |  |  |
| Relative $R/D$ | 1 | 8 | 6 | 120 | 50 | 24–52 |

[a] Reference 1.
[b] Reference 2.
[c] Reference 3.
[d] References 12 and 4.
[e] Reference 14.
[f] Reference 29.

with the rate being the largest at the smallest size. Clearly this can only apply after most of the 2D islands have disappeared.

As we have noted before, there should be a correlation between kinetics and energetics. Our kinetic MC simulation shows that the depletion process begins once strain enables 2D island edge atoms to be mobile enough to be promoted to the upper levels. But this process must be also favored by the energetics; we expect the corresponding 3D island to be energetically more favorable than the 2D island. For the systems studied here we have observed this correspondence. In a previous paper we studied the energetics of $(10n)$ facetted Ge hut clusters on Si substrate,[15] using the atomic configuration of (105) side facets suggested by Mo *et al.*[5] The general conclusion was that taller (105) facetted islands become energetically favorable at smaller sizes than islands with $(10n)$ facets for $n\geqslant7$. (103) facetted islands are excluded because these faces require costly double steps so that they are not observed experimentally. (105) facetted hut clusters become favorable only when they have at minimum, heights of 12 layers[15] (at size $s_o$, say). Islands with lower aspect ratios have to reach greater sizes to become energetically favorable. For systems which are growing with growth front roughness of 1–2ML's,[5] it is then necessary for 2D islands to grow well beyond the size $s_o$ before the transition to a 3D shape can begin. This size $s_c'$ may be comparable to the size at which a *two-level* island first becomes favorable. As we have noted in the simulations above, the actual transition size, $s_c$ is determined by kinetics but this size must be such that $s_c\geqslant s_c'$. So energetics sets the lower size limit at which a 2D–3D transition can occur. In Fig. 2 of Khor *et al.*[24] (111) facetted islands are shown to become energetically favorable at sizes and heights greater than those for (105) islands at size $s_o$. With increasing size, (111) facetted islands quickly become more favorable than the (105) hut clusters. These results are consistent with experimental observations of Hansson *et al.*[6] who obtained (111)-facetted islands under near equilibrium conditions and also the results of Mo *et al.* where macroscopic structures were seen to be the stable ones.

Three groups, Leonard *et al.*[1], Moisson *et al.*,[2] and Gerard *et al.*,[3] observe the presence of a critical coverage $\theta_c$ below which no 3D islands are seen. Leonard *et al.* characterize this transition to be like that of a first-order phase transition. We observe a similar transition in our simulations. However, in contrast to the results above, Polimeni *et al.*[14] report a smooth 2D–3D transition for the growth of InAs on GaAs(001). In Table IV, we compare the growth conditions for the different groups. The growth temperature used by Polimeni *et al.* at 420 °C, is substantially lower than those (500–530 °C) used by the other groups. At these temperature differences, the diffusion rate $D$ could differ by more than an order of magnitude. Assuming a behavior for $D$ to be similar to that observed for Si adatoms on Si(001),[18,19] we calculate the ratio $R/D$, where $R$ is the deposition rate. This ratio is $\sim N^3$ ($N=$island density), for the same coverage;[18,19] this tendency to nucleate islands should correlate with growth front roughness. We see from the last row of Table IV, this ratio for Polimeni *et al.* is about 50 times that for Leonard *et al.* Since higher effective deposition rates contribute to rougher growth, it may make level to level transitions less distinct than those we have seen in the simulations above. At some point kinetic roughness at the growth front arising from fast (slow) deposition (diffusion) may mask the phenomenon of depletion and give rise to an apparently smooth 2D–3D transition.

Solomon *et al.*[29] have shown that 3D island density, at fixed coverage and temperature, is increased when either the growth rate $R$ is reduced or the diffusion $D$ is increased (the latter is done by increasing the flux V/III ratio).[29] A related observation is made by Mo and Lagally[23] for Ge/Si(100) growth. When they deposited Ge at 850 K, they found the concentration of macroscopic clusters to be higher than that at $T<800$ K. This result is unexpected for as we have seen above from nucleation theory for regular island growth that island density goes as $R^p/D^p$ where $p$ is positive;[19] but this applies, in our case to the $2D$ islands. Increasing deposition rate or decreasing diffusion then increases the 2D island density and correspondingly decreases average 2D island size at a given coverage. Assuming that there is an average 2D-3D island transition size $s_c$ for the growth regimes of Solomon *et al.* (this must be the case since they observe the constant 3D island diameter throughout their experiments), this means that fewer 2D islands reach this size at that coverage. The density of $3D$ islands then increases by reducing growth rate or increasing $D$. We see from Table IV, that its relative $R/D$ ratio of 24–52 may put it in a ''rough'' growth regime closer to that of Polimeni *et al.* than to that of Leonard *et al.*, so that the existence of a sharp $\theta_c$ is uncertain.

In Table IV it is interesting to note that even for a relative $R/D=120$, much larger than that of Polimeni *et al.*,[14] Ruvimov *et al.*[12] still observed 3D island size uniformity $<20\%$. This is true even when their observations were carried out for coverages of $\theta=2-4$ ML's, which is much greater than the

$\theta_c$ of Leonard *et al.* They did not specifically study if the 2D-3D transition is sharp or smooth.

We have noted above that depletion is seen to occur on a timescale of minutes in Ge/Si systems compared to seconds for InAs/GaAs systems. We should expect to see results in the former similar to those observed for the latter if the deposition rates and growth temperatures are appropriately scaled. Shklyaev *et al.*[8] have carried out growth experiments of Ge on Si(111) at small increments of coverage; they used a growth rate of 0.004 bilayer (BL)/sec and a temperature of 480 °C ($R/D \sim 0.8$, see Table IV). They observed the growth of two types of islands which were called large flat islands and 3D islands. The latter appear ''abruptly,'' there being a distinct jump in 3D island density over a growth interval of 0.1 BL. Much of the material for the formation of these islands come from the substrate. Annealing experiments suggest that this depletion occurs over a time period of about 10 min. They did not measure island size distribution but their Fig. 1 shows 3D island images which appear quite uniform in size. Many of the experiments for the growth of Ge on Si were carried out with quite high relative $R/D$ values, for example, $R/D=7,7,$ and 4 for Voigtlander *et al.*,[9] Medeiros-Ribeiro *et al.*[13] and Kastner *et al.*,[9] respectively. It is in this growth regime that the last two groups observed rectangularly shaped hut islands.[5] Voigtlander *et al.* saw the aspect ratio of a single island change over a coverage interval of ~1 BL (20 min), indicating that depletion probably takes that long.

As noted above, the time ($\tau$) for depletion increases with decreasing lattice mismatch $x$. In general it probably goes as $\tau \sim x^{-\eta} D^{-\gamma}$, where $\eta$ and $\gamma$ are some positive constants. We have seen above that depletion occurs over a large range of growth conditions as determined by the ratio $R/D$. We would expect depletion to fail to occur only when it is completely overwhelmed by deposition, that is, when $1/R \gg \tau$, or when $x \ll R^{1/\eta} D^{-\gamma/\eta}$. This must be the condition for smooth (nonislanding) growth at low temperatures or high deposition rates. The relationship must be applicable to the temperature-concentration phase curve, delineating smooth from rough growth for the deposition of $\mathrm{Si}_{1-x}\mathrm{Ge}_x$ on Si(001) obtained by Bean *et al.*[30] In Fig. 3 of Khor *et al.*,[24] a replot of the experimental data of Fig. 1 of Bean *et al.* shows a linear relationship of ln($x$) versus $1/T$, (except for the point at $x=1$ where a minor temperature change from 550 to 527 °C would put the point on the line), which supports this conclusion.

## V. CONCLUSION

In conclusion, we find that in general, for strained heteroepitaxial growth of semiconductors, there exists an effective kinetic barrier for the 2D to 3D transition. Under conditions of slow deposition and fast diffusion, islands initially grow two dimensionally to a size $s_c \gg s_o$ well beyond the size $s_o$ at which a 3D island *first* becomes energetically favorable. At this size $s_c$, atoms at the edge of the 2D island become mobile as a result of strain, and are promoted to the next level. Promotion of atoms to the next levels occurs in quick succession because edge atoms continue to be highly strained and so remain mobile. The process of depletion is completed when the island attains its highest aspect ratio. This size $s_c$ is sharply defined, and there is a correlation with energetics. This is a robust result that should apply to a wide range of semiconductor systems. For highly mismatched systems, it is the underlying microscopic reason for the uniformity in the sizes of islands seen experimentally. It is consistent with other experimental results such as the increase in island density with coverage with no corresponding increase in size, the phenomenon of depletion, the (initially unexpected), result that island density increases with reduced growth rate or enhanced diffusion.

Much work has been done on 1D simulations of molecular-beam epitaxy (MBE) growth. Our MC simulations are carried out in the same spirit. The test of the usefulness of the $(1+1)$ simulation is whether or not conclusions reached can be extended to the $(2+1)$ situation; the microscopic mechanism for fast depletion behavior that we described above is surely applicable to the higher dimension—edge atoms experience increasing stress with growth of 2D islands until they become mobile enough to be promoted to the higher levels, in turn rendering newly exposed edge atoms mobile until the whole process of building a 3D island is completed. We do not expect periphery motion, i.e., atoms that ''go around'' the islands to affect the applicability of the idea to $(2+1)$ growth.

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[1] D. Leonard, M. Krishnamurthy, C.M. Reaves, S.P. Denbars, and P.M. Petroff, Appl. Phys. Lett. **63**, 3203 (1993); D. Leonard, K. Pond, and P.M. Petroff, Phys. Rev. B **50**, 11 687 (1994).

[2] J.M. Moison, F. Houzay, F. Barthe, and L. Leprince, Appl. Phys. Lett. **64**, 196 (1994).

[3] J.M. Gerard, J.B. Genin, J. Lefebvre, J.M. Moison, N. Lebouche, and F. Barthe, J. Cryst. Growth **150**, 351 (1995); J.M. Gerard, O. Cabrol, J.Y. Marzin, N. Lebouche, and J.M. Moison, Mater. Sci. Eng., B **37**, 9 (1996).

[4] M. Grundmann, J. Christen, N.N. Ledentsov, J. Bohrer, D. Bimberg, S.S. Ruvimov, P. Werner, U. Richter, U. Gosele, J. Heydenreich, V.M. Ustinov, A.Yu. Egorov, A.E. Zhukov, P.S. Kop'ev, and Zh.I. Alferov, Phys. Rev. Lett. **74**, 4043 (1995).

[5] Y.-W. Mo, D.E. Savage, B.S. Swartzentruber, and M.G. Lagally, Phys. Rev. Lett. **65**, 1020 (1990).

[6] P.O. Hansson, M. Albrecht, H.P. Strunk, E. Bauser, and J.H. Werner, Thin Solid Films **216**, 199 (1992).

[7] M. Sopanen, H. Lipsanen, and J. Ahopelto, Appl. Phys. Lett. **67**, 3768 (1995).

[8] Alexander A. Shklyaev, M. Shibata, and M. Ichikawa, Phys. Rev. B **58**, 15 647 (1998); Surf. Sci. **416**, 192 (1998).

[9] Martin Kastner and Bert Voigtlander, Phys. Rev. Lett. **82**, 2745 (1999); Bert Voigtlander and Andre Zinner, Appl. Phys. Lett. **63**, 3055 (1993).

[10] V.A. Shchukin, N.N. Ledentsov, P.S. Kop'ev, and D. Bimberg, Phys. Rev. Lett. **75**, 2968 (1995).

[11] N. Moll, M. Scheffler, and E. Pehlke, Phys. Rev. B **58**, 4566 (1998).

[12] S. Ruvimov, P. Werner, K. Scheerschmidt, U. Gosele, J. Heydenreich, U. Richter, N.N. Ledentsov, M. Grundmann, D. Bimberg, V.M. Ustinov, A.Yu. Egorov, P.S. Kop'ev, and Zh.I. Alferov, Phys. Rev. B **51**, 14 766 (1995).

[13] G. Medeiros-Ribeiro, T.I. Kamins, D.A.A. Ohlberg, and R. Stanley Williams, Phys. Rev. B **58**, 3533 (1998).

[14] A. Polimeni, A. Patane, M. Capizzi, F. Martelli, L. Nasi, and G. Salviati, Phys. Rev. B **53**, R4213 (1996).

[15] K.E. Khor and S. Das Sarma, Phys. Rev. B **49**, 13 657 (1994); **50**, 18 382 (1994).

[16] C. Priester and M. Lannoo, Phys. Rev. Lett. **75**, 93 (1995).

[17] K.E. Khor, Bull. Am. Phys. Soc. **40**, 621 (1995).

[18] Y.-W. Mo, J. Kleiner, M.B. Webb, and M.G. Lagally, Surf. Sci. **268**, 275 (1992).

[19] J.A. Venables, Surf. Sci. **299/300**, 798 (1994).

[20] Y. Chen and J. Washburn, Phys. Rev. Lett. **77**, 4046 (1996).

[21] W. Theis and R.M. Tromp, Phys. Rev. Lett. **76**, 2770 (1996).

[22] Jacques Amar and Fereydoon Family, Phys. Rev. Lett. **74**, 2066 (1995).

[23] Y.-W. Mo and M.G. Lagally, Mater. Sci. Eng., B **14**, 311 (1992); J. Knall and J.B. Pethica, Surf. Sci. **265**, 156 (1992).

[24] K.E. Khor and S. Das Sarma, J. Vac. Sci. Technol. B **15**, 1051 (1997).

[25] B.G. Orr, D. Kessler, C.W. Snyder, and L. Sander, Europhys. Lett. **19**, 33 (1992).

[26] Deepak Srivastava and Barbara Garrison, Phys. Rev. B **46**, 1472 (1992).

[27] A.H. Harker, K. Pinardi, S.C. Jain, A. Atkinson, and R. Bullough, Philos. Mag. **A71**, 871 (1995).

[28] S. Christiansen, M. Albrecht, S. Strunk, and H.J. Maier, Appl. Phys. Lett. **64**, 3617 (1994).

[29] G.S. Solomon, J.A. Trezza, and J.S. Harris, Jr., Appl. Phys. Lett. **66**, 3161 (1995).

[30] J.C. Bean, T.T. Sheng, L.C. Feldman, A.T. Fiory, and R.T. Lynch, Appl. Phys. Lett. **44**, 102 (1984).