Quantum Dot Self-assembly in Growth of Strained-Layer Thin Films: a Kinetic Monte-Carlo Study

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Abstract

We use Monte-Carlo 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.
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) 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. ‘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 and finally to even larger but dislocated islands. It is possible to bypass the hut cluster stage by growing at slightly higher temperatures. Another much studied system is the growth of InAs on GaAs(001) (mismatch ∼7%); here the particular interest is in the uniformity in the size of islands formed. This uniformity, with dispersions of 10% in height and 7% in diameter of the islands at the initial stages of formation, decreases with coverage \( \theta \). There seems to be a distinct coverage \( \theta_c \) 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. shows that by substantially increasing the deposition rate, for example, it is possible to shift it from 1.7ML to 1.95ML. There is evidence that the material to build an island comes mainly by depleting its immediate environment: the thickness of the InAs layer before landing occurs, which is between one and two MLs, is reduced to one ML in the immediate region surrounding the island. 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. Island shapes and sizes can depend on growth conditions, so that kinetic effects are important. Under much higher deposition rates and lower growth temperatures than those used by Leonard et al. and Moisson et al., Ruvimov et al. found that islands also exhibit size uniformity (≤ 20%); however, island size increases with coverage. While Moisson et al. observed (104) and (110) facets on the islands, Grundmann et al. and Ruvimov et al. saw only (110) facets; Moll et al. showed that the equilibrium shape of an InAs island involves (111), (1\bar{1}1) 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 \( \theta \). 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 \( \theta_c \). There is also the phenomenon of fast depletion (of the order of seconds), where a 3D island is created quickly (compared to a deposition rate of .01ML/sec) 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 ∆\( \theta \)∼ 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), 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 and recent growth experiments were carried out at typical deposition rates of a few ML’s/sec. Under these growth conditions, even the sharp 2D-3D transition may disappear.

In this study then we focus on the early stages of growth for thin films which grow in the Straniski-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, 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. 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. 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) 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, who used a critical nucleus of $i = 1$ in the scaling function $\Phi(N/\bar{N})$ in fitting the island density results of Leonard. Island nucleation of Ge on Si(001) should be similar (in both cases the dimer is the stable nucleus). 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, after growth of about 3ML of Ge on Si at 500°C, a growth front roughness of three layers over an area of 60nm x 60nm. Gerard et al. observe one layer roughness over extensive 2D areas ($\sim 2000\AA^2$) for the growth of InAs on GaAs(001) at 520°C. We stress that $s_c >> s_o$, 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) faceted 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. 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 non-equilibrium 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. THE 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 activation energy depends on the bonding environment and the elastic energy associated with strain. The hopping rate is given by the expression,

$$R_n = R_o \exp \left( \frac{-E}{k_BT} \right),$$

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., 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)x7(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,

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

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. 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 750K. This is about 50-100 times the width of the islands that form. Using diffusion rates from Mo et al and others and experimental deposition rates and island sizes, we get comparable results of the ratio diffusion distance/island size $\sim 100$. We choose the spring constant $k = 200\text{eV} \sim 200$ times the diffusion barrier for a single adatom, and a deposition rate of $0.01$-$0.2$ MLs/sec. We carried out simulations for strained-layer lattices with misfits of $0-7\%$, at temperatures of 700 to 800K. We start with systems at thicknesses of 11MLs, 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, varying the diffusion barrier for atoms at the ends of islands, $E_{\text{end}}$, from 1.3 to 1.8eV, for temperatures $T$, from 700-800K, and deposition rates 0.1-0.2 MLs/sec and over coverages of $\theta$ from .5 to 0.8. In Table I, we display the results for two growth temperatures, 750 and 800K, with a deposition rate of 0.2MLs/sec, a system size of $10^4$ cells and a coverage $\theta \sim 0.6$. We calculate a roughness index (R.I.) as the percentage of sites in islands, which have heights $> 1$, i.e., R.I. is a rough measure of the deviation from “two-dimensionality” (one-dimensionality in our simulations) in the islands. For $E_{\text{end}} \geq 1.5\text{eV}$ growth is smooth, islands are flat (very small R.I.), but growth is distinctly rough for $E_{\text{end}} \leq 1.4\text{eV}$, there being much larger proportions of islands with 2 or more layers in height. Clearly the transition from smooth to rough growth is sharp.

Next we look at island-end energies $E_{\text{end}}$ for some island configurations when elastic interactions are included. Specifically we carry out calculations for misfits of 5%. In Fig. I, we plot $E_{\text{end}}$ against island volume (number of atoms), for seven island configurations, comprising (a) 1-level islands ($h=1$), (b) 2-level islands (with 1 atom ($h=2a$) and 2-atoms ($h=2b$) on the second level, (c) 3 level islands with 1 ($h=3a$) and 2 atoms ($h=3b$) on the third level and 3 and 4 atoms respectively on the second and (d) 4 and 5 level islands each with 1 atom on the top level ($h=4,5$ 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 $E_{\text{end}} < 1.5\text{eV}$ as the condition for rough growth, then islands with volumes $> 15$ will have end atoms with diffusion barriers $< 1.5\text{eV}$ for all the consecutive 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 $E_{\text{end}}$ increases 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.

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 factor $F_{\text{end}} = 1.0, 1.2$ and $1.5$ on the first level end atoms of islands. It is known that there is tremendous strain at the foot of islands. Our results are not particularly sensitive to variations in this strain, aside from making islands a little smaller as $F_{\text{end}}$ is increased. In Fig. 2, we follow the development of a single island over a growth period of about 0.2ML (<3secs.). Figs. 2a,b,c show a 1-level(2D) island of volume 19 atoms being folded up into a 2-level island in 0.5 secs. The material for this 2-level island (volume=20) comes almost completely from the original 1-level island. In the rest of Figs. 2d,e,f and g, we see similarly rapid buildups of the third and fourth levels after a brief waiting period. The whole process starting from Fig. 2a to Fig. 2g takes less than 3 secs. The bulk of the material ( ~ 80% for the 3-level island and ~ 65% for the 4-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). Fig. 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).

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 2 atoms in each case, showing that transitions occur at sharp distinct sizes.

We plot in Fig. 4 the total number of islands with 3 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. observed this experimentally and fitted the island density $\rho_{\text{isl}}$ with the function $\rho_{\text{isl}} = \rho_o(\theta - \theta_c)$. 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 2 or more levels instead of the $\geq 3$ levels we have chosen above.

In Table III, we show the energies of islands of various configurations comprising 2, 3 and 4 levels, relative to the energies of their corresponding 1-level configuration at the same volume. We see that the first energetically favorable 2-level island is the one whose volume is 8 atoms with a configuration of 3 atoms on the second level and 5 on the first. 3-level islands become favorable at a volume of about 12 atoms but for this volume the 2-level configuration has the best energetics. 3 and 4-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 2-level island becomes
energetically favorable at a volume of 8 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 $s_c >> s_o$ kinetically driven scenario we discussed before. Note also that because of the small size of the islands we encounter here, it is the 2-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 III. 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[1,28]. The first three groups concluded that there is depletion-like 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 $\sim 1000\AA$. They also show that the timescale of this mass movement to form an island is from 2 to 10 seconds. 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, $\sim$-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. Secondly, 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. [4], the diffusion barrier for an atom at the end of a 2D island of size 10-15 atoms is $\sim 1.5\text{eV}$ while it is $\sim 1.38 – 1.4\text{eV}$ at the bottom edge of a 3-4 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 $\theta - \theta_c \leq 0.5$[4], is due to the preferred growth of 2D over that of 3D islands. Chen and
Washburn\textsuperscript{20} obtained results of continuous increase in the size of 3D islands (see their Fig. 5) 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) faceted Ge ‘hut’ clusters on Si substrate\textsuperscript{15} using the atomic configuration of (105) side facets suggested by Mo \textit{et al.}\textsuperscript{5} The general conclusion was that taller (105) faceted islands become energetically favorable at smaller sizes than islands with (10n) facets for n≥7. (103) faceted islands are excluded because these faces require costly double steps so that they are not observed experimentally. (105) faceted hut clusters become favorable only when they have at minimum, heights of 12 layers\textsuperscript{15} (at size s\textsubscript{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 1-2MLs\textsuperscript{5}, it is then necessary for 2D islands to grow well beyond the size s\textsubscript{o} before the transition to a 3D shape can begin. This size s\textsubscript{c} may be comparable to the size at which a 2-level island first becomes favorable. As we have noted in the simulations above, the actual transition size, s\textsubscript{c} is determined by kinetics but this size must be such that s\textsubscript{c} ≥ s\textsubscript{c}'. So energetics sets the lower size limit at which a 2D–3D transition can occur. In Fig. 5 (see Khor \textit{et al.}\textsuperscript{23}) (111) faceted islands are shown to become energetically favorable at sizes and heights greater than those for (105) islands at size s\textsubscript{o}. With increasing size, (111) faceted islands quickly become more favorable than the (105) hut clusters. These results are consistent with experimental observations of Hansson \textit{et al.}\textsuperscript{6} who obtained (111)-facetted islands under near equilibrium conditions and also the results of Mo\textit{et al.}\textsuperscript{6} where macroscopic structures were seen to be the stable ones.

Three groups, Leonard \textit{et al.}\textsuperscript{1}, Moisson \textit{et al.}\textsuperscript{2} and Gerard \textit{et al.}\textsuperscript{3}, observe the presence of a critical coverage θ\textsubscript{c} below which no 3D islands are seen. Leonard \textit{et al.} characterise 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 \textit{et al.}\textsuperscript{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 \textit{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)\textsuperscript{18,19}, we calculate the ratio R/D, where R is the deposition rate. This ratio is ∼ N\textsuperscript{3} (N= island density), for the same coverage\textsuperscript{13,19}; this tendency to nucleate islands should correlate with growth front roughness. We see from the last column of Table IV, this ratio for Polimeni \textit{et al.} is about 50 times that for Leonard \textit{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 \textit{et al.}\textsuperscript{28} 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.) A related observation is made by Mo and Lagally\textsuperscript{22} for Ge/Si(100) growth. When they deposited Ge at 850K, they found the concentration of macroscopic clusters to be higher than that at $T<800K$. 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\textsuperscript{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 \textit{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 \textit{et al} than to that of Leonard \textit{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 \textit{et al}\textsuperscript{14}, Ruvimov \textit{et al}\textsuperscript{12} still observed 3D island size uniformity $<20\%$. This is true even when their observations were carried out for coverages of $\theta = 2$-4MLs, which is much greater than the $\theta_c$ of Leonard \textit{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 \textit{et al}\textsuperscript{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$^\circ$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 minutes. 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 \textit{et al}, Medeiros-Ribeiro \textit{et al}\textsuperscript{8} and Kastner \textit{et al}\textsuperscript{9} respectively. It is in this growth regime that the last two groups observed rectangularly shaped ‘hut’ islands. Voigtlander \textit{et al} saw the aspect ratio of a single island change over a coverage interval of $\sim$1BL(20min), 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 (non-islanding) 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 Si$_{1-x}$Ge$_x$ on Si(001) obtained
by Bean et al. In Fig. 6 (Khor et al.), 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 $s_c >> s_o$ 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.

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TABLES

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

| $E_{\text{end}}$ | $\theta$ | R.I. | $\theta$ | R.I. |
|-----------------|----------|------|----------|------|
| 0.3             | 0.56     | 29.3 | 0.56     | 41.0 |
| 0.4             | 0.63     | 7.4  | 0.65     | 17.6 |
| 0.5             | 0.61     | 1.3  | 0.63     | 3.2  |
| 0.6             | 0.64     | 1.1  | 0.62     | 1.3  |

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 |

TABLE III. Energetics of islands of various configurations relative to the energy of the corresponding 1 level island at the same volume.

| volume | 2-level | 3-level | 4-level | $\Delta E$  |
|--------|---------|---------|---------|-------------|
| 4      | 1,3     |         |         | -0.58eV     |
| 6      | 2,4     |         |         | -0.18eV     |
| 8      | 3,5     | 1,3,5   |         | -0.24eV     |
| 9      | 5,7     |         |         | 0.72eV      |
| 12     | 2,4,6   |         |         | 0.42eV      |
| 14     | 6,8     | 3,5,7   |         | 0.97eV      |
| 15     | 11,13   | 6,8,10  |         | 1.06eV      |
| 24     | 11,13   | 3,5,7,9 |         | 1.79eV      |
| 24     | 6,8,10  |         | 3,5,7,9 | 2.72eV      |
| 28     | 13,15   | 4,6,8,10|         | 2.6eV       |
| 28     |         |         |         | 1.28eV      |
|                          | Leonard<sup>a</sup> | Moison<sup>b</sup> | Gerard<sup>c</sup> | Ruvimov<sup>d</sup> | Polimeni<sup>e</sup> | Solomon<sup>f</sup> |
|--------------------------|---------------------|-------------------|-------------------|---------------------|----------------------|----------------------|
| 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                  | .18-.39              |
| Island uniformity        | 24%                 | 40%               | <20%              |                     |                      |                      |
| $\theta_c$               | 1.5                 | 1.75              | 1.7               | -                   | -                    | -                    |
| Relative R/D             | 1                   | 8                 | 6                 | 120                 | 50                   | 24-52                |

<sup>a</sup>Reference<sup>1</sup>.  
<sup>b</sup>Reference<sup>2</sup>.  
<sup>c</sup>Reference<sup>3</sup>.  
<sup>d</sup>Reference<sup>12</sup>,<sup>4</sup>.  
<sup>e</sup>Reference<sup>14</sup>.  
<sup>f</sup>Reference<sup>28</sup>.
FIGURES

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

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 2-level island in (b) and (c). Figs. (d), (e), (f) and (g) show rapid promotion to the upper layers.

FIG. 3. Height and width distributions of islands at coverages $\theta=0.4\text{ML},0.67\text{ML},0.79\text{ML}$ and $0.96\text{ML}$.

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

FIG. 5. Island energies in meV/atom versus cluster size. Dashed line: rebonded (105) islands; light solid: (107) islands; dot-dashed line: (111) islands; heavy solid line: two layer islands. (105),(107) and 2-layer island curves cross the abscissa at points A,B and C respectively.

FIG. 6. Replot of the film morphology curve of Bean et al (Fig. 1 in ref 29), $1/T$, in $^o\text{K}^{-1}\times1000$, versus $\ln(x)$.
