

Mechanism for Coherent Island Formation during Heteroepitaxy

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Abstract. — Monte Carlo simulations are reported for an atomistic model of heteroepitaxial growth. Dislocations are excluded but lattice misfit is assumed to encourage atom detachment from islands by reducing the barrier for this process by a morphology-dependent strain energy. Three-dimensional, coherent islands of nearly uniform size are found to form spontaneously for misfit above a critical value.

The thermodynamic analysis of Bauer [1] established that the stability of three-dimensional (3D) epitaxial islands on top of a uniform wetting layer (Stranski-Krastanov growth mode) can be attributed to the strain induced by lattice constant misfit between a substrate and a dissimilar deposited material. His result that island formation in this situation is linked to the introduction of misfit dislocations at the heteroepitaxial interface has been dogma for over thirty years. But this orthodoxy was overturned with the experimental demonstration by Eaglesham and Cerullo [2] that undislocated (coherent) islands can form readily under appropriate circumstances. They correctly identified the considerable relaxation of strain possible at the free surface of the island as a driving force for island formation. It turns out that such coherent islands can indeed be equilibrium structures [3, 4] but it is evident that kinetic considerations [5] cannot be ignored. Indeed, recent experimental activity has focused precisely on the manipulation of growth kinetics to tailor coherent island size distributions for quantum dot applications [6].

In this paper, we explore the kinetics of 3D coherent island formation with Monte Carlo growth simulations of a solid-on-solid model of heteroepitaxy. The model is a generalization of one we have used previously to study the distribution of two-dimensional (2D) heteroepitaxial

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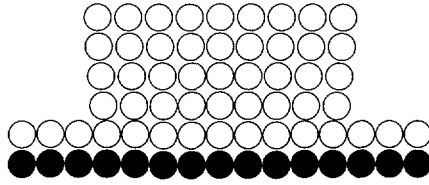


Fig. 1. — Schematic view of strain relief in a coherent island by progressive adjustment of the lattice constant in each layer. The island sits on a strained wetting layer so its lattice constant matches that of the substrate. The dotted lines are perpendicular to the substrate.

island sizes in the submonolayer regime [7]. In contrast to the work of Orr and co-workers [8] (who studied 2D coherent island formation on a one-dimensional substrate), we do not take explicitly into account harmonic forces between atoms. Instead, we use an approximate treatment of misfit strain relaxation at the edge of islands [4] that has been shown to account quantitatively for the height dependence of the lattice constant of coherent islands of Ge/Si(111) [9]. We believe this methodology to be more accurate than one applied to this problem recently by Grandjean and Massies [10].

The basic growth simulation used for studies of homoepitaxial growth [11] proceeds by the random deposition of atoms at an average rate F onto the sites of a square lattice. Neither vacancies nor overhangs are allowed, so every configuration of the lattice is completely described by the column heights at each lattice site. Surface diffusion occurs by the movement of a surface atom from a given site to the top of the column of atoms at a nearest neighbor site. The rate at which *any* surface atom migrates in this way is $D \exp(-nE_N/k_B T)$ where $n = 0, 1, 2, 3, 4$ is the number of lateral nearest neighbors before the hop occurs, $D = (2k_B T/h) \exp(-E_S/k_B T)$ is the hopping rate of a free adatom, E_S is a substrate bond energy, and E_N is an effective pair bond energy.

We retain the lattice character of this growth model to study heteroepitaxy. Misfit dislocations thus are excluded *a priori*. This is not a serious problem so long as any coherent islands that form are small enough to ensure their stability against dislocation generation [4]. A single value of E_S is sufficient since we will always assume that deposition begins after at least one complete wetting layer has formed. We assume that the main effect of misfit strain is to reduce the quantity E_N . Justification for this choice (rather than a change of the free adatom hopping rate) can be found elsewhere [12]. The crucial point is that the strain felt by an atom in a coherent 3D island is not uniform because the island relaxes by the gradual adjustment of its in-plane lattice spacing as its height increases (Fig. 1). The calculations of Ratsch and Zangwill [4] treated the energetic of this problem using a sequence of vertically coupled Frenkel-Kontorova (FK) [13] chains of finite but variable length. In effect, every atom in a given horizontal layer of a 3D island adopts an average relaxed lattice constant that reflects the lateral accommodation of the edge atoms of that layer to the sinusoidal potential established by the layer just beneath. From this it is clear that laterally small islands with relatively more edge atoms relieve strain more efficiently than large islands.

To incorporate heteroepitaxial strain, we reduce the contribution of every lateral nearest-neighbor to the detachment barrier of an atom at a given site i by an amount equal to the local strain energy per atom at that site, i.e., $E_N \rightarrow E_N - \epsilon_i$. Our layerwise FK model [12, 14, 15] provides an estimate of ϵ_i that takes account of the fact that the local strain within the p^{th} 2D layer of an (generally 3D) island depends on both the number of atoms in the 2D layer and the effective misfit of that layer. Figure 2 illustrates this effect qualitatively for several

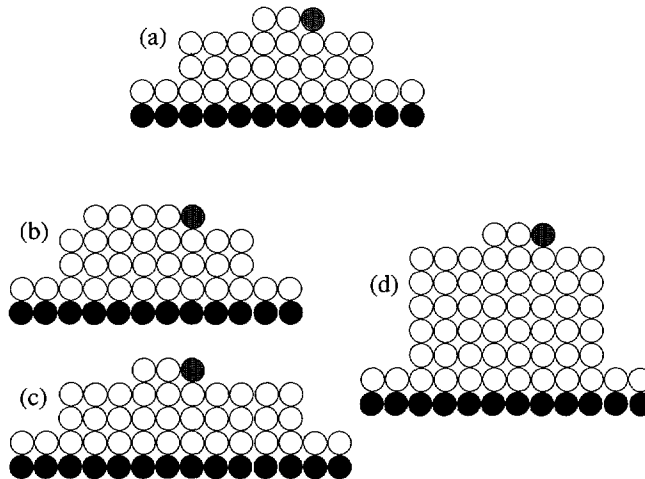


Fig. 2. — Examples of strain and morphology dependent changes to the detachment rate of an atom. Let R_a denote this rate for the grey atom in (a). Although it is at the same height, the corresponding grey atom in (b) detaches at a rate $R_b > R_a$ because it is laterally bonded to a larger 2D island that relieves strain less well. Similarly, $R_c > R_a$ because the topmost layer (of the same size as in (a)) is supported by larger and thus relatively less relaxed layers than in (a). On the other hand, the island in (d) differs from (a) only in its absolute height. The progressive relaxation shown in Figure 1 then implies that $R_d < R_a$.

typical island configurations. Non-trivial bookkeeping is required to implement this scheme because the effective misfit depends on the arrangement of all the other atoms beneath layer p of the island under consideration. Moreover, the required strain energies must be updated as morphological evolution proceeds.

It is not difficult to see that the effects of strain built into our model will lead to 3D islanding. In the submonolayer regime, the island-size-dependent hopping rates lead to the formation of relatively more 2D islands than for the corresponding homoepitaxial situation [7]. These islands grow laterally as deposition proceeds and eventually second layer islands begin to nucleate on top of them. Because the effective misfit decreases as the island height increases, the nearest-neighbor bond strength is reduced by a smaller amount in higher layers. Thus, it is less likely for an atom in a higher layer to detach (compared to an atom in a layer below with the same number of nearest neighbors), so a strained system has a greater tendency to form 3D islands. The energetic preference of one island shape over another is determined by the competition between the decrease of surface energy (here reflected by the gain of nearest neighbor bonds) and the decrease of strain energy. As usual, this thermodynamic effect is reflected indirectly by the kinetics.

The model parameters used for all simulations reported below are $E_S = 1.3$ eV, $E_N = 0.3$ eV, $T = 750$ K, and $F = 0.1$ s⁻¹ so that $D/F \approx 6 \times 10^5$ — a typical value for laboratory studies. Figure 3 shows the surface morphology for 4.5% misfit at four different coverages Θ up to one monolayer (ML). Only a few islands with second layer atoms can be seen until $\Theta = 0.25$ ML. But as growth proceeds further, 3D islands start to form, and at $\Theta = 0.5$ ML cover only slightly more lateral area than at $\Theta = 0.25$ ML. This is in contrast to results with 0% misfit (not shown), where islands at $\Theta = 0.5$ cover twice as much of the substrate as those at $\Theta = 0.25$. After

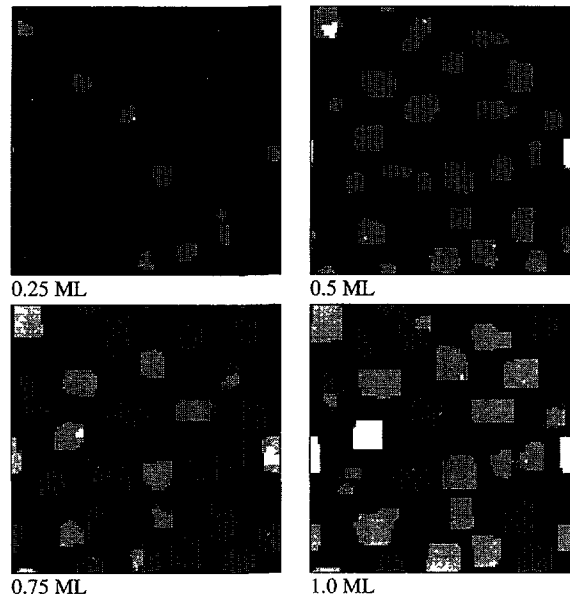


Fig. 3. — Surface morphology with 4.5% misfit at different coverages. The substrate is shown in black and different gray scales correspond to different island heights. For maximum contrast the gray scales do not correspond to the same heights in different panels.

1 ML has been deposited (Fig. 3) 3D islands up to four layers in height have formed. While the 3D islands grow, some of the remaining small 2D islands shrink and eventually dissolve.

The scenario illustrated in Figure 3 occurs for all values of misfit larger than a critical misfit f_c . This critical misfit depends both on the growth conditions and on the material parameters chosen; here, we find $f_c \approx 3\%$. The morphologies for different values of misfit after 2 ML have been deposited are shown in Figure 4. Our results for $f = 0\%$ and $f = 3\%$ are almost indistinguishable because for $f < f_c$ the strain energy is not large enough to drive the system into a 3D growth mode. However, when $f > f_c$, islanding supplants layer-by-layer growth. This is apparent already for $f = 3.5\%$, but the islands are very large and tend to connect at lower layers. Clearly separated coherent islands are seen for larger values of misfit with a typical size that decreases as misfit increases. This observation is in qualitative accord with experimental results for $\text{In}_x\text{Ga}_{1-x}\text{As}/\text{GaAs}(001)$ (Ref. [16]) and can be understood easily within our model. Atoms have to detach from island edges and hop onto a higher layer often enough so that a sufficient adatom density is achieved there to form a new, essentially unstrained nucleus. This small nucleus provides energetically favorable sites for adatom attachment and grows rapidly. For islands of a fixed lateral size the detachment rate increases progressively as misfit increases. Therefore, the lateral island size at the time of nucleation in a higher layer shrinks with increasing misfit as seen in Figure 4.

The transition from 2D growth for $f < f_c$ to 3D growth for $f > f_c$ can also be illustrated by studying the evolution of the surface width defined as $w = \langle \sqrt{h^2 - \bar{h}^2} \rangle$, i.e. the variance of the surface height h . The surface width as a function of coverage is shown in Figure 5. For $f < f_c$, w oscillates exhibiting minima when complete layers are deposited as expected for layer-by-layer growth. However, for $f > f_c$ we find that the surface width increases with

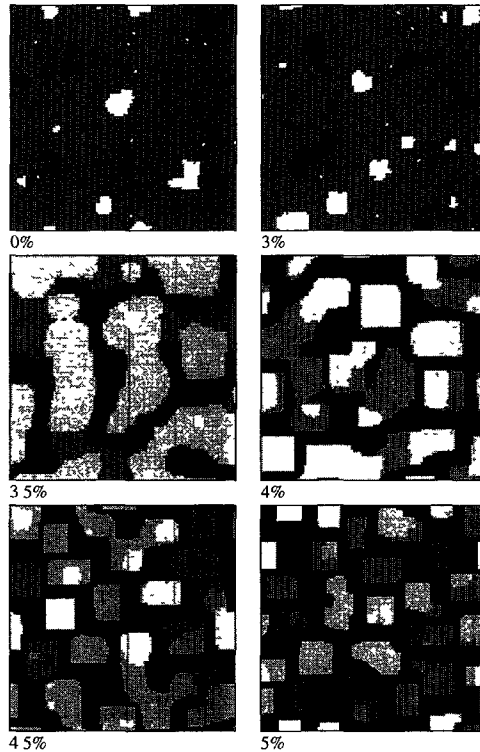


Fig. 4. — Surface morphology after 2 ML have been deposited for different values of misfit. Different gray scales correspond to different layer heights (the lowest exposed layer is shown in black). For maximum contrast the gray scales do not correspond to the same heights in different panels.

coverage according to $w \propto \Theta^\beta$. The best fit of our data for $f = 5\%$ can be obtained for $\beta \approx 0.7$. This is in good agreement with a recent study of $\text{CoSi}_2/\text{Si}(001)$ growth [17] where $\beta \approx 0.66$ has been measured. The inset in Figure 5 shows the dependence of w on misfit at the total coverage of 2 ML and reveals that the transition from 2D to 3D growth is rather sharp with w increasing for $f > f_c$ approximately as $(f - f_c)^{0.5}$. A more detailed study of the character of this transition and the dependence of the critical misfit f_c on the material parameters and growth conditions would be an interesting extension of the present work.

The morphologies in Figure 4 also show that the distribution of lateral sizes of the simulated coherent islands becomes narrower as misfit increases. This occurs despite the fact that our model admits no deformation of the substrate and hence no long-range elastic repulsion between islands [18]. The reason is again the increased detachment rate at island edges. Those atoms that do not hop into an upper layer re-enter the common pool of adatoms on the wetting layer. A communication path thus exists between islands that independently are trying to attain the same strain-minimizing size and shape by mass redistribution. We remark here that a theory of coherent island sizes recently proposed by Priester and Lannoo [19] emphasizes elastic repulsion between islands but tacitly assumes sufficiently rapid detachment processes for local equilibrium to be established.

Close inspection of the gray scales in Figure 4 reveals that our relaxed coherent islands resemble towers as misfit (and coverage) increases. This is an artifact that results entirely

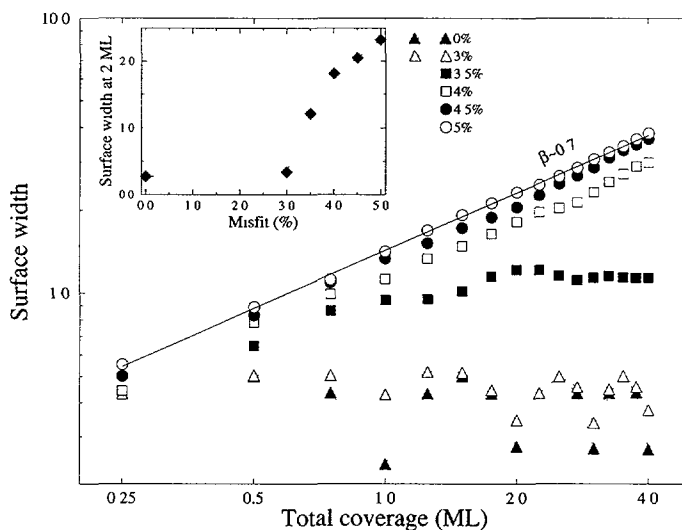


Fig. 5. — Time evolution of the surface width at different values of misfit. The inset shows the misfit dependence of the surface width at 2 ML deposited (cf. Fig. 4). The dotted lines are guides for the eye.

from the simplicity of our simulation model designed to focus exclusively on the effect of a strain-dependent weakening of nearest neighbor bonds. For example, interactions of longer than nearest neighbor range [20] readily could add inclined facets to the free surface of coherent islands. Additional kinetic process not considered here such as enhanced barriers to atomic migration over step edges also are well known to influence island morphologies [21]. However, the simple model proposed here correctly reproduces some of the key features observed in experiments on strained systems.

Acknowledgments

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