ELASTIC ENERGIES OF COHERENT GERMANIUM ISLANDS ON SILICON

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ABSTRACT

Motivated by recent observations of coherent Ge island formation during growth of Ge on Si (100), we have carried out a theoretical study of the elastic energies associated with the evolution of a uniform strained overlayer as it segregates into coherent islands. In the context of a two-dimensional model, we have explored the conditions under which coherent islands may be energetically favored over both uniform epitaxial films and dislocated islands. We find that if the interface energy (for dislocated islands) is more than about 15% of the surface energy, then there is a range of island sizes for which the coherent island structure is preferred.

INTRODUCTION

Until recently, it was generally believed that the growth mode of Ge on Si (100) was of the Stranski-Krastanov (SK) type [1] in which initial layer-by-layer growth is followed by the formation of dislocated islands. (By "dislocated islands" we mean islands at the relaxed Ge lattice constant, with dislocations at the Si/Ge interface to relieve the misfit.) However, Eaglesham and Cerullo [2] have made the surprising discovery that in the initial stages of island growth, the islands can be *coherent* (dislocation-free); only later, as the islands grow in size, do they become dislocated. The coherent islands appear somewhat rounded in shape, and can be up to $\sim$ 1500 Å in size and $\sim$ 500 Å high.

In a possibly related development, Mo *et al.* [3] have presented STM observations of a metastable cluster (island) phase in the same system. In addition to the large dislocated islands expected in SK growth, these workers observed smaller metastable prism-shaped islands of low aspect ratio [formed of (501) facets] and typical dimension $\sim$ 1000 Å. The island surfaces were found to have the relaxed Ge lattice constant to within 1.5% uncertainty. While one might therefore be tempted to conclude that the islands are dislocated in this case, we suggest below that such a conclusion *need not* follow.

Motivated by these developments, we have carried out an analysis of the elastic relaxation energies of coherent islands for a model lattice-misfit system. Our goal will be to determine whether, and under what conditions, the coherent island structure can be energetically preferred in comparison to both the uniformly strained epilayer and dislocated island morphologies.

SPECIFICATION OF THE MODEL SYSTEM

In order to arrive at a tractable model system, we have made a number of simplifying assumptions. Our most severe approximation is that we take the islands to be uniform in one direction along the surface, and work in the 2D plane of cross section. (Thus, the islands are really "ridges.") Also, we assume that Si and Ge are identical in all respects, except that the preferred lattice constant of Ge exceeds that of Si by a reference strain $\epsilon_0 = 0.05$ (i.e., 5%).

We have studied geometries in which the island structure is repeated periodically in the direction along the surface, as shown in Fig. 1(a). The actual calculations are carried out on a system in which the continuous medium has been replaced by a finite-element

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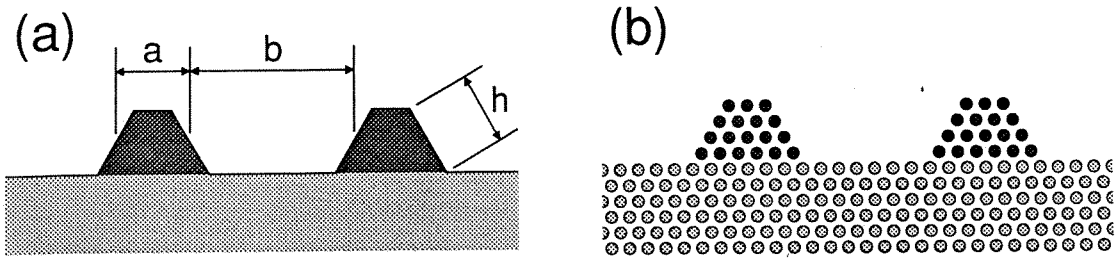

Figure 1. (a) Continuum model of islands, illustrating the definitions of the island half-width a, trough half-width b, and height h. Heavily and lightly shaded regions indicate Ge and Si respectively. (b) Same, but showing hexagonal finite-element discretization actually used in the calculations.

discretization, as shown in Fig. 1(b). The discrete lattice is hexagonal, connected by nearest-neighbor springs of spring constant k, and with lattice spacing d. [Si-Si, Ge-Ge, and Si-Ge bonds have preferred lengths d, $(1+\epsilon_0)d$, and $(1+\epsilon_0/2)d$, respectively.] The resulting elastic constants are isotropic, with Poisson ratio $\nu = 1/4$. For ease of modeling by the discrete lattice, we have only studied islands whose sides are inclined at 60°. With these approximations, the geometry of an island structure can be specified by giving the island height h, island size a, and trough size b, as shown in Fig. 1. Thus, the goal of our calculations will be to obtained the relaxation energy $\Delta E_{\rm el}$ of the structure of Fig. 1(a), in the continuum limit, as a function of h, a, and b.

CALCULATION OF ELASTIC RELAXATION ENERGIES

For given values of h, a, and b, we constructed a series of finite-element representations of decreasing lattice constant d, with up to ~ 60 mesh points along the periodic direction (parallel to the surface). We used a finite number of rows in the vertical direction, with free boundary conditions on the bottom row. However, we kept the number of rows in the vertical direction somewhat greater than the horizontal repeat distance; under these conditions, the results were found to be independent of the depth of the sample, to a high degree of accuracy. The relaxed structure was determined by a simple iterative scheme in which the updates for the displacements were just taken parallel to the calculated forces; typically several thousand such iterations were required to obtain good convergence.

Only the mesh points in the islands were identified as Ge sites (see Fig. 1). In the real system, of course, several atomic layers of Ge do cover the remainder of the surface. While our mesh points do not really correspond to atoms, we nevertheless experimented with setting the top one or two rows of mesh points to be Ge sites. However, we found very little difference in the results. This is easily understood; the presence of an entire Ge row, in the presence of periodic boundary conditions, does not result in any forces on the atoms (in the unrelaxed configuration). Thus, in the limit $\epsilon_0 \to 0$, the results should be rigorously independent of the presence of entire Ge rows (i.e., the effect of such entire rows on $\Delta E_{\rm el}$ enters at cubic and higher order in ϵ_0).

Finally, we obtained the relaxation energy in the continuum limit, $\Delta E_{\rm el}$, by extrapolation from a series of discrete calculations, by fitting the results to a polynomial in d and then taking the limit $d \to 0$.

It is convenient to express the relaxation energy $\Delta E_{\rm el}$ in terms of a dimensionless function of dimensionless arguments

$$F(h_a, h_b) = \frac{2\Delta E_{\rm el}}{k h^2 \epsilon_0^2} \ , \qquad h_a = \frac{h}{a} \ , \qquad h_b = \frac{h}{b} \ . \tag{1}$$

Table I. Calculated dimensionless relaxation energy F and the combination $h_a F$ for a representative sample of geometries specified by $h_a = h/a$ and $h_b = h/b$.

h_a	h_b	F	$\Delta\overline{E}_{\rm el}$	h_a	h_b	F	$\Delta\overline{E}_{\rm el}$
0.083	1.000	3.220	0.268	0.500	1.000	1.492	0.746
0.125	0.125	3.508	0.438	1.000	0.500	0.828	0.828
0.125	1.000	3.020	0.377	1.000	1.000	0.822	0.822
0.250	0.250	2.500	0.625	1.000	2.000	0.804	0.804
0.250	1.000	2.360	0.590	1.500	1.000	0.560	0.840
0.500	0.250	1.532	0.766	2.000	1.000	0.420	0.840
0.500	0.500	1.524	0.762	2.000	2.000	0.412	0.824

Then F remains finite in the limit $\epsilon_0 \to 0$, i.e., negligible non-linear distortion. (Incidentally, we did some tests which indicated that the values of F obtained with our $\epsilon_0 = 0.05$ are in fact very close to those that would apply in the $\epsilon_0 \to 0$ limit.) Moreover, F also remains finite in the limits $h_b \to 0$ (isolated islands), $h_a \to 0$ (isolated troughs), $h_b \to 2$ (V-shaped troughs), and $h_a \to 2$ (inverted V-shaped islands). However, we expect a logarithmic singularity when $h_b \to 0$ and $h_a \to 0$ simultaneously. In this limit (which can be thought of as $h \to 0$ for fixed a and b), the problem can be mapped onto an array of line forces of alternating sign acting parallel to the surface. This problem has been solved in the context of a theory of stress domains on Si (100) by Alerhand *et al.* [4], [5]. Thus in this limit we expect

$$\lim_{h_a,h_b\to 0} F(h_a,h_b) = c_0 + \frac{2\sqrt{3}}{\pi} \ln\left[\frac{1}{\pi}\frac{h_a+h_b}{h_a h_b} cos\left(\frac{\pi}{2}\frac{h_a-h_b}{h_a+h_b}\right)\right] . \qquad (2)$$

Some of the results of our numerical calculations are given in Table I. A notable feature of these results is the asymmetry under the interchange $h_a \leftrightarrow h_b$ (islands $\leftrightarrow$ troughs), which is a symmetry of Eq. (2). The relaxation energy is greater for a trough configuration (e.g., $h_a = 1/2$, $h_b = 1$) than for the corresponding island configuration (e.g., $h_a = 1$, $h_b = 1/2$). This might have been expected: while the forces that result from the material stresses are similar in both cases, the trough acts like a region of enhanced elastic compliance, and the island like one of reduced compliance, in the vicinity of the forces. However, the magnitude of the asymmetry is striking. For example, starting from $h_a = h_b = 1$, the change in F as we let $h_a \to 0$ is approximately two orders of magnitude greater than the change as $h_b \to 0$. This indicates that the repulsive interaction between islands is very much weaker than would be the repulsive interaction between troughs of similar aspect. We also find that the departure from the limit expressed in Eq. (2) is so rapid that this limit is *not* a useful starting point for understanding the energetics of islands.

Returning to the energetics of islands, a useful way of expressing the results is to calculate the ratio of the relaxation energy to the maximum relaxation energy possible in a fully relaxed (dislocated) island of the same dimensions:

$$\Delta\overline{E}_{\rm el} = \frac{2\Delta E_{\rm el}}{k\epsilon_0^2 h a} = h_a F(h_a,h_b) . \qquad (3)$$

These results are also given in Table I. Moreover, we have extrapolated our results to the $h_b \to 0$ axis, corresponding to the case of an isolated island, by assuming an approximately

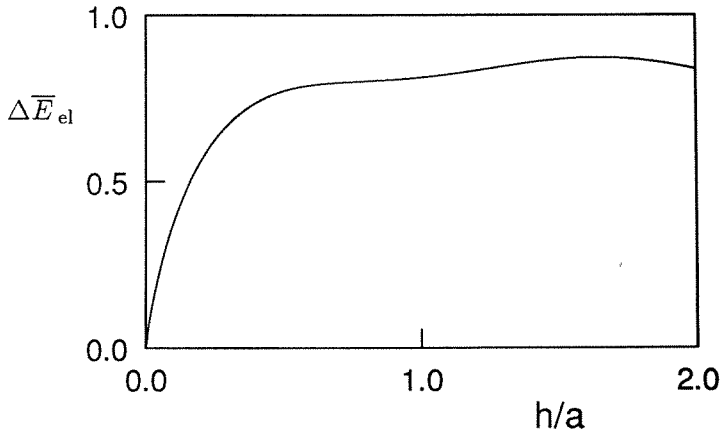

Figure 2. Reduced elastic relaxation energy $\Delta\overline{E}_{el}$ of an isolated island as a function of $h_a = h/a$ as defined in Fig. 1. The maximum value $\Delta\overline{E}_{el} = 1$ would correspond to a fully relaxed (e.g., dislocated) island.

parabolic dependence upon h_b for $h_b \leq h_a$. (Because of the weakness of the dependence of $\Delta\overline{E}_{el}$ upon h_b, the details of this extrapolation procedure are not very important.) We then fitted the resulting $F(h_a) = F(h_a, 0)$ to a function of the form

$$F(h_a) = -\frac{2\sqrt{3}}{\pi}\ln(h_a) + c_0 + c_1 h_a + c_2 h_a^2 + c_3 h_a^3 + c_4 h_a^4 \ . \tag{4}$$

(The first two terms are required for consistency with Eq. (2) in the limit $h_a \to 0$.) We obtained the best overall fit with $c_0 = 1.470$, $c_1 = -2.601$, $c_2 = 3.059$, $c_3 = -1.315$, and $c_4 = 0.2$. The resulting function $\Delta\overline{E}_{el}(h_a) = h_a F(h_a)$ is plotted in Fig. 2.

Recalling that $\Delta\overline{E}_{el}$ has the limiting values 0 and 1 for completely unrelaxed and completely relaxed (dislocated) islands respectively, we notice immediately that even islands of rather flat aspect ratio can relieve most of their strain energy by relaxing in the coherent mode. For example, an island with $h_a = h/a = 1/4$ can get rid of approximately 60% of its strain energy by relaxing *without* the introduction of dislocations. Moreover, for the same case ($h_a = 1/4$) the strain at the center of the top surface of the island is found to be 3.6%, and averaged over the entire top surface it is 4.1%, to be compared to the 5% assumed for completely relaxed Ge. This suggests that the islands observed in STM by Mo *et al.* are *most likely coherent after all.*

The above results suggest that coherent islands may actually be energetically favored under some conditions, since they are able to relieve much of the strain energy without paying the cost of introducing dislocations. Whether this is in fact possible is explored in the next section.

COMPARISON OF ENERGETICS OF THREE GROWTH MODES

In this section we compare the energy cost of depositing a given quantity of Ge in three different ways: as a single dislocated island (DI), as a single coherent island (CI), and incorporated into a uniformly strained coherent overlayer (UF, for "uniform film"). The quantity of Ge to be deposited is represented by its cross-sectional area A_{Ge} (for our islands, $A_{Ge} = \frac{\sqrt{3}}{2}ha$). In addition to the elastic relaxation energy ΔE_{el} discussed above,

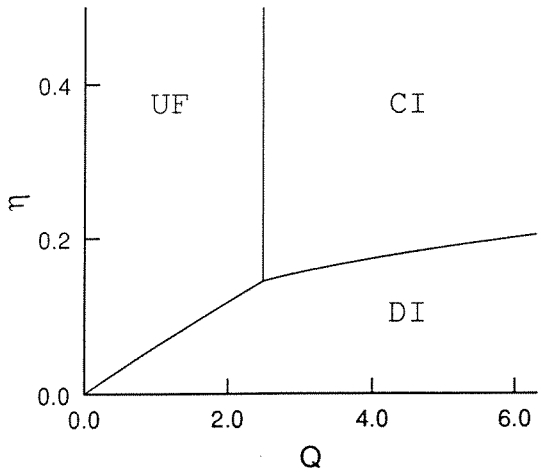

Figure 3. Phase diagram showing the preferred morphology as a function of amount of Ge deposited (horizontal axis) and the ratio of interface to surface energy (vertical axis). Labels DI, CI, and UF refer to "dislocated island," "coherent island," and "uniform film," respectively.

we suppose that there is an energy cost per unit length γ associated with the additional surface exposed on islands, and a similar cost λ associated with the dislocation density which must exist at the dislocated Si-Ge interface. Then the energies of these three modes of deposition are given by

$$E_{\rm DI} = \gamma h + \lambda\left(a + \frac{h}{2}\right) \ , \qquad E_{\rm CI} = \gamma h + \frac{1}{\sqrt{3}} k\epsilon_0^2 A_{\rm Ge} - \Delta E_{\rm el} \ , \qquad E_{\rm UF} = \frac{1}{\sqrt{3}} k\epsilon_0^2 A_{\rm Ge} \tag{5}$$

Introducing dimensionless quantities η (ratio of interface to surface energy), Q (quantity of Ge deposited), and $\overline{E}$ (reduced energy, following Eq. (3)) via

$$\eta = \lambda/\gamma \ , \qquad Q = \frac{2}{\sqrt{3}}\left(\frac{k\epsilon_0^2}{\gamma}\right)^2 A_{\rm Ge} \ , \qquad \overline{E} = \frac{\sqrt{3}E}{\epsilon_0^2 k A_{\rm Ge}} \tag{6}$$

we then obtain

$$\overline{E}_{\rm DI} = (2+\eta)\sqrt{\frac{h_a}{Q}} + 2\eta\frac{1}{\sqrt{h_a Q}} \ , \qquad \overline{E}_{\rm CI} = 2\sqrt{\frac{h_a}{Q}} + 1 - \Delta\overline{E}_{\rm el} \ , \qquad \overline{E}_{\rm UF} = 1 \ . \tag{7}$$

For the two kinds of islands, we minimize the energy with respect to variation in the aspect ratio h_a (holding Q and η constant). (For the case of coherent islands, this is done numerically using the function $\Delta\overline{E}_{\rm el}$ obtained earlier.) Then the reduced energies in Eq. (7) become functions of Q and η only.

In Fig. 3 we have constructed a phase diagram showing which of the three morphologies is energetically preferred, for given values of Q and η. Note that η is a *material property* (the ratio of the dislocated-interface energy to the surface energy), whereas Q is a measure of the *amount of Ge deposited.* We find that for small η, the system makes a transition directly from the uniform film to the dislocated island morphology with increasing Q. In contrast, for η above a critical value $\eta_c = 0.145$, we find that *coherent islands are favored* for intermediate values of Q. There is first a transition from uniform film to coherent island at $Q_c = 2.49$, and then a second transition to an incoherent island at a larger Q which depends upon η. (Both transitions are discrete in our model, but the second CI $\rightarrow$ DI transition may well be continuous in the real physical system.)

We can get an order-of-magnitude estimate of the physical value of η by assuming that the interface and surface energies are $\lambda = \epsilon_0 E_{\rm db}/a_0$ and $\gamma = E_{\rm db}/a_0$ respectively,

where a_0 is a lattice spacing and $E_{\rm db}$ is the energy of a dislocation-core or surface dangling bond. Then $\eta \approx \epsilon_0 = 0.05$, which is below the critical value, but only by a factor of ~ 3. Moreover, in a real three-dimensional analysis, λ would increase (because dislocations would run in both directions), and so would $\Delta E_{\rm el}$ (since coherent islands could relax more efficiently). Thus, given the numerous approximations that have been made, we think it is quite plausible that the real Si/Ge system may be in the $\eta > \eta_c$ regime.

DISCUSSION

While the model presented here involves many simplifying assumptions, we expect the main qualitative features of the analysis to survive in a more exact treatment. Specifically, we have provided theoretical support for the idea that coherent islands may be locally stable with respect to both dislocated islands and uniform film growth, for certain materials systems. This will be most likely in systems with large lattice misfit, where the dislocation density necessary for incoherent islands is relatively costly.

Under these conditions, we imagine that the system evolves through three stages with increasing deposition of the mismatched species. The first stage is the nucleation of coherent islands whose size exceeds the critical size necessary to prevent these islands from "evaporating" back into the uniform film phase (i.e., $Q > Q_c = 2.49$ in the model). In the second stage, coherent islands grow by Ostwald ripening until they reach the size at which dislocated islands are preferred. In the third and final stage, dislocated islands grow by Ostwald ripening. In this scenario, the rounded islands observed by Eaglesham and Cerullo [2] and the prism-shaped islands seen in STM by Mo *et al.* [3] would be associated with the second and third stages, respectively. The difference in the observed shapes may be associated with differences in the relative importance of energetic and kinetic factors during these latter two stages.

In summary, we have analyzed the energetics of the relaxation of coherent islands in a model system. We find that even coherent islands of relatively low aspect ratio are surprisingly efficient at reducing their elastic strain energy. For appropriate materials parameters, we find that coherent islands can be favored over both dislocated island and the uniform film morphology. These results provide theoretical support for the hypothesis that Stranski-Krastanov growth proceeds via an intermediate coherent-island phase for the Si/Ge system.

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