Surface strains in epitaxial systems

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The stress state of heteroepitaxial film systems is examined using a boundary integral method together with boundary conditions that allow deflections at the substrate/film interface. It is found that for geometries that deviate from planar structures significant variations in surface strain and film energy arise. These calculations explain recent important experimental results for Ge growth on Si, including observations for Ge islands on Si that show the surface lattice constant can exceed the bulk Ge value, and observations that the preferred region for growth on terraced films is not necessarily at the steps. © *1995 American Institute of Physics.*

Strained heteroepitaxial structures are becoming increasingly important as a means of enhancing the electronic/optical properties of semiconductor devices. While significant potential improvements in performance are possible through the use of increasingly dissimilar materials, the resulting misfit strain makes it more difficult to grow the ideal structures envisioned by device designers. This difficulty arises because strained films are not stable structures.[1–3] In addition to strain relief by dislocation formation,[4] it has been shown both experimentally and theoretically that films can become rough to reduce their strain energy.[5–15] This result, only relatively recently applied to the study of thin films, has been previously examined in the context of the effect of stress on sintering, precipitate shapes, and other phenomena.[16–19] We explore the effects that large amplitude deviations from planar geometries have on the surface strain and film energy. We predict behaviors that have significant impact on the understanding of thin film growth and shed light on some remarkable experimental results for Ge growth on Si. In particular, observations for Ge islands on Si indicate that the surface lattice constant can exceed the bulk Ge value by more than 1.5% despite the 4% compressive strain at the film/substrate interface,[20] and that there is a preferred region for adatom collection on a Ge growth surface.[21] These two observations and reflect behavior predicted by our two-dimensional model. In addition, we find for nonplanar systems that relatively small deflections of the substrate/film interface can result in significant reductions in the elastic energy of the films. There is also considerable enhancement of the variability of the surface stress when the substrate is allowed to relax, as opposed to results when the substrate is constrained to remain flat.

A boundary integral analysis was used to evaluate the stress state of film/substrate systems.[22] In the boundary integral formulation, the partial differential equations for two-dimensional elasticity are replaced by an equivalent integral equation on the boundary. For each boundary point either surface displacement or traction is prescribed, and the unknown value is determined through numerical approximation of the integral equation. The present discussion is limited to a computationally simple two-dimensional plane strain model, and a differentiable isoparametric Overhauser approximation was employed.[23]

Recent calculations[10,24] and experiments[25] for heteroepitaxial systems have suggested that the effect of the substrate is significant. We examine the stress distribution in heteroepitaxial systems with large amplitude deviations from planar morphologies. We consider nonplanar continuous layer films and films consisting of discrete islands on a fully deformable substrate. Interface boundary conditions were developed to represent coherent epitaxial growth of a strained thin film. The lattice mismatch at the interface was modeled by setting the initial position of the film nodes to be shifted from the position of the corresponding substrate node, the amount of shift determined by the lattice mismatch. The film and substrate are then relaxed by requiring that the traction at the interface balances, and that corresponding nodes for substrate and film in the interface region, end up at the same location.[26] The remaining boundary conditions are standard. The bottom of the substrate is fixed, the sides of the substrate and film are periodic, and all other surfaces are free. The solution yields the strain of the coupled substrate and film system while realistically treating the interface boundary condition.

In order to compare our results with experiment, we use elastic constants derived from silicon and germanium with a lattice mismatch of 4%. The materials are assumed isotropic, with the effects of crystal symmetry approximated by using Voigt averaged elastic constants.[27] The values of the elastic constants are listed in Table I.

We consider first the role of the substrate on the stress state of nonplanar thin film morphologies. The importance of the substrate, and thus the need to use realistic interface boundary conditions, can be appreciated by considering the simple example of a film of Ge with a surface initially set to follow a cosine wave on a thick Si substrate (100 nm thick) (see Fig. 1). The system is relaxed for two different interface boundary conditions: (1) the interface is constrained to remain flat at $y=0$, and (2) the interface is allowed to relax as described above. For a film with its surface initially set to a cosine wave with amplitude 3.5 nm, 32 nm wavelength, and

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TABLE I. Elastic constants (dyn/cm^2) for silicon and germanium.

	C_{11}	C_{12}	C_{44}	G	ν
Ge	12.85×10^{11}	4.83×10^{11}	6.68×10^{11}	5.612×10^{11}	0.2006
Si	16.58×10^{11}	6.39×10^{11}	6.814×10^{11}	6.814×10^{11}	0.2174

an average thickness of 4 nm (Fig. 1), the system with the deformable interface (2) has 85% of the strain energy in the Ge film and 15% in the Si substrate. Fixing the interface in (1) increases the total strain energy by 17% above that for the more realistic interface conditions of (2). The strain energy stored just in the film is increased 37% when the interface is constrained to remain flat. Despite the fact that the strain energy is reduced in (2), the maximum stress is a factor of 6.6 greater than for the fixed flat interface model. This effect can clearly be seen in the plots of the strain energy density in Fig. 1. The interface deflections responsible for these very large changes in strain energies and peak stress are surprisingly small, -0.075 nm to $+0.067$ nm. Thus, for systems in which the substrate can accommodate some of the misfit strain, the strain energy stored in the film is reduced and the stress concentration in the valleys is enhanced. This is an important consideration for any realistic thin film calculation involving strain, especially when the stress concentration in the film is close to a deformable substrate, e.g., the nucleation of dislocations at edges of islands. The nearly one order of magnitude increase in stress seen in the above calculation can provide sufficient energy to overcome the barrier to dislocation nucleation. It also has important implications for the morphology of the growing film. The strain distribution at the film surface results in gradients in the chemical potential that can significantly influence surface

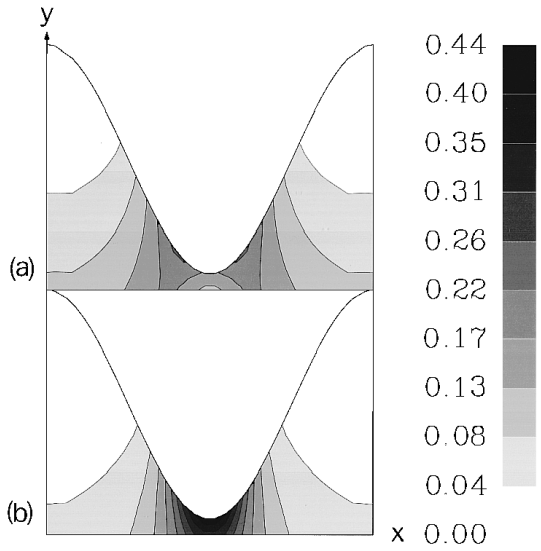

FIG. 1. Strain energy density in a film with the surface initially set to follow a cosine wave [$y=4+3.5\cos(2\pi x/32)$]. y is the normal distance from the initial position of the interface prior to relaxation, x is the distance parallel to the interface, and the units are nanometers. In (a) the interface is constrained to be flat while in (b) the substrate is allowed to deform. Only the film is shown. The strain energy in the Ge film in (a) is 9.43×10^{-4} erg/cm and 6.86×10^{-4} erg/cm in (b). Note that the x and y distances are scaled differently.

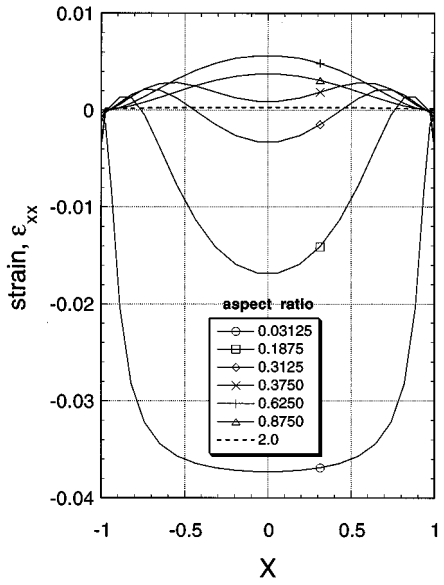

FIG. 2. For a rectangular island the diagonal component of the strain matrix, ϵ_{xx}, on the top surface of the island in the direction parallel to the initially flat interface (x direction) is plotted as a function of lateral position x for various aspect ratios α=height/width. $x=0$ is the center of the film.

diffusion[16] of the deposit and thus the growth pattern of the film.

One of the most striking aspects of the present calculations is illustrated in Fig. 2, where the lateral strain on the top surface of a rectangular Ge island on a Si substrate is plotted for a number of aspect ratios, α=height/width. For the smallest α in Fig. 2, the entire island surface is negatively strained with a small region of reduced strain at the edge. As α increases, a region of positive strain develops, moves away from the edge, and broadens. The maximum positive strain occurs at approximately $\alpha=0.5$. Further increase in α reduces the surface strain while still leaving it positive. At $\alpha=2$, nearly all the strain at the surface is relieved and the top surface of the Ge island is very close to the bulk lattice constant. It is important to note that these results for rectangular islands do not change qualitatively by either altering the island shape or by allowing interactions between islands. We find that for trapezoidal islands with sides and a top corresponding to {113} and {001} facets observed for Ge on Si(001), the strain distributions are quite similar to those found for the rectangular islands. The strain energy of the island and the maximum stress in the island show negligible increases of about 2% and 4%, respectively, as the distance between islands is reduced from 3 times to 1 times the island width.

The average strain on the top surface of the island is plotted in Fig. 3. For the compressively strained film, the surface strain, initially, is relatively uniform and nearly identical to the interfacial strain. As the island thickens, the surface strain decreases and can even change sign, becoming tensile for our model Ge island on Si. The behavior predicted here provides an excellent explanation of experiments of Theiss *et al.*,[20] who have studied the change in lattice constant on the top surface of Ge islands on Si(111) using a scanning tunneling microscope. They observed that the aver-

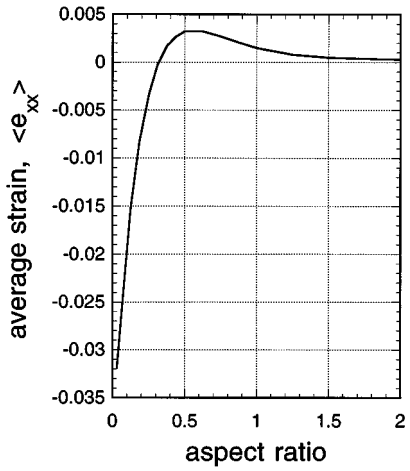

FIG. 3. The average lateral strain, $\langle \epsilon_{xx} \rangle$, on the top surface of rectangular islands is plotted as a function of island aspect ratio.

age lattice constant of the flat top surface of Ge islands on Si(111) gradually increases from a value near the Si value until it exceeded the bulk Ge value with increased island height. Upon further growth, the island's surface lattice constant dropped below the Ge bulk value once again before finally returning to its bulk value. The reduction in the surface lattice constant is correlated with a reduction in island aspect ratio, which, as our calculations indicate, leads to an increased compressive surface strain and, thus, a reduced lattice constant on the island surface. Dislocations are observed just after the lattice constant drops below the Ge bulk value, indicating that in this regime more than one strain relief mechanism is involved.

These results for the strain on the top surface of the Ge islands have additional implications concerning Ge growth on terraced films. The surface strain energy is an important component of the chemical potential along the surface. A nonuniform chemical potential produces surface diffusion fluxes proportional to the chemical potential gradient.[16] Adatoms will tend to move to regions of low strain (low chemical potential) enhancing collection and sustained growth in these regions. For our model rectangular islands with very small aspect ratios (less than 0.03), the strain is relatively constant along most of the island surface, with a sizable gradient only near the edges. As the aspect ratio increases, the surface strain becomes less uniform, with significant gradients from the center out to a region near but not at the edge. It can be expected that adatoms should collect in this region near the edge. For the elastic constants used in this study, at an aspect ratio of 0.38, the center of the island is less strained than the rest of the island surface; thus, growth is expected to be enhanced in the center of the island. As the island aspect ratio increases beyond this value, the surface strain becomes tensile everywhere, with the maximum strain at the center of the island. The surface strain gradient will tend to move adatoms from the center of the island out to and off the edge. For aspect ratios of 2 or more the surface strain and surface strain gradient becomes negligible. This predicted behavior is strikingly similar to recent observations by Chen *et al.*[21] for Ge islands on vicinal Si(001). They observe that new Ge layers form on the top surface near but not at the front of sawtooth growth fronts of S_B steps.[21] We analyze this behavior by taking two-dimensional sections normal to the growth direction for sawtooth growth fronts and applying the strain analysis for the 2-D rectangular islands. The intermediate aspect ratios correspond to regions near the front of the sawtooth. As discussed above, it is only for this intermediate regime, $0.1<\alpha<0.4$, that the diffusional forces (due to the strain gradients) tend to move the atoms into the low strain regions away from the islands edges. It is in these regions where one would expect the next Ge layer to begin to form, and this corresponds well with the atomic force microscopy observations of Chen *et al.*[21]

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