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EVOLVING SURFACE CUSPS DURING STRAINED-LAYER EPITAXY

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ABSTRACT

We have combined Z-contrast imaging and Ge marker layer experiments to study the evolving surface morphology of $\text{Si}_x\text{Ge}_{1-x}$ alloys grown by molecular beam epitaxy (MBE). Surface cusps are seen to arise as the intersection lines between coherent islands. The potential implications of stress concentrations associated with cusps are considered with a view to strain relaxation in the film via dislocation nucleation.

INTRODUCTION

The shape evolution of a thin overlayer film subjected to substrate-induced misfit strain remains a fundamental issue in strained-layer epitaxy. In the general sense, it is known that flat surfaces are unstable to the formation of waves [1] or coherent islands [2]. However, outstanding questions concerning the details of evolving surface shapes, the essential physics underlying surface instabilities, and the nucleation of strain relieving misfit dislocations still remain unanswered. Here, we report the surprising result that strained $\text{Si}_x\text{Ge}_{1-x}$ films are unstable to the formation of cusp-like features. The cusps are associated with high stress concentrations, and we consider the potential implications of this morphological development as a means of strain relaxation in the film via dislocation nucleation.

MARKER LAYER EXPERIMENTS

To investigate the evolving surface morphology of $\text{Si}_x\text{Ge}_{1-x}$ alloys, we performed a $\text{Si}_{0.5}\text{Ge}_{0.5}/\text{Si}(001)$ MBE growth experiment in which two monolayer Ge marker layers were deposited at selected intervals. These marker layers were then imaged along [110] in cross section using the Z-contrast technique [3,4] to provide a direct map of the evolving nonequilibrium surface morphology. In comparison with conventional "quench-and-look" techniques, this method has the distinct advantage that highly metastable features should be preserved under the conditions of high supersaturation.

A typical experimental result is shown in Fig. 1(a). Here, the Ge marker layers appear as bright horizontal lines. The morphological development can be broadly classified into three distinct regions. Initially, the alloy grows with a kinetically enforced flat surface morphology preceding the development of a wave-like morphology at around 25 nm. However, the surprising feature of the image is the appearance of cusp-like shapes at around 40 nm. The cusps, which we interpret as intersection points between coherent islands, persist for 20 nm or so before rapidly flattening out. This is highly suggestive that these features are linked to strain relaxation in the film.

THE ORIGIN OF CUSPS

To explain how the cusps arise, we refer to the basic concept of stress concentration as indicated in Fig. 2. If a thin film under compressive (or tensile) stress contains a surface undulation, then a stress concentration will develop at the center of the valley given by $\sigma(1 + 2(d/\rho_v)^{1/2})$ where σ is the applied stress, d is the depth of the undulation, and ρ_v is the radius of curvature at the valley. The lateral velocity of adatoms is governed by the gradient in surface chemical potential given by [1,5]

$$\mu = \mu^s + \gamma \Omega \kappa(x) + \sigma_{\tau}^2(x) \frac{\Omega}{2E} \quad (1)$$

where μ^s is the chemical potential of the unstressed surface, Ω is the atomic volume, γ is the surface free energy per unit area, and the x -axis is defined in Fig. 1(b).

Fig. 1. (a) [110] Z-contrast image of a Si_{0.5}Ge_{0.5} alloy grown by MBE with 0.25 nm Ge marker layers deposited at selected intervals. The growth temperature was 400°C and the deposition rate 2 Ås⁻¹. The profile simulations in (b) correspond to the period of cusp formation in (a) between vertical ordinates Y₁ and Y₂. The solid circles denote the positions of the peak, valley, and inflexion position as determined by the simulation method described in the text.

The second term, involving the surface curvature $\kappa(x)$, reflects the fact that the surface would like to be flat to minimize the surface energy. However, the third term involves the elastic energy cost of locating an atom at a surface site under tangential stress $\sigma_\tau(x)$, which has a maximum at the center of the valley. Therefore, if the elastic energy term dominates, adatoms will prefer to migrate away from regions of high chemical potential, such as at the valley, to the more relaxed regions of lower chemical potential at the peaks. Clearly, this situation can rapidly run away with itself because as adatoms migrate, the valley will further sharpen, which accentuates the stress concentration and in turn increases the rate of migration.

Fig. 2. Schematic representation of the origin of a surface stress concentration arising at the valley of a surface undulation in a film under compressive stress.

This qualitative picture of cusp formation can be investigated more quantitatively using the time evolution equation for the surface profile [1,5] with the addition of a deposition term. Using a simple model for the surface stresses and curvatures at the peak and valley, it has been possible to solve the evolution equation analytically and obtain instantaneous profile velocities at the peak, valley, and point of inflexion [6]. By approximating the initial modulation (Y_1) in Fig. 1(a) as a sinusoid, it is then possible to simulate the evolution of a cusp between Y_1 and Y_2 . The simulation in Fig. 1(b) clearly reproduces all of the experimental features in Fig. 1(a), indicating that cusp formation is a physically reasonable process.

An interesting prediction of the simulation is that at a critical stage of evolution, the cusp sharpness and tip stress will accelerate rapidly. This would seem to be intimately connected to the Griffith criterion for the unstable propagation of surface cracks. A Griffith crack propagates when the energy cost of creating a new surface is outweighed by the elastic energy released [7]. Cusp formation is merely the result of the same energy balance applied to mass transport over the surface of a strained film. In the limit of a sharp crack, our simple analysis gives the critical depth for rapid propagation as $E\gamma/2\sigma^2$ which is slightly less than the Griffith value of $2E\gamma/\pi\sigma^2$.

STRAIN RELAXATION

In reality, of course, the tip stress cannot increase indefinitely, and the most obvious candidate to relieve the stress is the nucleation of a dislocation. In Fig. 3, for example, we consider the nucleation of a 60° half-loop at the cusp tip. The total energy for the nucleation process is given as a function of loop radius R_L by

$$U(R_L) = R_L \frac{b^2 \mu}{8} \frac{(2-\nu)}{(1-\nu)} \log \left(\frac{\alpha R_L}{b} \right) - \frac{R_L \mu b^2}{4} \cos \beta - \left[5 \cos \frac{\phi}{2} - \cos \frac{5\phi}{2} \cos \lambda \cos \phi \right] \\ \times \frac{1}{4} \frac{(1+\nu)}{(1-\nu)} \epsilon \mu b \rho_v^{1/2} (1 + 2(d/\rho_v)^{1/2}) \int_{-R_L}^{R_L} [\rho_v + 4(R_L^2 - z^2)^{1/2}]^{1/2} - \rho_v^{1/2} dz. \quad (2)$$

The first term specifies the energy cost of a dislocation of core parameter α and Burgers vector b in a medium of shear modulus μ and Poisson's ratio ν . The second term is the energy gained by the removal of a surface step, β being the angle between $\mathbf{b}$ and the direction in the slip plane which is perpendicular to the intersection of the slip plane with the surface. The last term represents the elastic energy released by the loop where λ and ϕ are the angles defined by Matthews [8]. Here, we have utilized a blunt crack model [9] for the radial fall off in stress away from the tip.

Fig. 3. Geometry of dislocation half loop nucleation at a surface cusp (see text).

The nucleation energy given by Eq. (2) is plotted as a function of expanding loop radius in Fig. 4 (lower curve) and compared with the usual situation for nucleation at a flat surface (upper curve). In the case of nucleation at a flat surface, the total energy increases until a critical radius R_c is reached at which there is a large (~ 70 eV) activation barrier for the nucleation process. Under these circumstances, nucleation is therefore effectively impossible. In complete contrast, however, we find that nucleation at a cusp geometry corresponding to Fig. 1(a) is almost barrierless, indicating that there is enough elastic energy stored within the cusp stress field to nucleate and inject a loop into the film (and possibly into the substrate, explaining the recent observation of LeGoues et al. [10]).

It is clear from our discussion that the shape evolution of the surface is of critical importance when discussing dislocation nucleation. To quantify this concept, we note that R_c is appreciably reduced by the cusp stress field so that for $\rho_v \gg R_L \approx R_c$ in Eq. (2), we can consider dislocation nucleation at a cusp to be similar to dislocation nucleation at the flat surface of a film which is uniaxially stressed to the tip stress. As a reference point, taking ϵ^* as the system dependent flat film strain at which the nucleation becomes barrierless, then the critical cusp geometry or shape $G_c = d/\rho_v$ giving rise to barrierless nucleation is

$$G_c = \frac{1}{4} \left(\frac{\epsilon^*}{\epsilon} - 1 \right)^2. \quad (3)$$

The critical curve G_c is plotted in Fig. 5 as a function of misfit ϵ for the Si-Ge system ($\epsilon^* = 0.088$). If, during a given growth experiment, we form cusps with a subcritical geometry lying below the critical curve, then we would expect no dislocations. However, Eq. (3) would suggest that cusp geometries approaching the line might be expected to nucleate dislocations. In particular, the experimental cusps seen in Fig. 1(a) lie very close to the G_c curve for a 50% alloy consistent with the possibility that cusps act as nucleation sources. Additional images of cusps can also be seen in the literature which have previously been interpreted as sinusoids. For example, the triangle in Fig. 5 represents a subcritical cusp observed by Pidduck et al. [11], where in this case the film was fully coherent as expected.

It is interesting that the development of cusps can explain many of the puzzling aspects of dislocation nucleation at low misfits. For example, high stress concentrations can develop as a result of sharp cusps (small ρ_v) without the need for very large islands (large d). Furthermore, cusp formation is clearly very sensitive to growth conditions. It would, therefore, seem appropriate to redefine the so-called critical thickness as that film thickness at which a critical surface stress concentration develops capable of nucleating a dislocation. It is also apparent from Fig. 1(a) that cusps can act as multiple sources for perhaps 20 nm in the growth direction. Once the critical geometry is attained, dislocations will be generated in

Fig. 4. Total energy curves for nucleation of a 60° half loop in a $\text{Si}_{0.5}\text{Ge}_{0.5}$ alloy as a function of loop radius R_L . The top and bottom curves, respectively, correspond to nucleation at a flat surface and cusp of geometry ($d = 7$ nm, $\rho = 3$ nm) appropriate to Fig. 1(a). The calculation assumes $\alpha = 4$.

Fig. 5. Critical cusp shape G_c required for the barrierless nucleation of 60° half loops. Relevant experimental data points are superimposed (see text).

closely bunched (111) planes, the rate of nucleation limited by glide away from the tip. The cusps then rapidly flatten out to lower the surface energy in response to the strain relief in the film. Finally, it is important to emphasize that our calculations have been performed for the most pessimistic case involving barrierless nucleation of complete 60° dislocations with a core parameter of four. In reality, the thermally activated introduction of a sequence of partial dislocations could be considerably easier.

SUMMARY

We have imaged evolving surface cusps during the MBE growth of $\text{Si}_x\text{Ge}_{1-x}$ alloy films. The origin of cusps can be explained by surface diffusion in response to gradients in the surface chemical potential induced by surface stress. The nucleation of surface half-loops can then be explained by the stress concentration at the cusp tip [12].

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