

Phase boundary between ripple and hut in the initial roughening stage in heteroepitaxy

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A regularly undulating surface topography has been observed during growth of heteroepitaxial layers such as $\text{Si}_{1-x}\text{Ge}_x/\text{Si}^2$ and $\text{In}_x\text{Ga}_{1-x}\text{As}/\text{GaAs}^5$. We present a modified evolution mechanism of this ripple structure, which consists of initial roughening and evolving stages. A theoretical relationship is derived through energy minimization, which indicates that the ratio of the amplitude to the square of the period of the ripple structure is constant in the evolving stage. Also derived is a criterion for determining the phase boundary between the ripple and hut phases in the Stranski–Krastanov growth. © 1998 American Institute of Physics. [S0021-8979(98)00209-6]

In heteroepitaxy, Stranski–Krastanov (SK) growth can occur:¹ a few monolayers grow layer by layer and then induced by the strain in the epilayer, further growth proceeds by islanding. The SK growth may proceed in two different modes: ripple and hut growth modes. One interesting feature found in the surface morphology of $\text{Si}_{1-x}\text{Ge}_x/\text{Si}$ is that the film structure evolves from a rippled surface in the beginning stage to a three-dimensional island morphology.^{2–4} This regularly undulating surface topography has also been observed in $\text{In}_x\text{Ga}_{1-x}\text{As}/\text{GaAs}$ systems.⁵ Most theoretical works^{6,7} have been devoted to the slightly wavy surface where the strain energy is always reduced when an initially flat surface is slightly perturbed in an arbitrary manner. The reduced strain energy competes with the surface energy which stabilizes the surface at short wavelengths. As a result, the instability can simply be regarded as a competition between the elastic energy and the surface energy. These perturbation results, however, cannot give information on the film morphology after the film evolves. For this evolving stage, several authors^{8,9} have simulated numerically the stress-driven surface instability based on chemical potential analysis. Due to large misfit in their system, a nominally flat surface profile of an elastically stressed solid can rapidly evolve into a cusped surface with a fixed period by surface diffusion. Small misfit results in a rather smooth sinusoidal curve because {501} facet hinders the cusp formation.¹⁰ Recent experimental data³ have revealed that the ratio of undulation amplitude to the square of wavelength is constant for a given Ge fraction as the ripple structure evolves.

On the other hand, a microscopic study of Ge on $\text{Si}(001)$ ¹¹ has disclosed the existence of a metastable three-dimensional phase consisting of small (hut) clusters that have a {501} facet structure. Mo *et al.*¹¹ speculate that the hut clusters are an intermediate phase in the formation of macroscopic clusters (islands).

These experimental observations reveal that there are

two intermediate steps leading to the macroscopic islands. One is surface rippling and the other is hut cluster.

In this communication, we propose an evolving mechanism of the ripple structure and derive an expression for the phase boundary between the ripple and hut structures in Si/Ge heteroepitaxy. We restrict our result here to the system where kinetic barriers can be neglected due to high temperature and where misfit is small.

The ripple structure of our interest is schematically shown in Fig. 1. Gao⁶ showed that in the initial roughening stage, the strain along the two-dimensional sinusoidal surface is given by

$$\frac{\epsilon}{\epsilon_0} = 1 - 2\sqrt{2}\pi(1+\nu)\frac{t_1}{\lambda}\cos kx\cos ky, \quad (1)$$

where ϵ_0 is the bulk strain induced by the lattice mismatch between film and substrate and k is $2\pi/\lambda$. Then the excess volume strain energy ΔF_v over the area λ^2 is given by

$$\begin{aligned} \Delta F_v &= 2\mu\left(\frac{1+\nu}{1-\nu}\right)\int_0^\lambda\int_0^\lambda\epsilon(x,y)^2t(x,y)dxdy, \\ &= -2\sqrt{2}\pi\frac{(1+\nu)^2}{(1-\nu)}\mu\epsilon_0^2\lambda t_1^2, \end{aligned} \quad (2)$$

where μ is the shear modulus for the epilayer material and $t(x,y) = t_1\cos kx\cos ky$, where t_1 being the half amplitude shown in Fig. 1. It is now necessary to balance this energy gain against the change in surface free energy, ΔG_s , due to the increase in surface area resulting from the undulations. The increase in the surface area ΔS is

$$\Delta S = \int_0^\lambda\int_0^\lambda\sqrt{1+\left(\frac{\partial t}{\partial x}\right)^2+\left(\frac{\partial t}{\partial y}\right)^2}dxdy - \lambda^2 \approx \pi^2 t_1^2, \quad (3)$$

where the approximation is based on the fact that $t_1^2 \ll (\lambda/2\pi)^2$ in the perturbation stage. Then ΔG_s is

$$\Delta G_s = \pi^2 t_1^2 \gamma, \quad (4)$$

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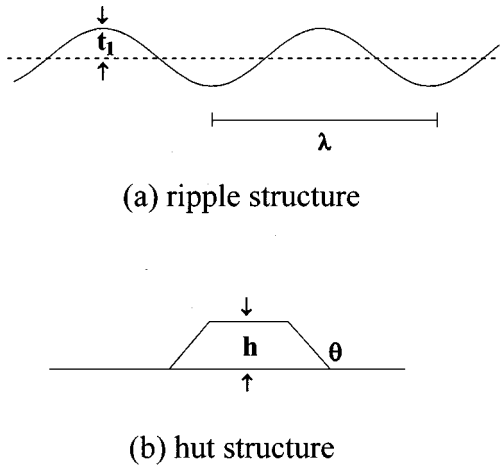


FIG. 1. Schematic diagram of ripple and hut structures.

where γ is the surface free energy per unit area. The net energy change is then

$$\Delta E = -2\sqrt{2}\pi \frac{(1+\nu)^2}{(1-\nu)} \mu \epsilon_0^2 \lambda t_1^2 + \pi^2 t_1^2 \gamma. \quad (5)$$

Equation (5) shows that the critical period λ_c scales as $\gamma/\mu\epsilon_0^2$, which corresponds to the earlier works of D. Srolovitz¹² and P. Voorhees.¹³ Differentiating Eq. (5) with respect to t_1 gives $\lambda = \lambda_c(\text{const})$. This fact reveals that in the initial roughening stage, the wavelength λ is fixed at λ_c and t_1 increases, which coincides with the simulation result.

We have so far concentrated on the initial roughening stage. These results, however, cannot give information on the film morphology after the film evolves. For the information, we deal here with the evolving stage to which the perturbation method is no longer applicable. Equation (1) shows that if λ is fixed and t_1 increases, there is a point where the strain at the ripple peak is completely relaxed. Cullis *et al.*¹⁴ showed that near-complete relaxation takes hold in the ripple peaks in the direction normal to the ripple rows. In the near-complete relaxation state, ϵ is simply given by

$$\epsilon = \epsilon_0(1 - \cos kx \cos ky) \quad (6)$$

and Eq. (2) reduces to

$$\Delta F_\nu = -\frac{(1+\nu)}{(1-\nu)} \mu \epsilon_0^2 \lambda^2 t_1. \quad (7)$$

Equation (7) is justified in that the strain energy scales as $\lambda^2 t_1$ because the volume of ripple also scales as $\lambda^2 t_1$. Equation (7) is valid for low misfit system or smooth sinusoidal surface morphology. In large misfit system, the strain is considerably large at the trough but the trough exists as a point singularity such that the integration is not affected substantially. Chiu and Gao⁶ obtained a similar result for the strain energy of fully developed cusp using a complex mapping method. The net energy change is then

$$\Delta E = -\frac{(1+\nu)}{(1-\nu)} \mu \epsilon_0^2 \lambda^2 t_1 + \pi^2 t_1^2 \gamma. \quad (8)$$

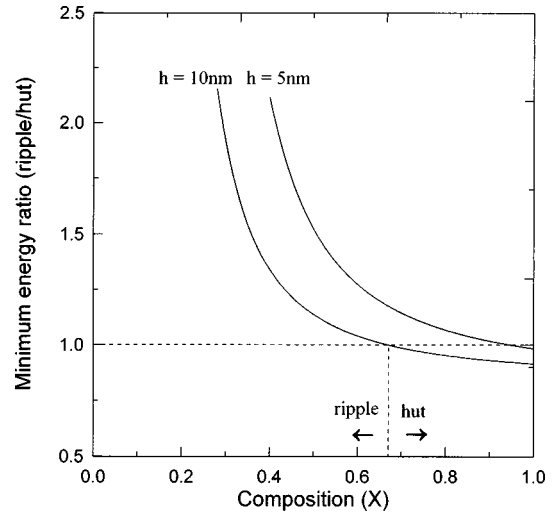


FIG. 2. The ratio of minimum energies with h as the parameter. Note that at a certain composition, $X = X_c$, there is a transition from the ripple to the hut structure.

Minimizing Eq. (8) with respect to t_1 gives the following minimum energy expression:

$$\Delta E_{\min} = -\frac{(1+\nu)^2 \mu^2 \epsilon_0^4 \lambda^4}{(1-\nu)^2 4 \pi^2 \gamma}. \quad (9)$$

The value of t_1 that minimizes ΔE can be rearranged to give

$$\frac{t_1}{\lambda^2} = \frac{(1+\nu) \mu \epsilon_0^2}{(1-\nu) 2 \pi^2 \gamma}. \quad (10)$$

Equation (10) reveals that the ratio of amplitude to the square of wavelength is constant in the evolving stage, which is supported by the experimental data by Pidduck *et al.*³ The energy per unit volume is given by

$$\Delta E_{\min}/V = -0.94 \left(\frac{1+\nu}{1-\nu} \right) \mu \epsilon_0^2, \quad (11)$$

where V is the ripple volume, which is $0.467\lambda^2 t_1$.

Tersoff and Tromp¹⁵ derived the energy change in the hut cluster formation. When their results are used for the minimum, there results

$$\Delta E_{\min}/V = -\frac{8\sqrt{e}}{\pi} \frac{(1+\nu)^2}{1-\nu} \tan \theta \mu \epsilon_0^2 \times \exp \left[\frac{-\pi\Gamma(1-\nu)}{2\mu\epsilon_0^2(1+\nu)^2 h} \right], \quad (12)$$

where h is the hut height, $\Gamma = \gamma_e \csc \theta - \gamma_s \cot \theta$ (γ_e and γ_s , respectively, are the surface energies of the substrate and the edge facets), ν and μ , respectively, are the Poisson ratio and the shear modulus of the substrate, and θ is the facet angle of the hut island, which is 11.2° for {501} facet. Dividing Eq. (11) by Eq. (12) yields

$$\frac{(\Delta E_{\min}/V)_r}{(\Delta E_{\min}/V)_h} = \frac{0.22}{\tan \theta (1+\nu)} \exp \left[\frac{\pi\Gamma(1-\nu)}{2\mu\epsilon_0^2(1+\nu)^2 h} \right], \quad (13)$$

where the subscripts r and h are for the ripple and hut structures.

Equation (13) is a criterion that can be used to determine which structure is favored in the film growth. When the ratio is less than unity, the hut structure is favored whereas the ripple structure is when it is larger than unity. Figure 2 shows the ratio plotted as a function of the composition X for $\text{Si}_{1-x}\text{Ge}_x$ system with h as the parameter. It can be seen that there is a transition from the ripple to the hut structure at a certain composition, $X=X_c$, for a given h . Here X_c is the value of X at which the curve crosses unity value of the energy ratio. Typical parameter values for $\text{Si}_{1-x}\text{Ge}_x$ are used in the plot. These are: $\mu=0.7\times 10^{11}$ N/m²,¹⁶ $\nu=0.25$,¹⁶ $\theta=11.2^\circ$, and $\Gamma=0.12$ N/m (assuming $\gamma_e=\gamma_s=1.2$ N/m¹⁶). It is shown in Fig. 2 that a larger X (nearly $X=1$) results in the hut structure as reported in Mo *et al.*¹¹

In summary, a modified evolution mechanism of the ripple structure has been presented, which consists of distinctly different initial roughening and evolving stages. In the evolving stage, the ratio of the amplitude to the square of the period of the ripple structure is constant. In the SK growth, the formation of macroscopic islands is preceded by either surface rippling or hut cluster formation. A criterion has been derived that can provide the conditions under which one of the two intermediate steps is favored.

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