CHIN. PHYS. LETT. Vol. 36, No. 1 (2019) 016801

Instability of Epitaxially Strained Thin Films Based on Nonlocal Elasticity *

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(Received 7 October 2018)

We perform a linear analysis of the elastic fields and stability of epitaxially strained thin films based on nonlocal elasticity. We derive expressions of perturbed stresses to the first order of perturbation amplitude, which show that the stresses are directly proportional to the lattice mismatch and the perturbation amplitude, and decrease with an increase in the perturbation wavelength. The critical perturbation wavelength distinguishes whether the flat film for the perturbation is stable, which is inversely proportional to the square of the mismatch and decreases with the thickness of the film.

PACS: 68.35.Gy, 62.20.D−, 68.35.Fx, 61.66.Dk DOI: 10.1088/0256-307X/36/1/016801

The growth of epitaxially strained thin films on lattice mismatch substrates is of major importance in the fabrication of semiconductor and optoelectronic devices. The lattice mismatch generates strain in deposited films, which can cause film instability, which is unfavorable to uniform flat film growth. The strained films can relax either by the introduction of dislocations or by the formation of coherent (dislocation-free) ripples or islands on the film surface via surface diffusion. These coherent ripples or islands can self-organize to create periodic arrays, which can be utilized to create quantum wire or quantum dot structures of electronic significance. Consequently, understanding and predicting strained thin-film evolution is important for the improved fabrication of semiconductor devices.

Early film growth tends to occur via coherent ripples or islands formation because there is an energy barrier to the introduction of dislocations. Dislocations occur at island edges once islands reach a certain size because the large stress at island edges provides a pathway for dislocations formation.[1] It has been observed experimentally that dislocation-free flat films of less than a certain thickness are stable to surface perturbations, while thicker films are unstable.[2−10] We only consider dislocation-free films because many experiments show an absence of dislocations especially in early film evolution.[1−10]

The instability of an initial planar system was first investigated by Asaro and Tiller,[11] and later in great detail by Grinfeld[12] and other researchers.[13−16] These works are based on classical (local) elasticity, which are scale free and cannot reflect the nanoscale physical laws of thin films; for example, the critical wavelength that is derived is independent of film thickness. In this Letter, we report on a linear analysis of the elastic fields and stability of epitaxially strained thin films deposited on mismatched substrates, based on Eringen's nonlocal elasticity theory,[17,18] and we derive expressions of the perturbed stresses to first order of perturbation amplitude, which show that the stresses are directly proportional to the lattice mismatch and the perturbation amplitude, and decrease with increasing the perturbation wavelength. The critical perturbation wavelength distinguishes whether the flat film for the perturbation is stable, which is inversely proportional to the square of the mismatch and decreases with the thickness of the film.

We consider a perturbation of a thin film on a substrate, as shown in Fig. 1. The surface of the film is at $y = h(x,t)$, the film is in the $y > 0$ region, and the substrate is in the $y < 0$ region with the film-substrate interface at $y = 0$. The lattice mismatch $\varepsilon_{\rm m}$ in the x and z directions between the film and the substrate creates a strain ε in the film. The system is modeled to be invariant in the z direction, and all quantities are calculated for a section of unit width in that direction. This is consistent with the plane strain where the solid extends infinitely in the z direction and hence all strains in this direction vanish; i.e., $\varepsilon_{xz} = \varepsilon_{yz} = 0$. We assume that there is no material mixing between the film and the substrate.

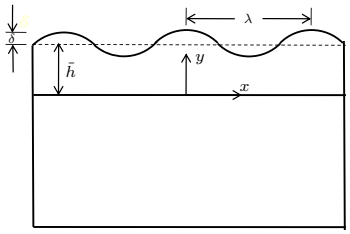

Fig. 1. Diagram of a perturbation of a thin film on a substrate.

Under nonlocal elasticity, which states that the stress at a point in a body depends not only on the local stress at that particular point, but also on the spatial integrals, with weighted averages, of the local stress contribution from all other points in the body, the relationship between stress and strain is given by[17,18]

$$\sigma_{ij} = \iint_V \alpha(x - x', y - y') C_{\rm ijkl} \varepsilon_{kl} dx' dy', \qquad (1)$$

*Supported by the Zhejiang Provincial Natural Science Foundation of China under Grant Nos LY18A020011 and LQ17A020001, and the National Natural Science Foundation of China under Grant No 11702247.

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where $i, j, k, l = x, y$, and

$$\alpha(x-x', y-y') = \frac{k^2}{a^2\pi/2} \exp\Big\{ -\frac{k^2}{a^2}[(x'-x)^2 + (y'-y)^2]\Big\} \quad (2)$$

is a nonlocal kernel function, with a being the crystal constant of the film, and $k = 1.65$. The equation of mechanical equilibrium in the absence of body forces is given by

$$\partial_j \sigma_{ij} = 0, \quad \text{for} \quad y < h(x,t). \quad (3)$$

Initial nonlocal equilibrium stresses are present in the film due to the mismatch

$$\sigma^0_{xx} = \sigma^0_{zz} = \sigma_{\rm m} S_{\rm i}(y, \bar{h}) = E/(1-\nu)\varepsilon_{\rm m} S_{\rm i}(y, \bar{h}), \quad (4)$$

where $S_{\rm i}(y, \bar{h}) = \frac{1}{\sqrt{\pi}/2} \int_{k(y-\bar{h})/a}^{ky/a} \exp(-z^2) dz$, E is the Young modulus, ν is the Poisson ratio, and $\sigma^0_{xy} = \sigma^0_{yy} = 0$ in the reference state, which corresponds locally to a flat film of thickness $\bar{h}$ constrained to have the lateral lattice constant of the substrate.

The total stress σ_{ij} in the perturbed state then is the sum of the initial stress and the stress σ^h_{ij} caused by the surface perturbation; i.e.,

$$\sigma_{ij} = \sigma^0_{ij} + \sigma^h_{ij}, \quad i, j = x, y. \quad (5)$$

Because the strains ε_{ij} are not independent but are linked via the displacements of the elastic body, they must also satisfy the equation of compatibility,

$$(\partial^2_{xx} + \partial^2_{yy})(\sigma^h_{xx} + \sigma^h_{yy}) = 0. \quad (6)$$

The boundary conditions are given by

$$\sigma_{ij} n_j = 0, \quad \text{at} \quad y = h(x,t), \quad (7)$$

$$\sigma_{ij} \to 0, \quad \text{when} \quad y \to -\infty, \quad (8)$$

where n is the exterior normal. Equations (1)–(8) give us a system of equations, which are sufficient for the complete determination of the equilibrium stress and strain in the system.

We assume that surface diffusion is the dominant mass transport mechanism. Gradients in the chemical potential produce a drift of surface atoms with an average velocity v given by the Nernst–Einstein relation

$$v = -\frac{D_{\rm s}}{k_{\rm B}T} \frac{\partial \chi}{\partial s}, \quad (9)$$

where $D_{\rm s}$ is the surface diffusion coefficient, s is the arc length, T is the temperature, $k_{\rm B}$ is the Boltzmann constant, and χ, given by an equation

$$\chi = (U - kU_{\rm s})\Omega, \quad (10)$$

is the chemical potential at the surface; i.e., it is the increase in free energy when an atom is added to the solid surface at the point of interest. Here U is the strain energy density (evaluated at the surface), $U_{\rm s}$ the surface energy density, k the mean curvature, and Ω the atomic volume. Taking the divergence of the surface current produced by the atom drift gives an expression for the surface movement,[19]

$$\frac{\partial h}{\partial t} = \frac{D_{\rm s}\eta\Omega}{k_{\rm B}T} \frac{\partial}{\partial x} \frac{\partial \chi}{\partial s}, \quad (11)$$

where η is the number of atoms per unit area at the solid surface.

In the following we carry out a linear stability analysis of Eq. (11) against a sinusoidal perturbation of wavelength λ and amplitude δ, similar to that carried out in Ref. [3] for an infinitely thick stressed film. We thus look for a height profile of the form $h(x,t) = \bar{h} + \delta(t)\cos\frac{2\pi x}{\lambda}$ with average thickness $\bar{h}$, which solves Eq. (11) to first order in δ.

To calculate the stress and strain, we use stress function of the form

$$A(x,y) = f(y)\cos\frac{2\pi x}{\lambda}, \quad (12)$$

where $f(y)$ is a function to be determined, taking into account the perturbed profile and the boundary conditions of the system. The stresses of perturbed state are then given by

$$\sigma^h_{xx} = \partial^2_{yy} A, \ \sigma^h_{xy} = -\partial^2_{xy} A, \quad \sigma^h_{yy} = \partial^2_{xx} A. \quad (13)$$

Substituting Eq. (12) into Eq. (6) gives

$$f^{(4)}(y) - 2(2\pi/\lambda)^2 f''(y) + (2\pi/\lambda)^4 f(y) = 0. \quad (14)$$

From the boundary conditions (7) and (8), the solution to Eq. (14) is given by

$$f(y) = -\frac{\delta\sigma_{\rm m} S_{\rm i}}{\sqrt{\pi}/2}(y - \bar{h})\exp(2\pi y - \bar{h})/\lambda, \quad (15)$$

to first order in δ.

The perturbed stresses are then as follows:

$$\sigma^h_{xx} = -\frac{4\pi\delta\sigma_{\rm m} S_{\rm i}}{\lambda\sqrt{\pi}/2}\Big[1 + \frac{\pi}{\lambda}(y-\bar{h})\Big] \cdot \exp\frac{2\pi(y-\bar{h})}{\lambda}\cos\frac{2\pi x}{\lambda}, \quad (16)$$

$$\sigma^h_{xy} = -\frac{2\pi\delta\sigma_{\rm m} S_{\rm i}}{\lambda\sqrt{\pi}/2}\Big[1 + \frac{2\pi}{\lambda}(y-\bar{h})\Big] \cdot \exp\frac{2\pi(y-\bar{h})}{\lambda}\sin\frac{2\pi x}{\lambda}, \quad (17)$$

$$\sigma^h_{yy} = \frac{4\pi^2}{\lambda^2}\frac{\delta\sigma_{\rm m} S_{\rm i}}{\sqrt{\pi}/2}(y-\bar{h})\exp\frac{2\pi(y-\bar{h})}{\lambda}\cos\frac{2\pi x}{\lambda}, \quad (18)$$

where $S_{\rm i} = \int_0^{k\bar{h}/a} \exp(-y^2) dy$.

Equations (16)–(18) show that the perturbed stresses are in proportion to the mismatch $\varepsilon_{\rm m}$ and the perturbation amplitude δ.

The total stresses at the surface in the perturbed state are evaluated to first order in δ, from Eqs. (5) and (16)–(18), and are given by

$$\sigma_{xx} = \frac{\sigma_{\rm m} S_{\rm i}}{\sqrt{\pi}/2}\Big(1 - \frac{4\pi}{\lambda}\Big)\delta\cos\frac{2\pi x}{\lambda}, \tag{19}$$

$$\sigma_{xy} = -\frac{2\pi}{\lambda}\frac{\sigma_{\rm m} S_{\rm i}}{\sqrt{\pi}/2}\delta\sin\frac{2\pi x}{\lambda}, \tag{20}$$

$$\sigma_{yy} = 0. \tag{21}$$

The strain energy density at the surface is given by

$$U = U_{\rm m}\Big[1 - \frac{4\pi}{\lambda}(1+\nu)\delta\cos\frac{2\pi x}{\lambda}\Big], \tag{22}$$

where $U_{\rm m} = \frac{(1-\nu)\sigma_{\rm m}^2}{E}\frac{S_{\rm i}}{\sqrt{\pi}/2} = \frac{E\varepsilon_{\rm m}^2}{1-\nu}\frac{S_{\rm i}}{\sqrt{\pi}/2}$ is the nonlocal strain energy density of the film in the reference state, and depends on the film thickness $\bar{h}$ through $S_{\rm i}$.

The curvature of the surface, which neglects terms of second order and higher in δ, is given by $k = h_{xx} = -\frac{4\pi^2\delta}{\lambda^2}\cos\frac{2\pi x}{\lambda}$. The chemical potential at the surface is given by, from Eq. (10),

$$\chi = \Big[U_{\rm m} + \Big(\frac{4\pi^2}{\lambda^2}U_{\rm s} - \frac{4\pi(1+\nu)}{\lambda}U_{\rm m}\Big)\delta\cos\frac{2\pi x}{\lambda}\Big]\Omega. \tag{23}$$

From Eqs. (11) and (23), one obtains an equation that is satisfied by the amplitude of perturbation,

$$\delta'(t) = -\frac{4\pi^2 D_{\rm s}\eta\Omega^2}{\lambda^2 k_{\rm B}T}\Big[\frac{4\pi^2}{\lambda^2}U_{\rm s} - \frac{4\pi(1+\nu)}{\lambda}U_{\rm m}\Big]\delta. \tag{24}$$

The solution to Eq. (24) gives

$$\delta(t) = \delta_0\exp\Big\{-\frac{4\pi^2 D_{\rm s}\eta\Omega^2}{\lambda^2 k_{\rm B}T}\Big[\frac{4\pi^2}{\lambda^2}U_{\rm s} - \frac{4\pi(1+\nu)}{\lambda}U_{\rm m}\Big]\Big\}t, \tag{25}$$

where δ_0 is the initial amplitude of perturbation. Equations (24) and (25) show that the perturbation amplitude δ increases with t if $\frac{4\pi^2}{\lambda^2}U_{\rm s} - \frac{4\pi(1+\nu)}{\lambda}U_{\rm m} < 0$, and decreases with t if $\frac{4\pi^2}{\lambda^2}U_{\rm s} - \frac{4\pi(1+\nu)}{\lambda}U_{\rm m} > 0$. The former means that the flat film is unstable for the perturbation at perturbation wavelength $\lambda > \frac{\pi U_{\rm s}}{(1+\nu)U_{\rm m}}$, and the later means that the flat film is stable at $\lambda < \frac{\pi U_{\rm s}}{(1+\nu)U_{\rm m}}$. The critical perturbation wavelength that distinguishes the stability of the flat film is therefore

$$\lambda_{\rm cr} = \frac{\pi U_{\rm s}}{(1+\nu)U_{\rm m}}, \tag{26}$$

which is inversely proportional to the square of the mismatch $\varepsilon_{\rm m}^2$, or $\sigma_{\rm m}^2$.

For a description of the $Si_{1-x}Ge_x$ layers deposition on the silicon surface, it is essential to take the dependence of the thermodynamic parameters on the composition x into account in the formulae. For this purpose, the values of the physical constants ξ, such as the lattice mismatch, the elastic constants and the surface energy density for pure materials (Si, Ge) and Vegard's law are used[20]

$$\xi(x) = \xi({\rm Ge})x + \xi({\rm Si})(1-x). \tag{27}$$

The existing works experimentally report on the morphological evolution of SiGe film grown on Si, reveal that the surface morphology has sinusoidal features,[21–24] and estimate that the critical wavelength $\lambda_{\rm cr} \approx 230\,{\rm nm}$ for $x = 0.21$ and $\bar{h} = 40\,{\rm nm}$, and $\lambda_{\rm cr} \approx 315$ nm for $x = 0.19$ and $\bar{h} = 50\,{\rm nm}$, which are in good agreement with the calculated results $\lambda_{\rm cr} \approx 233\,{\rm nm}$ and $\lambda_{\rm cr} \approx 296\,{\rm nm}$, respectively, based on Eqs. (26) and (27).

Figure 2 shows the critical wavelengths $\lambda_{\rm cr}$ versus average thickness $\bar{h}$ and composition x, for the alloying film $Si_{1-x}Ge_x$ on the Si substrate (001). For comparison, the results of local elasticity, where the wavelengths $\lambda_{\rm cr}$ are independent of the film thickness $\bar{h}$, and correspond to the asymptotes of the curves, are also given in Fig. 2.

Equation (26) or Fig. 2 shows that the critical wavelength decreases dramatically with the thickness of the thin film in the earlier stages of growth, and changes slightly when the thickness of the film is over a certain thickness, which depends on composition x. The larger the composition, the smaller the thickness of instability.

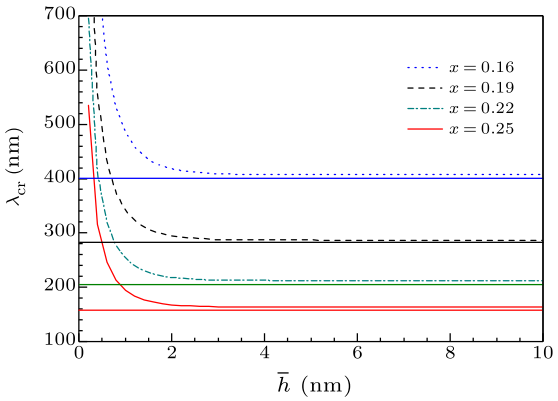

Fig. 2. Critical perturbation wavelength $\lambda_{\rm cr}$ versus thickness $\bar{h}$. The curves and straight lines correspond to the results of nonlocal and local elasticity, respectively.

In summary, we have carried out a linear analysis of the nonlocal elastic fields and stability of epitaxially strained thin films. According to the expressions (19)–(21) of the stresses at the surface in the perturbed state, we see that the valleys of the perturbed surface are the regions of high stress, correspondingly, have large chemical potential. We also see that the hills are the regions of low stress and correspondingly have small chemical potential, hence atoms tend to detach from valleys and attach to hills increasing perturbation amplitude. However, surface energy acts to reduce the surface curvature, thereby reducing perturbation amplitude. Thus equilibrium between the surface stress and surface tension is the critical condition of stability shown by Eq. (26). From Eqs. (16)–(21) and (26), the stresses and critical perturbation wave-

length depend not only on the mismatch $\varepsilon_{\rm m}$ between the deposited film and substrate, and the perturbation amplitude δ, but also on the average film thickness $\bar{h}$, which is not done by classical (local) elasticity. These results reflect the nanoscale physical laws of thin films in heteroepitaxial growth. Instabilities are observed in heteroepitaxial growth and can be explained by the mechanism mentioned in the present study.

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