Growth Instability of Strained Film: An Elastic Green's Function Force Monopole Approach

Hao Hu and Feng Liu

Abstract We analyze the growth stability of an epitaxial strained film on a flat substrate by the elastic Green's function force monopole approach. We calculate and compare the strain energies and growth instability of three different forms of surface undulations: sinusoidal waviness, surface faceting and island formation. In general, the instability occurs beyond a critical length scale, in agreement with the conventional analysis of ATG instability based on the stress function approach. For isotropic surface energies, the critical length scale for the island formation is the smallest, because it offers the most effective mode of strain relaxation.

Keywords Strain • Thin film growth and stability • Elastic Green's function

1 Introduction

The morphology of epitaxially grown strained films has drawn much attention for its scientific and technological importance. Development of stress in the surface of a strained thin film can greatly change the surface morphology. Self-assembly and self-organization of step-flow growth [1, 2], quantum dots [3–5], quantum wires [6–8], and surface phase separation [9, 10] are all different manifestations of stress (strain) induced surface growth instabilities. Surface stress can also lead to the formation of a surface periodic domain structure [11, 12]. These phenomena open a new way to synthesis of nanostructures by strain induced self-assembly.

The stability of strained film grown on a flat substrate has been studied extensively, known as ATG instability [13–15], where a strained film becomes unstable to surface undulation beyond a critical wavelength λ_c. The strain undulation may

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take different forms: surface reconstruction [16], stress domain formation [11, 12], step bunching [1, 2], and 3D faceted island formation [3–5], etc. In general, the critical wavelength λ_c scales with the ratio of surface energy (γ) over strain energy ($Y\varepsilon^2$, where Y is Young's modulus, ε is misfit strain).

Conventionally the ATG instability is usually analyzed using the stress function approach [12–14]. Here, we re-analyze the strained film instability using an elastic Green's function force monopole approach. We calculate the strain relaxation energy in the undulated film relative to flat film in reciprocal space by Fourier transformation [11], and derive the critical length scale (wavelength and size) for instability. We consider three forms of surface undulation: sinusoidal waviness, periodic facets, and array of isolated islands, and among them the island formation is shown to have the smallest critical length scale for isotropic surface energies because of its most effective mode of strain relaxation. The paper is organized as the following: we introduce and detail the methodology in Sect. 2, calculate the total energies of strained films with the three different surface profiles and analyze their properties in Sect. 3. We conclude in Sect. 4.

2 Elastic Green's Function Force Monopole Method in Reciprocal Space

For a heteroepitaxially grown strained film, the surface stress of the film induced by the misfit strain can be calculated by the strain induced bulk stress times the local film thickness, i.e. $\sigma^f = C\varepsilon h(x,y)$, within the shallow-angle approximation [3, 17], where C is elastic modulus, ε is the misfit strain induced by lattice mismatch; these two quantities are assumed uniform within the film; $h(x,y)$ is the surface height profile function. Then, if the surface profile $h(x,y)$ is not flat, as shown in Fig. 1, the surface stress is non-uniform.

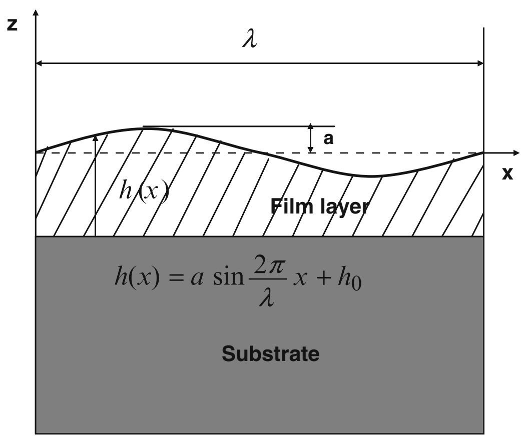

Fig. 1 Schematic illustration of sinusoidal strained film on a flat substrate

A surface with non-uniform stress will generate a surface elastic force monopole density, defined as [17, 18]

$$f_i\left(\vec{r}\right) = \frac{\partial}{\partial j}\sigma_{ij}\left(\vec{r}\right), \tag{1}$$

where $\sigma_{ij}(\boldsymbol{r})$ is the surface stress tensor, the indices i and j label directions in the plane of the surface, $\boldsymbol{r} = (x, y)$ is the position vector in the surface. Since we adopt the shallow-angle approximation, these elastic force monopoles can be projected onto the film surface plane, in parallel to the substrate surface plane. A homogeneous flat strained film on a flat substrate free of defects is free of surface elastic forces, because the surface stress is uniform, experiencing no strain relaxation.

The elastic force monopoles, induced by a non-uniform surface stress, in turn induce a displacement field $\boldsymbol{u}(\boldsymbol{r}, z)$ in the medium, which can be expressed in terms of elastic Green's function $\chi_{ik}(\boldsymbol{r}, z)$ [17, 18]:

$$u_i\left(\vec{r}\right) = \int d^2r' \sum_j \chi_{ij}(\vec{r} - \vec{r'}, z) f_j\left(\vec{r'}\right), \tag{2}$$

$\chi_{ik}(\boldsymbol{r}, z)$ depends on the film and substrate elastic properties, Young's modulus Y and Poisson ratio ν. Since the surface elastic forces defined in Eq. (1) only exist on the surface plane, the surface strain relaxation energy per unit area E_{el} is the integral of the force distribution multiplies the displacement over the surface [17]:

$$\begin{aligned} E_{el} &= -\frac{1}{2L^2}\int d^2r \sum_i f_i\left(\vec{r}\right) u_i\left(\vec{r}, 0\right) \\ &= -\frac{1}{2L^2}\iint d^2r d^2r' \sum_i \sum_j f_i\left(\vec{r}\right) \chi_{ij}\left(\vec{r} - \vec{r'}, 0\right) f_j\left(\vec{r'}\right), \end{aligned} \tag{3}$$

where L is the system size. When we treat surface morphology of periodic undulations with period (λ_x, λ_y), $f_i(\boldsymbol{r})$ and $\boldsymbol{u}(\boldsymbol{r}, 0)$ would all be periodic functions of (λ_x, λ_y), and their Fourier Transforms are:

$$u_i\left(\vec{r}\right) = \sum_{\vec{G}} u_i\left(\vec{G}\right) e^{i\vec{G}\bullet\vec{r}}, \tag{4a}$$

$$f_i\left(\vec{r}\right) = \sum_{\vec{G}} f_i\left(\vec{G}\right) e^{i\vec{G}'\bullet\vec{r}} = \sum_{\vec{G}} f_i^*\left(\vec{G}\right) e^{-i\vec{G}\bullet\vec{r}}, \tag{4b}$$

and

$$u_i\left(\vec{G}\right) = \frac{1}{\Omega}\int_\Omega d^2r u_i\left(\vec{r}\right) e^{-i\vec{G}\bullet\vec{r}}, \tag{4c}$$

$$f_i\left(\vec{G}\right) = \frac{1}{\Omega}\int_\Omega d^2r f_i\left(\vec{r}\right) e^{-i\vec{G}\bullet\vec{r}}, \tag{4d}$$

where Ω is the surface area of one period, G is the lattice vector in reciprocal space, and

$$\vec{G} = (G_x, G_y) = (2m\pi/\lambda_x, 2n\pi/\lambda_y), \quad m, n = 0, \pm 1, \pm 2 \ldots. \tag{5}$$

Substituting Eqs. (4) and (5) into Eq. (3), the strain relaxation energy per unit area E_{el} can be expressed in reciprocal space as

$$E_{el} = -\frac{1}{2} \sum_i \sum_{\vec{G}} u_i \left(\vec{G}\right) f_i^* \left(\vec{G}\right). \tag{6}$$

Using the convolution theorem for Eq. (2), we obtain the Fourier Transform of the displacement field as

$$u_i(\vec{G}) = \sum_j \chi_{ij}(\vec{G}) f_j(\vec{G}). \tag{7}$$

Substituting Eq. (7) into Eq. (6), we obtain the expression of E_{el}:

$$E_{el} = -\frac{1}{2} \sum_i \sum_j \sum_{\vec{G}} \chi_{ij}(\vec{G}) f_i^*(\vec{G}) f_j(\vec{G}). \tag{8}$$

The elastic Green's functions $\chi_{ij}(\boldsymbol{r}, 0)$ have the following forms [19]:

$$\chi_{xx}\left(\vec{r}, 0\right) = \frac{1-\nu^2}{\pi Y}\frac{1}{r} + \frac{\nu(1+\nu)}{\pi Y}\frac{x^2}{r^3}, \tag{9a}$$

$$\chi_{yy}\left(\vec{r}, 0\right) = \frac{1-\nu^2}{\pi Y}\frac{1}{r} + \frac{\nu(1+\nu)}{\pi Y}\frac{y^2}{r^3}, \tag{9b}$$

$$\chi_{xy}\left(\vec{r}, 0\right) = \chi_{yx}\left(\vec{r}, 0\right) = \frac{\nu(1+\nu)}{\pi Y}\frac{xy}{r^3}. \tag{9c}$$

Their Fourier Transforms are

$$\chi_{xx}(\vec{G}) = \frac{1-\nu^2}{\pi Y}\frac{2\pi}{G} + \frac{\nu(1+\nu)}{\pi Y}\frac{2\pi G_y^2}{G^3}, \tag{10a}$$

$$\chi_{yy}(\vec{G}) = \frac{1-\nu^2}{\pi Y}\frac{2\pi}{G} + \frac{\nu(1+\nu)}{\pi Y}\frac{2\pi G_x^2}{G^3}, \tag{10b}$$

$$\chi_{xy}(\vec{G}) = \chi_{yx}(\vec{G}) = -\frac{\nu(1+\nu)}{\pi Y}\frac{2\pi G_x G_y}{G^3}, \tag{10c}$$

Equations (8) and (10) are valid for any kind of periodic domain structures of a strained film surface. Particularly, for a one-dimension (1D) periodic domain structure, $G_y = 0$, Eqs. (8) and (10) can be simplified as [11]

$$E_{el} = -\frac{1}{2}\sum_{\vec{G}} \chi_{xx}(\vec{G})\left|f_x(\vec{G})\right|^2, \quad G_x = \frac{2m\pi}{\lambda}, \quad m = 0, \pm 1, \pm 2..., \tag{11}$$

$$\chi_{xx}(\vec{G}) = \frac{1-\nu^2}{\pi Y}\frac{2\pi}{G_x}. \tag{12}$$

For any given surface profile, we can calculate the elastic force density using Eq. (1) and the strain relaxation energy using Eqs. (8) and (10) (or Eqs. (11) and (12) for 1D systems). Combining the strain energy with surface energy, we can perform a thermodynamic analysis of film stability.

3 Total Energy of Strained Film with Undulated Surface

In this section, we present the properties of three 1D surface profiles of a strained film grown on a flat substrate. The strain induced surface undulation is characterized by a critical wavelength λ_c. For different surface profiles, the general scaling relation of λ_c with the surface energy and strain energy is the same, but having different geometric coefficients. We will derive the total energy for a sinusoidal film surface, a faceted film surface and a flat surface with faceted islands in Sects. 3.1, 3.2 and 3.3, respectively.

3.1 Sinusoidal Undulated Surface

For a sinusoidal surface profile of a strained film growing on a flat substrate (Fig. 1), the surface undulation is expressed as

$$h(x) = a\sin\frac{2\pi}{\lambda}x + h_0 = a\sin kx + h_0, \tag{13}$$

where $k = 2\pi/\lambda$ is the wave number. It leads to a distribution of elastic force monopoles in the surface, which can be calculated from the differential of bulk stress at the film surface as

$$f(x) = C\varepsilon\partial_x h(x) = C\varepsilon ak\cos kx, \tag{14}$$

where C is the Young's modulus and ε is the misfit strain, $C\varepsilon$ is the bulk stress in the film. To calculate the strain energy, we derive the Fourier Transform of the force density,

$$f(G_x) = \begin{cases} \frac{C\varepsilon ak}{2}, G_x = \pm k \\ 0, other \end{cases}. \tag{15}$$

Substituting Eqs. (15) and (12) into Eq. (11), we obtain the strain energy per unit area:

$$E_{el} = -(C\varepsilon)^2 \cdot \frac{(a\pi)^2}{\lambda} \cdot \frac{1-\nu^2}{\pi Y}. \tag{16}$$

On the other hand, the surface undulation will increase surface energy. As shown in Fig. 1, the surface energy per period of a flat surface is $E_s{}^0 = \gamma\lambda$. The surface energy per period of a sinusoidal surface can be expressed as

$$E_s = \gamma \cdot l = \gamma \cdot \int_0^\lambda \sqrt{1 + h'^2(x)}dx. \tag{17}$$

Since we adopt a shallow-angle approximation and only consider the critical wavelength in this subsection, we will neglect the surface energy anisotropy effect for simplicity. (The surface energy anisotropy effect would not affect the qualitative results, but change the coefficients). Then substituting Eq. (13) into Eq. (17) and assuming that $a \ll \lambda$, we have

$$E_s = \gamma\left(\lambda + \frac{a^2\pi^2}{\lambda}\right). \tag{18}$$

And the increase of surface energy is

$$\Delta E_s = E_s - E_s^0 = \gamma \cdot \frac{a^2\pi^2}{\lambda}. \tag{19}$$

The total energy per period is then

$$E = \Delta E_s + E_{el} = \gamma \cdot \frac{a^2\pi^2}{\lambda} - (C\varepsilon)^2 \cdot (a\pi)^2 \cdot \frac{1-\nu^2}{\pi Y}. \tag{20}$$

Notice that for a sufficiently large λ, E becomes negative, which implies that there exists a critical wavelength λ_c beyond which the undulated surface is always more stable than the flat surface. The critical wavelength λ_c obtained by using the condition $E = 0$ is

$$\lambda_c = \frac{\gamma}{E_s}, \tag{21}$$

where $E_s = (1-\nu^2)(C\varepsilon^2)/(\pi Y)$ represents the unit strain energy.

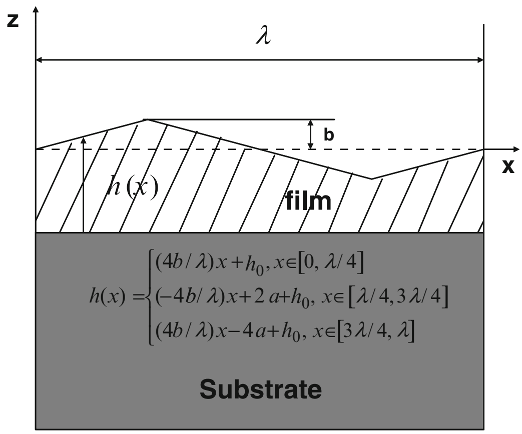

Fig. 2 Schematic illustration of faceted strained film on a flat substrate

3.2 *Faceted Surface*

For a faceted surface profile, as shown in Fig. 2, we can do the same analysis as in Sect. 3.1. The surface undulation is expressed as

$$h(x) = \begin{cases} (4b/\lambda)x + h_0, x \in [0, \lambda/4] \\ (-4b/\lambda)x + 2a + h_0, x \in [\lambda/4, 3\lambda/4] \\ (4b/\lambda)x - 4a + h_0, x \in [3\lambda/4, \lambda] \end{cases}. \tag{22}$$

The force monopole distribution is

$$f(x) = C\varepsilon \cdot \begin{cases} tg\theta, x \in [0, \lambda/4] \\ -tg\theta, x \in [\lambda/4, 3\lambda/4] \\ tg\theta, x \in [3\lambda/4, \lambda] \end{cases}. \tag{23}$$

The Fourier transformation of the force density is

$$f(G_x) = \frac{1}{\lambda}\int_0^{\lambda} f(x) e^{iG_x \cdot x} dx = \frac{C\varepsilon tg\theta}{\lambda} \cdot \frac{1}{mk} \cdot \begin{cases} 4(-1)^{(m+1)/2}, \ m - odd \\ 0, \ m - even \end{cases}. \tag{24}$$

The strain relaxation energy is then

$$E_{el} = -\frac{7(C\varepsilon)^2 tg^2\theta\lambda}{4\pi^2} \cdot \frac{1-\nu^2}{\pi Y} \cdot \zeta(3), \tag{25}$$

where ζ(3) is the Riemann zeta function, defined by

$$\zeta(n) = \sum_{k=1}^{\infty} \frac{1}{k^n}, \tag{26}$$

and $\zeta(3) = 1.2020569032\ldots$

The increase of surface energy is

$$\Delta E_s = E_s - E_s^0 = \gamma \cdot 4 \cdot \sqrt{\left(\frac{\lambda}{4}\right)^2 + b^2} - \gamma \cdot \lambda. \tag{27}$$

We also use the assumption that $b \ll \lambda$, but in this case, the surface contact angle of the facet is usually constant for a specific system, so we use θ as the variable instead of b ($b = \lambda \mathrm{tg}\theta/4$):

$$\Delta E_s = \gamma\lambda tg^2\theta/2. \tag{28}$$

The surface total energy per period is then

$$E = \Delta E_s + E_{el} = \gamma\lambda tg^2\theta/2 - \frac{7\lambda^2(C\varepsilon)^2 \cdot \zeta(3) tg^2\theta}{4\pi^2} \cdot \frac{1-\nu^2}{\pi Y}, \tag{29}$$

and the critical wavelength is

$$\lambda_c = \frac{2\pi^2}{7\varsigma(3)} \frac{\gamma}{E_s}, \tag{30}$$

Note that in the above derivation, we neglect the corner effect.

3.3 Isolated Faceted Island

For a faceted island grown on a flat substrate, as shown in Fig. 3, the surface profile can be expressed as

$$h(x) = \begin{cases} 0, x \in [-\lambda/2, -s] \\ (a/s)\, x + a, x \in [-s, 0] \\ (-a/s)\, x + a, x \in [0, s] \\ 0, x \in [s, \lambda/2] \end{cases}. \tag{31}$$

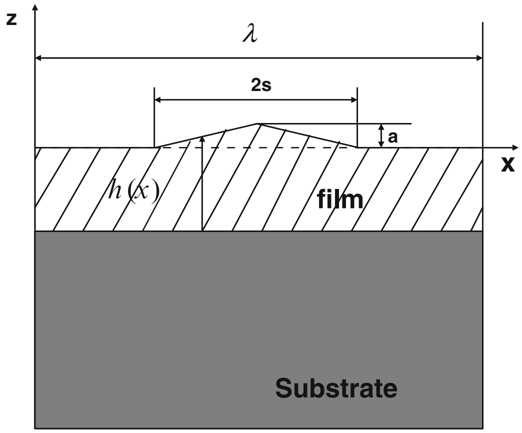

Fig. 3 Schematic illustration of periodic SK-grown faceted island on a flat substrate

where a is the height of the island, and s is length of the island, $a = s$*tgθ. The force monopoles are only distributed on the surface of island,

$$f(x) = C\varepsilon \cdot \begin{cases} 0, x \in [-\lambda/2, s] \\ tg\theta, x \in [-s, 0] \\ -tg\theta, x \in [0, s] \\ 0, x \in [s, \lambda/2] \end{cases}. \tag{32}$$

The Fourier transform of the force monopole density is

$$f^*(G_x) = -\frac{4C\varepsilon tg\theta}{\lambda} \cdot \frac{i}{G_x}\sin^2\frac{G_x s}{2}. \tag{33}$$

The strain relaxation energy is then

$$E_{el} = -(C\varepsilon)^2 \cdot \frac{1-\nu^2}{\pi Y} \cdot \left[4\ln 2s^2 tg^2\theta - \frac{\pi^2}{3\lambda^2}s^4 tg^2\theta\right]. \tag{34}$$

To obtain Eq. (34), we assume $s \ll \lambda$, which means the island-island distance is much larger than the island size, to differentiate from the case of a faceted surface in Sect. 3.2. The first term in Eq. (34) is the strain relaxation energy of an isolated island; the second term, which is proportional to $(1/\lambda)^2$, comes from the interaction between islands, indicating that islands interact with each other like two elastic force dipoles [9]. The higher order terms are neglected.

The surface energy increases by

$$\Delta E_s = \gamma s tg^2\theta. \tag{35}$$

The total energy per period is then

$$E = \gamma s t g^2\theta - 4\ln 2 s^2 t g^2\theta (C\varepsilon)^2 \cdot \frac{1-\nu^2}{\pi Y} + \frac{\pi^2}{3\lambda^2} s^4 t g^2\theta (C\varepsilon)^2 \cdot \frac{1-\nu^2}{\pi Y}. \quad (36)$$

If we neglect the third term in Eq. (36), i.e., the dipolar island-island interaction, it reduces to the energy of an isolated island on the surface. Then, there is no critical wavelength involved. But there is a critical island size, defining the smallest stable island size beyond which the island will keep growing [4]. The critical size is calculated as

$$s_c = \frac{1}{4\ln 2}\frac{\gamma}{E_s}. \quad (37)$$

Inclusion of the repulsive island-island interaction (third term in Eq. (36)) would increase the critical island size, and also prevent the further growth of the island after nucleation favoring formation of islands with uniform size [19].

From Eqs. (21), (30) and (37) we can see that the critical length scale for different surface profiles has the same scaling dependence on the ratio of surface energy (γ) over strain energy (E_s), but different coefficients. The critical wavelength of sinusoidal undulation has a coefficient 1, and that of a faceted undulation has a coefficient $2\pi^2/7\zeta(3)$, larger than 1. If we assume that the surface energy is isotropic and neglect corner effects of facets, the faceted surface will have a larger critical wavelength than that of a sinusoidal surface undulation. The coefficient for the critical island of an isolated island is the smallest [1/(4ln2)], because the island formation is the most effective mode of strain relaxation with the largest undulation magnitude for the same film volume.

In our analysis, we neglect the surface energy anisotropy effect, so that the surface energy and strain relaxation energy of a sinusoidal film are both proportional to the square of the undulation amplitude. If taking the surface energy anisotropy into account, the form of strain relaxation energy remains the same, but the surface energy is more complicated and increases much faster than the second-order power dependence on undulation amplitude. Then for a given wavelength, when the magnitude of the surface undulation is very small, the sinusoidal surface is more stable; but the faceted surface and the isolated faceted islands become more stable when surface the undulation becomes larger. This suggests that the surface may first undulated by forming stepped mounds in a sinusoidal profile. As the mounds grow higher, they transform into faceted islands as observed in some experiments and explained theoretically before [20, 21].

4 Conclusion

In conclusion, we perform a thermodynamic analysis for the strained film grown on a flat substrate, using a Green's function force monopole approach in reciprocal space. Our results agree generally with the previous work based on a stress function approach. We compared three different surface undulations: sinusoidal waviness, surface faceting and island formation. The critical length scale for the strain induced instability has the same linear dependence on the ratio of surface energy and strain energy, but different geometric coefficients. Among them, the isolated islands have the smallest critical length scale without consideration of surface energy anisotropy, because it offers the most effective mode of strain relaxation. Inclusion of surface energy anisotropy, however, may favor surface sinusoidal undulations first, followed by surface faceting or faceted island formation, as observed in some systems. We are extending our approach to non-flat substrate surfaces.

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