Enhanced growth instability of strained film on wavy substrate

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We demonstrate that the growth of a strained film is inherently less stable on a wavy substrate than on a flat substrate. For small surface undulation, the lowest strain energy state is for the film surface to adopt the same wavelength as the substrate surface in an antiphase configuration at the early stage of growth. The critical wavelength (λ_c) of growth instability on a wavy substrate is half of that on a flat substrate (λ_0). It increases linearly with increasing film thickness (t) as $\lambda_c = \lambda_0/2 + \pi t$. Implications for strain directed self-assembly on patterned substrate are discussed.
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I. INTRODUCTION

The study of morphological instability of strained thin film is of great scientific interest as well as of technological importance. Strain induced self-assembly provides an attractive route to nanofabrication of quantum dots and quantum wires.[1] However, when grown on a flat substrate, the self-assembled nanostructures are in general not yet uniform enough to be used in practical applications. Recent efforts have been made to combine the strain induced self-assembly with surface patterning in an effort to further improve the size uniformity and spatial ordering of nanostructures.[2–6]

The growth instability of a strained film on a flat substrate is well understood, generally referred to as Asaro-Tiller-Grinfeld (ATG) instability.[7–9] Due to lattice mismatch, misfit strain drives a flat surface to an undulated surface or a surface containing faceted islands, which are, respectively, characterized by a critical wavelength[10,11] or a critical size.[12–14] However, the growth instability of a strained film on a nonflat substrate is much less understood with rather limited experimental and theoretical studies. Here, we perform a linear stability analysis of a strained film on a wavy surface, which will not only advance our fundamental knowledge of thin film growth but will also shed some light on the understanding of strain directed self-assembly on patterned substrate.

On a flat surface, the growth stability is defined by the critical wavelength $\lambda_0 = \pi M \gamma / \sigma^2$, where M is the elastic modulus, γ is the surface tension, and σ is the nominal film stress proportional to misfit strain.[10,11] The growth is unstable if the wavelength of surface undulation is larger than λ_0 and stable vice versa. Physically, the larger (smaller) the surface (strain relaxation) energy is, the more stable the growth is, having a longer critical wavelength. In contrast, on a wavy substrate, we show that the growth becomes inherently less stable, having a critical wavelength (λ_c) essentially half of that on a flat substrate (λ_0). In general, the growth is always stable if the film surface undulates with a wavelength smaller than $\lambda_0/2$ and unstable with a wavelength larger than λ_0, independent of substrate surface undulation. For the film surface undulating with a wavelength in between $\lambda_0/2$ and λ_0, the growth is unstable if the substrate undulation contains the same wavelength component and stable otherwise.

We perform a two-dimensional (2D) linear stability analysis for a compressively strained thin film growing on a sinusoidal substrate surface as an arbitrary surface undulation can be Fourier transformed into a sinusoidal series of different wavelengths. We consider that the amplitude of undulation is small compared to the wavelength. The elastic constants of the film and substrate are assumed to be the same. In general, the film surface may adopt an undulation of different wavelengths and phases from the substrate, and we can express the film surface profile as $h_f(x) = A_f \sin(k_f x)$ and the substrate surface profile as $h_s(x) = A_s \sin(k_s x + \alpha) - t$ as shown in Fig. 1, where A_f (A_s) is the amplitude and k_f (k_s) is the wave number of the film (substrate) surface undulation. Note that the minimum t must be larger than A_s to retain a continuous film for our analysis to be applicable.

II. DERIVATIONS AND RESULTS

First, we show that at the early stage of growth when the film is thin the elastic interaction energy between a film surface undulation and a substrate surface undulation of different wavelengths vanishes so that to minimize the strain energy the film tends to adopt the same wavelength as the substrate in an antiphase configuration. The stress in the x-direction on the film surface can be calculated to the first order as[15–17]

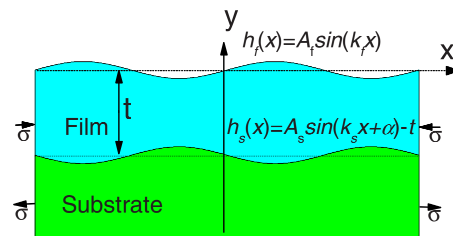

FIG. 1. (Color online) A schematic diagram of a film with a sinusoidal surface growing on a sinusoidal substrate.

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$$\sigma_{xx} = \sigma - 2\sigma A_f k_f \sin(k_f x) + 2\sigma A_s k_s e^{-k_s t} \sin(k_s x + \alpha), \quad (1)$$

where σ is the nominal biaxial stress in the film. The first two terms are stresses on a flat substrate as derived before.[10] The additional third term results from the buried substrate surface waviness, which is derived from the stress field induced by embedded stressors of any shape (such as embedded islands or wires) through Fourier transformation as discussed in Ref. 17. The stress in the y-direction is neglected since the amplitude of undulation is small. Note the sign difference of the second and third terms in Eq. (1), which can be understood by thinking of the case with a wavy film surface on a flat substrate surface giving rise to the second term versus the case with a flat film surface on a wavy substrate giving rise to the third term. In the former (latter) case, the local film volume is increased (decreased) in the peak region of the film (substrate) surface undulation but decreased (increased) in the valley region so that the normal compressive surface stress is relaxed (enhanced) in the peak region but enhanced (relaxed) in the valley region.

The strain energy density along the top surface can then be calculated as[11]

$$w(x) = w_0[1 - 4k_f A_f \sin(k_f x) + 4A_s k_s e^{-k_s t} \sin(k_s x + \alpha)], \quad (2)$$

where $w_0 = (1-\nu)\sigma^2/4\mu$, ν is Poisson's ratio, and μ is Young's modulus. In deriving Eq. (2), we neglected the higher-order terms of A_f^2, A_s^2, and $A_f A_s$.

To minimize strain energy, we use a variational approach[11] and take the strain energy variation with respect to the film surface undulation as

$$\frac{\delta U}{\delta A_f} = \int w(x) \sin(k_f x) dx. \quad (3)$$

For the general case of $k_f \neq k_s$ and assuming that the film surface consists of m (integer) periods of sinusoidal waves, we integrate Eq. (3) over the whole surface and obtain the average energy variation over one period,

$$\frac{\delta U}{\delta A_f} = -4\pi w_0 A_f - \frac{4w_0 A_s e^{-k_s t}}{m} \frac{k_f k_s}{(k_f^2 - k_s^2)} \times \left[\sin\left(\frac{2m\pi k_s}{k_f} + \alpha \right) - \sin \alpha \right]. \quad (4)$$

One can show that if the film surface waviness (wavelength) is commensurate with the substrate surface waviness, i.e., $m\lambda_f = n\lambda_s$ (n is an integer), then the second term in Eq. (4) vanishes. If the two are incommensurate, i.e., $m\lambda_f \neq n\lambda_s$, then we must take $m \to \infty$, and the second term also vanishes. Therefore, for $k_f \neq k_s$, we always have $(\delta U / \delta A_f) = -4\pi w_0 A_f$, i.e., the film strain energy varies with the film surface undulation only, independent of the substrate surface undulation. In other words, the elastic interaction energy between a film surface wave of k_f and a substrate surface wave of k_s is zero, to the first order, when $k_f \neq k_s$. Thus, the dominant contribution to varying strain energy comes from the film surface undulation adopting the same wave number as the substrate.

For $k_f = k_s = k$, the strain energy variation (i.e., strain relaxation energy) per period can be evaluated as

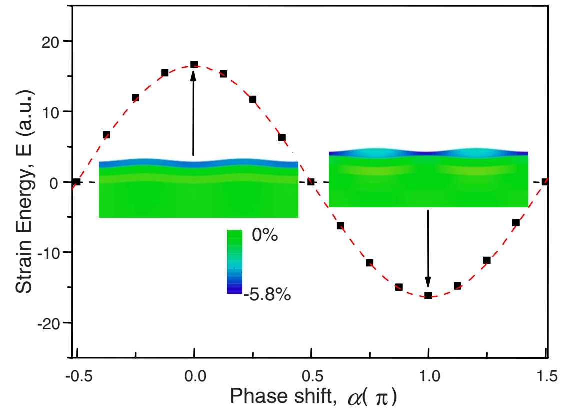

FIG. 2. (Color online) The strain energy E as a function of phase shift α obtained from FEA calculations. The energy on a flat substrate is set as the reference energy ($E=0$). The (red) dashed line is a fit using a cos function to the calculated data (squares). The insets show the stress distributions at the maximum-energy ($\alpha=0$) and minimum-energy ($\alpha=\pi$) configurations, respectively.

$$\Delta U = \int_0^{A_f} \frac{\delta U}{\delta A_f} dA_f = -2\pi w_0 [A_f^2 - 2A_f A_s e^{-kt} \cos \alpha]. \quad (5)$$

The first term is the strain relaxation energy on a flat substrate. The second term results from the buried substrate surface undulation interacting with the film surface. Equation (5) shows that for $-\pi/2 < \alpha < \pi/2$, the strain energy on a wavy substrate is higher than that on a flat substrate, and the maximum energy occurs at $\alpha = 0$, i.e., the in-phase configuration. On the contrary, for $\pi/2 < \alpha < 3\pi/2$, the reverse is true, and the minimum strain energy occurs at $\alpha = \pi$, the antiphase configuration.

The analytical results are further confirmed by finite element analysis (FEA) calculations[18,19] as shown in Fig. 2. The calculated data follow almost exactly the analytical expression of Eq. (5). The FEA provides useful hints on understanding the physical origin of strain energy variation. The two insets in Fig. 2 show the calculated stress distributions at the two extreme configurations. At the in-phase configuration (left inset), the stress in the film is rather uniform because of the uniform film thickness, so the film is uniformly stressed without much relaxation. Whereas at the antiphase configuration (right inset), the stress is highly modulated into a stress-domain pattern caused by the largest film thickness variation with alternating tensile and compressive domains in the valley region of the substrate (the thick film region) and in the peak region of the substrate (the thin film region), respectively. Consequently, the antiphase configuration has the minimum strain energy through the formation of stress domains as an effective mechanism for strain relaxation.[1]

The above results indicate that if the film surface undulates with a wavelength different from the substrate surface undulation, the film growth stability will be the same as that on a flat substrate because the elastic energy will contain only the strain relaxation energy due to the film surface undulation as if the substrate was not undulated. So, for any $\lambda_f \neq \lambda_s$, the film growth stability is defined by the same criti-

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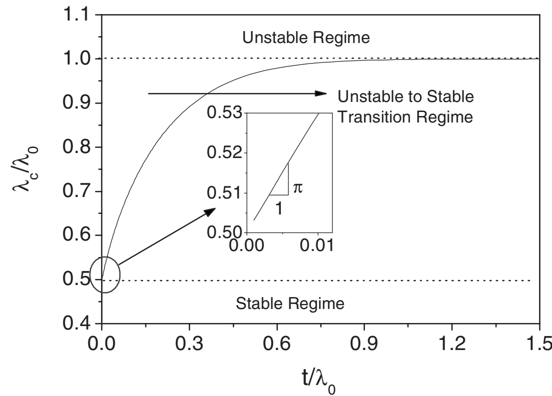

FIG. 3. The dependence of the critical wavelength λ_0 on the film thickness (t), both normalized with λ_0.

cal wavelength λ_0 on the flat substrate. The only relevant film wavelength that needs to be examined is the one that adopts the substrate wavelength.

We perform a linear stability analysis by taking $k_s=k_f=k$ and $\alpha=\pi$. At the limit of small undulation, the chemical potential in the film surface can be calculated as[10,20]

$$\mu=\mu^*+\gamma\Omega\kappa-\frac{2k\Omega\sigma^2}{M}(A_f+A_s e^{-kt})\sin(kx), \quad (6)$$

where μ^* is the chemical potential of the flat film surface bounding the substrate, Ω is the atomic volume, and κ is the curvature of the film surface. The stability can be analyzed by evaluating the chemical potential difference at the peak and the valley of the film surface as

$$\Delta\mu=2A_f k^2\gamma\Omega-\frac{4k\Omega\sigma^2}{M}(A_f+A_s e^{-kt}). \quad (7)$$

The sign of $\Delta\mu$ determines growth stability. For $\Delta\mu>0$, the chemical potential at the peak is higher than that at the valley so atoms diffuse from the peak to the valley, smoothening the surface and stabilizing the growth. For $\Delta\mu<0$, the reverse is true and the growth becomes unstable. The critical condition defining the growth instability is set by $\Delta\mu=0$, which leads to

$$\lambda_c\left(1+\frac{A_s}{A_f}e^{-2\pi t/\lambda_c}\right)=\lambda_0. \quad (8)$$

From Eq. (8), we see that when A_s goes to zero, $\lambda_c=\lambda_0$ as desired for the limiting case of a flat substrate. One interesting point is that the strain induced interaction between the film and substrate surface undulation makes critical wavelength λ_c dependent on film thickness t. To better reveal such a dependence, in the following analysis we will assume that $A_f=A_s=A$ for simplicity. We note that when the film becomes thick, A_f may not be the same as A_s, but the qualitative features that we obtain remain the same (see simulation results in Fig. 3 below). Then, at the initial stage of growth, when t is very small, the critical wavelength on a wavy substrate is half of that on a flat substrate ($\lambda_c\simeq\lambda_0/2$). It increases linearly with increasing film thickness with the slope of π,

$$\lambda_c=\frac{\lambda_0}{2}+\pi t \quad (t/\lambda_0\ll 1). \quad (9)$$

We note that on a flat surface, the critical wavelength may also depend on film thickness if the elastic constants of the film and substrate are different.[21] However, the physical origin and consequence of that thickness dependence are different from what we discuss here.

III. DISCUSSIONS

In principle, the linear stability analysis is valid only for small film thickness because nonlinear high-order effects and other forms of instability (e.g., dislocation formation) take place when the film grows thick. Nevertheless, it is useful to understand the general behavior of λ_c over the whole range of t and examine its asymptotic limit within our theoretical model. In Fig. 3 λ_c is shown to vary from $\lambda_0/2$ to λ_0 with increasing film thickness, approaching λ_0 when $t\gg\lambda_0$. This is consistent with our physical intuition that for a very thick film, the effect of the film/substrate interface undulation is negligible, as if it were a flat interface.

Theoretically, for the film surface to undulate with the same wavelength as the substrate surface undulation (λ_s), there could be three distinct regimes of growth stability defined by λ_s. If $\lambda_s\leq\lambda_0/2$, the growth is stable, evolving toward a smooth surface. If $\lambda_s\geq\lambda_0$, the growth is unstable, evolving toward a larger magnitude of undulation. In between if $\lambda_0/2<\lambda_s<\lambda_0$, the growth is initially unstable because $\lambda_s>\lambda_c\simeq\lambda_0/2$ when t is small, and then converts to be stable because $\lambda_s<\lambda_c\simeq\lambda_0$ when t is large. The transition from unstable-to-stable growth occurs at a critical film thickness $t_c=-(\lambda_s/2\pi)\ln(\lambda_s/\lambda_0-1)$.

We have carried out computer simulations to confirm the conclusions drawn from the linear stability analysis. We simulate the surface evolution of a strained film grown on a wavy substrate of different undulation wavelengths. Consider the case when surface evolution is dominated by surface diffusion. The equation of motion of the surface height profile is given by[22]

$$\frac{\partial h(x)}{\partial\tau}=M\frac{\partial^2\mu}{\partial s^2}+R, \quad (10)$$

where $h(x)$ is the surface height at position x, τ is the evolution time, M is the mobility constant (related to surface diffusivity, atomic volume, and temperature), s is the arc length, and R is the deposition rate. To obtain a close-to-equilibrium growth morphology, a small R is used. In the simulation, A_f changes with time while A_s remains fixed.

Figure 4 shows the simulation results for three typical cases illustrating different regimes of instability, consistent with theoretical analysis. In essence, the early stage of growth (i.e., for small t) is characterized by a critical wavelength $\lambda_s\simeq\lambda_0/2$, below which the growth is stable [Fig. 4(a)] and above which unstable [Figs. 4(b) and 4(c)]. Theoretically, if t were allowed to grow big, the growth would be divided into three regimes as shown, respectively, in Figs. 4(a)–4(c), which is consistent with Fig. 3.

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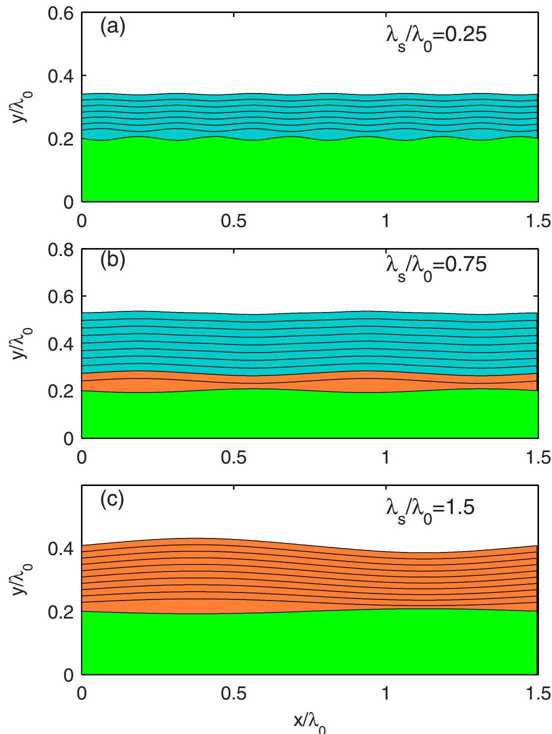

FIG. 4. (Color online) Typical simulation results ($M=0.01$) illustrating growth instability of a strained film on a sinusoidal substrate with the wavelength λ_s: (a) $\lambda_s/\lambda_0=0.25$, stable growth (cyan colored); (b) $\lambda_s/\lambda_0=0.75$, unstable-to-stable growth (cyan to orange); and (c) $\lambda_s/\lambda_0=1.5$, unstable growth (orange).

Now, we can put together all the results to have a general picture of growth stability for the film to undulate with an arbitrary wavelength. For the initial film growth on a sinusoidal substrate surface, the stability for the film surface to undulate with the same wavelength as the substrate surface is defined by $\lambda_0/2$ while the stability for the film surface to undulate with any other wavelength is defined by λ_0. If the film surface undulates with a wavelength longer than λ_0, its growth is always unstable independent of substrate undulation. If the film surface undulates with a wavelength shorter than $\lambda_0/2$, its growth is always stable independent of substrate undulation. If the film surface undulates with an intermediate wavelength between $\lambda_0/2$ and λ_0, its growth stability depends on the wavelength of substrate undulation: stable on the substrate undulation having a different wavelength but unstable on the substrate undulation having the same wavelength. So, effectively the critical wavelength is $\lambda_0/2$ on a wavy substrate containing all possible wavelengths including those in between $\lambda_0/2$ and λ_0. However, the critical wavelength is still λ_0 (same as on a flat substrate) if the wavy substrate contains only the wavelengths smaller than $\lambda_0/2$ and/or wavelengths larger than λ_0. This includes the case when the wavy substrate wavelength goes to infinity approaching the limiting case of flat substrate; hence, the critical wavelength is λ_0 as expected. All these possible scenarios can be summarized in a stability diagram shown in Fig. 5.

For the film growth on a substrate surface of arbitrary shape, which can be expressed as a linear combination of sinusoidal waves of different wavelengths,[23] the film surface will initially undulate in a manner also having a linear com-

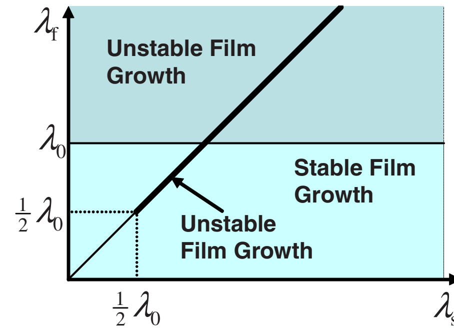

FIG. 5. (Color online) Stability diagram for arbitrary film wavelength (λ_f) and substrate wavelength (λ_s). λ_0 is the critical wavelength on a flat substrate.

bination of waves of different wavelengths. Then, those wave components with a wavelength longer than λ_0, which are always the unstable ones, will remain all the time. Those wave components with a wavelength shorter than $\lambda_0/2$, which are always the stable ones, will quickly die out. Those wave components with an intermediate wavelength between $\lambda_0/2$ and λ_0, which are initially unstable and then become stable, will gradually die out. So, the film surface will evolve with a changing overall undulation composed of surviving wave components.

Our results are obtained for the case of small substrate undulation magnitude. For growth on a patterned substrate with large surface undulation, an exact analytical treatment is difficult as the problem becomes more complex and physically less transparent.[24,25] Nevertheless, some implications for strain directed self-assembly on patterned substrates may be drawn from our analysis. The critical wavelength is reduced on a patterned substrate, which may explain the experimental observation that the average size of quantum dots on patterned substrates is smaller than that on flat substrates. Experiments show that quantum dots may form either at the peak[2,3] or at the valley/sidewall[4] of the patterns under different conditions. This can be qualitatively understood in terms of directed adatom diffusion from valley to peak or from peak to valley, favoring island nucleation at the peak or valley, respectively, as identified by our analysis in different regimes of growth instability. Also, local variations in surface undulation (wavelength or curvature) may create local chemical potential minima, guiding the island nucleation locally.[5]

IV. CONCLUSIONS

In conclusion, we have provided a linear stability analysis for strained thin films grown on wavy substrates. We demonstrate that the growth is inherently less stable as manifested by a smaller critical wavelength about half of that on flat substrates. This is caused by more effective strain relaxations by film surface undulation interacting with substrate surface undulation in the lowest strain energy state when the two surfaces adopt the same wavelength and are antiphase with each other. We show that the critical wavelength increases linearly with film thickness at the initial stage of

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growth. Our analysis also gives some qualitative understanding of certain aspects of directed self-assembly of quantum dots on patterned substrates.

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