Strain effect on the instability of island formation in submonolayer heteroepitaxy

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Abstract – A theoretical model is developed to study the strain effect on the instability of island formation in submonolayer heteroepitaxy in both thermodynamic and growth kinetic regimes. By using the linear-stability analysis, the elastic energy change of a circular island is derived. Combined with a rate equation theory, the interplay between growth kinetics and strain effect on the shape instability is analyzed and illustrated with the constructed morphological phase diagrams. Critical island sizes beyond which islands grow unstable are also derived and can be used to estimate the energy parameters. Well-defined scaling properties are also obtained for the shape instability.

Understanding the growth mechanisms in the early stages of epitaxial growth is crucial for developing advanced high-quality devices [1–3]. For example, the mechanism of strain-induced self-organization of nanostructures in heteroepitaxy has been explored extensively since it offers a promising route for the fabrication of optoelectronic devices [4]. While various growth models have been developed for submonolayer homoepitaxy [1–3], these models provide very limited understanding for the growth in heteroepitaxy where strain arising from the lattice mismatch plays an important role [4]. So far, submonolayer heteroepitaxy still remains as a poorly understood subject.

One of key issues in submonolayer heteroepitaxy is the shape of strained 2-Dimensional (2D) islands. Strain-induced shape instability during island formation has been studied experimentally and theoretically [5–10], but most theoretical studies focused on the thermodynamic regime [5,9,10]. It has been found that the competition between strain energy and step free energy leads to a shape transition from compact to elongated or fractal [5–8]. However, what roles do the growth kinetics play in the shape instability? How does strain interplay with

kinetics during island formation? These issues are very important because the desired surface morphologies can be obtained via proper manipulation of growth kinetics, which has been demonstrated in recent studies for the step bunching instability observed in a strained film grown on a vicinal surface [11–13]. Unfortunately, these issues are still not clear in strain-induced shape instability and no comprehensive model was proposed.

In this letter we develop a comprehensive model to study the strain effect on the instability of island formation in heteroepitaxy in both thermodynamic and kinetic regimes. The elastic energy change is derived analytically for a strained 2D circular island using the linear-stability analysis [14]. Combined with a self-consistent rate equation theory [15], we analyze the shape instability in various conditions. Morphological phase diagrams are constructed to illustrate the interplay between growth kinetics and strain effect on the shape instability, providing a guide for manipulating growth kinetics in heteroepitaxy to obtain the desired surface morphologies. The critical island size beyond which islands grow unstable is derived, which can be used to estimate the energy parameters of given systems. More analyses show well-defined scaling properties in the strain-induced shape instability.

First we consider a circular island with radius R and height h under isotropic stress on a substrate for

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simplicity. In order to analyze the shape instability, we examine small perturbations to this circular island. Here only the shape perturbation is considered [16]. The radius of the perturbed island can be expressed as $r(\phi) = R + A_k \cos(k\phi)$. Here R is the unperturbed radius, $A_k (\ll R)$ is the amplitude of the perturbations, and k is the wave number of the perturbations. If the perturbation increases the energy, the island will be stable; otherwise, it is unstable. We will investigate the energy change of the island under perturbations.

For a strained island, if there is no dislocation at the interface, its elastic energy is [17]

$$E_e = C \int \mathrm{d}^2\mathbf{r} \int \mathrm{d}^2\mathbf{r}' \frac{\Theta(\mathbf{r})\Theta(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|^3}, \qquad (1)$$

where $\Theta(\mathbf{r})$ is the step function, which is one, if $|\mathbf{r}| < r(\phi)$ and zero, otherwise. The constant C is determined by

$$C = \frac{Y_a^2(1-\nu_s^2)}{\pi Y_s(1-\nu_a)^2} h^2 f^2. \qquad (2)$$

Here Y is Young's modulus and ν is Poisson's ratio. The subscripts s and a represent the substrate and film, respectively. $f = (a_a - a_s)/a_s$ is the lattice mismatch with lattice constants a_s and a_a for the substrate and the film, respectively. h is the island height. In the submonolayer growth regime $h \ll R$. Here for simplicity, the edge stress and interfacial stress are neglected in our model [5]. In our analysis the lattice constants on the fcc(100) surface are used as the unit length.

Under a small perturbation of A_k, the change of the elastic energy can be derived as

$$\delta E_e = -\pi C A_k^2 \alpha(R,k)/R, \qquad (3)$$

here

$$\begin{aligned} \alpha(R,k) = {}& (k^2-1)\ln\left(\frac{4R}{a}\right) + 1 \\ & + \sum_{n=1}^{k}\left[\frac{2n(n-1)}{2k-2n+1} - \frac{4k^2-1}{2n-1}\right]. \end{aligned} \qquad (4)$$

Note that the area of the island keeps constant before and after perturbation. Here a is a cutoff length in the range of the surface lattice constant.

It is obvious that δE_e is positive as $k=1$. For other modes, δE_e is positive or negative depending on the island radius. Figure 1 shows that α is negative when islands are very small, but becomes positive as the island size increases. Therefore, as an island is very small, δE_e is positive, so the perturbation increases the elastic energy of the island, and the island will be stable. Otherwise, the island will become unstable. This indicates that a critical island radius exists.

The perturbations also make the periphery length of the perturbed island longer than that of the circular one, which attempts to increase the step energy of the island by
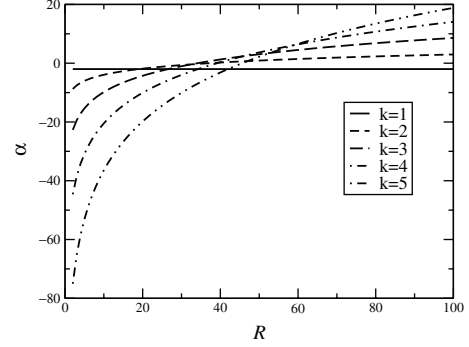
$$\delta E_s = \pi A_k^2(k^2-1)\gamma/(2R), \qquad (5)$$

Fig. 1: Variation of α with island radius R with different k.

where γ is the step energy per unit length. It is clear that the change of step energy attempts to stabilize the island shape.

As a result, the energy change caused by the perturbations is

$$\Delta E = \delta E_s + \delta E_e. \qquad (6)$$

For a given mode k, if $\Delta E > 0$, the circular island is stable against the perturbation; if $\Delta E < 0$, it is unstable. As mentioned above, the circular island is always stable under the perturbation of mode $k=1$, so the first unstable mode is $k=2$. We notice that δE_e is proportional to $\ln(R)/R$, while the step energy change $\delta E_s \propto 1/R$. When R is large enough, ΔE is always negative. Therefore, as $\Delta E(k=2) = 0$, the critical island size can be derived as

$$R_c = \frac{e^5 a}{4} \exp\left(\frac{\gamma}{2C}\right). \qquad (7)$$

Thus, once the radius of an island exceeds R_c, the strain will make the island grow unstable. As mentioned above, the strain-induced shape instability during island formation has been studied theoretically in the thermodynamic regime, where islands tend to grow along one direction [5,9].

Equation (7) shows that a larger step energy γ or a smaller lattice mismatch f leads to bigger stable islands. If $C = 0$, R_c will be infinite, which implies that unstrained circular islands are always stable in the thermodynamic regime. Note that R_c does not depend on temperature explicitly. According to eq. (6), the critical island radius increases with increasing k. This indicates that high-k modes are stable for very large islands, which is similar to the step meandering in the step flow growth [18].

The above analysis is for the island formation in equilibrium conditions. In most epitaxial experiments, the growth is far from equilibrium. The kinetic effect on island formation will be important. Next we will investigate the strain effect on the shape instability in the growth kinetic regime. Here we examine the time dependence of small perturbations to a strained 2D circular island $R(t)$ for its shape stability, which is determined by a relative stability function ω_k, so the radius of the island is replaced by

$r(\phi, t) = R(t) + A_k(t)\exp(ik\phi)$, where $R(t)$ is the unperturbed island radius. To the first order in $A_k(t)$, we have

$$\frac{\mathrm{d}}{\mathrm{d}t}\left(\frac{A_k}{R}\right) = \omega_k\left(\frac{A_k}{R}\right). \qquad (8)$$

If $\omega_k > 0$, the island grows unstable; if $\omega_k < 0$, it is stable. Thus, $\omega_k = 0$ defines a critical size. It is clear that once $\mathrm{d}R/\mathrm{d}t$ and $\mathrm{d}A_k/\mathrm{d}t$ are known, ω_k can be determined. $\mathrm{d}R/\mathrm{d}t$ and $\mathrm{d}A_k/\mathrm{d}t$ will be derived in the following in terms of the adatoms density surrounding the island and the mass conservation.

Here we consider a standard epitaxial growth including deposition, adatom diffusion on the substrate, aggregation to existing islands, and edge diffusion. We also consider reversible aggregation, where adatoms can overcome an energy barrier to detach from islands. On the surface, each island is surrounded by some others, and the environment is changing with time during growth. In order to describe the growth properties accurately, a self-consistent rate equation theory [15] is used to account for the influence of the environment. In this model, the strain-induced island-island interaction is not considered [19].

In the steady state, the adatom density $n(r,\phi)$ surrounding the island can be obtained by solving the diffusion equation [15]

$$\frac{1}{r}\frac{\partial}{\partial r}\left[r\frac{\partial n(r,\phi)}{\partial r}\right] + \frac{1}{r^2}\frac{\partial^2 n(r,\phi)}{\partial \phi^2} - \xi^{-2}\left[n(r,\phi) - n_0\right] = 0. \qquad (9)$$

Here ξ is the diffusion length and is defined as $\xi^{-2} = \sigma n_0 + \sigma_{\mathrm{av}}(N + n_0)$ [15]. N is the island density, n_0 is the average adatom density, and the average capture number σ_{av} measures the rate of islands capturing the adatoms on the surface. The solution to the above equation is

$$n(r,\phi) = n_0 + B_0 K_0\left(\frac{r}{\xi}\right) + B_k K_k\left(\frac{r}{\xi}\right)\exp(ik\phi), \qquad (10)$$

where $K_k(k = 0, 1, 2, \ldots)$ is the modified Bessel function of order k. $B_k(k = 0, 1, 2, \ldots)$ is a coefficient which can be determined by the boundary condition to eq. (9) at the island edge where the mass should be balanced,

$$\frac{\partial n(r,\phi)}{\partial r}\Big|_s = \beta\left[n(r,\phi)|_s - n_0(r,\phi)|_s\right]. \qquad (11)$$

Here s denotes the boundary of the island. β is a step kinetic coefficient which is related to the attachment barrier as adatoms aggregate to islands [15]. We assume no attachment barrier. $n_0(r,\phi)|_s$ is the equilibrium adatom density of the island.

On the other hand, doing the Taylor expansion, the adatom density $n(r,\phi)$ around the island can be written as

$$n(r,\phi)|_s \approx n(r,\phi)|_s + A_k \exp(ik\phi)\frac{\partial n(r,\phi)}{\partial r}\Big|_s, \qquad (12)$$

with first-order approximation.

For $n_0(r,\phi)|_s$, according to the Gibbs-Thomson relation, it can be written as [14]

$$n_0(r,\phi)|_s \approx n_{\mathrm{eq}}\left[1 + \frac{\Gamma}{R} + \frac{\Gamma}{R^2}(k^2 - 1)A_k\exp(ik\phi)\right], \qquad (13)$$

in the first order of A_k. Here n_{eq} is the equilibrium adatom density of an infinite island, and $\Gamma = \gamma\Omega/(k_{\mathrm{B}}T)$. Ω is the atomic volume.

Substituting eq. (12) and eq. (13) into the boundary condition eq. (11), then making use of eq. (10), we can determine the coefficients B_0 and B_k in eq. (10).

The rate of growth of the island is determined by the equation of mass conservation at the interface. With the considered perturbation the mass conservation equation can be written as [14]

$$\frac{\mathrm{d}R}{\mathrm{d}t} + \frac{\mathrm{d}A_k}{\mathrm{d}t}\exp(ik\phi) = j\Omega. \qquad (14)$$

Thus, $\mathrm{d}R/\mathrm{d}t$ and $\mathrm{d}A_k/\mathrm{d}t$ can be determined when the flux j is known. In fact, the flux j consists of two parts, the diffusive current of adatoms aggregating to islands as shown in eq. (11) and the diffusive current along island edges which is [20]

$$\mathbf{j}_e = D_e n_e \nabla\delta\mu/(k_{\mathrm{B}}T), \qquad (15)$$

where $\delta\mu(\phi) = (\Omega/R^2)[\gamma(k^2 - 1) - 2C\alpha(R, k)]A_k\cos(k\phi)$.

According to eq. (14) and eq. (8), finally one can obtain

$$\omega_k = \omega_k^1 + \omega_k^2 + \omega_k^3 - \frac{kF\Omega}{2}, \qquad (16)$$

in which

$$\omega_k^1 = D_t\Omega\xi^{-2}\left[n_0 - n_{\mathrm{eq}}\left(1 + \frac{\gamma - 2C\ln\frac{4R}{e^2 a}}{k_{\mathrm{B}}TR}\right)\right]$$
$$\times\frac{K_1(\frac{R}{\xi})}{K_0(\frac{R}{\xi})}\left[\frac{\xi(k-2)}{R} + \frac{K_{k-1}(\frac{R}{\xi})}{K_k(\frac{R}{\xi})} - \frac{K_0(\frac{R}{\xi})}{K_1(\frac{R}{\xi})}\right],$$

$$\omega_k^2 = \frac{D_t n_{\mathrm{eq}}\Omega}{k_{\mathrm{B}}TR^2}\left[2C\alpha - \gamma(k^2 - 1)\right]\left(\frac{K_{k-1}(\frac{R}{\xi})}{\xi K_k(\frac{R}{\xi})} + \frac{k}{R}\right),$$

$$\omega_k^3 = \frac{2D_e n_e\Omega C\alpha k^2}{k_{\mathrm{B}}TR^4} - \frac{D_e n_e\gamma\Omega k^2(k^2 - 1)}{k_{\mathrm{B}}TR^4}.$$

Here F is the deposition rate, T is the substrate temperature, k_{B} is Boltzmann's constant, and n_e is the number of atoms per unit length along the island edge. D_t and D_e are the diffusion rates on the substrate and along the island edge, respectively. In eq. (16), ω_k^1 is related to the instability caused by terrace diffusion. The second and third terms, ω_k^2 and ω_k^3, represent the competition between strain and step energy on the shape instability in the attachment-detachment–limited and edge-diffusion–limited growth regimes, respectively. The last term corresponds to the deposition on top of the

island, which can be neglected in further analysis. Note that for an irreversible homoepitaxy, eq. (16) recovers the equation derived in ref. [21]. This indicates that our theoretical model is more general.

For a compact island, R/ξ is a simple function of θ, here θ is the coverage [15]. Typically $\xi \gg R$, and since n_{eq} is very small compared to n_0, eq. (16) can be simplified as

$$
\omega_k \approx \frac{g_1(\theta)F\Omega}{NR^2}(k-2) + \frac{2D_e n_e \Omega C \alpha k^2}{k_{\text{B}}TR^4}
$$
$$
+ \frac{2D_t n_{\text{eq}}\Omega C \alpha k}{k_{\text{B}}TR^3}g_2(\theta) - \frac{D_e n_e \Omega \gamma k^2}{k_{\text{B}}TR^4}(k^2-1)
$$
$$
- \frac{D_t n_{\text{eq}}\Omega \gamma k}{k_{\text{B}}TR^3}(k^2-1)g_3(\theta). \tag{17}
$$

In the above equation, the first three terms are related to the deposition and strain effect which attempt to destabilize the island growth. The last two terms are related to the step energy which attempts to stabilize the island growth. Therefore, eq. (17) can be used to analyze the shape instability of an island caused by the interaction of deposition, strain effect, step energy, and surface kinetic processes. Here $g_i(\theta)$ $(i=1,2,3)$ are functions of θ and independent of system properties (C, γ, and α) and growth kinetics (D_e, D_t, F, n_{eq}).

According to eq. (17), deposition and strain may stabilize or destabilize the island growth, depending on the island radius and the perturbation modes (different k). However, edge diffusion and detachment processes in the last two terms attempt to decrease the step energy and stabilize the island growth. Therefore, the stability of the island growth depends on the perturbation modes. For the mode of $k=1$, the first two terms are negative and the last two terms are zero, so $\omega_k < 0$, and the island growth is always stable. In this case, both deposition and strain effect stabilize the island growth. For the modes of $k \geqslant 3$, deposition starts to destabilize the island growth, since more deposited atoms prefer to aggregate to the corners or protrusions of the existing larger islands [22], leading the island to grow unstable. Here the case of $k=3$ and $k=2$ for deposition and strain, respectively, will be discussed. For the last two terms, $k=2$ is used. In this case, the deposition flux causes the diffusion instability of the island shapes, and the strain effect destabilizes the island growth by forcing adatoms to diffuse to the less strained areas via edge diffusion or detachment processes.

As mentioned above, both deposition and strain can lead to island shape instability. But how do they interplay with each other? When deposition dominates the strain effect to destabilize the island growth, that is, the first term in eq. (17) is much larger than the second and third terms, we have

$$
\frac{F}{N} \gg \frac{24D_e n_e C}{k_{\text{B}}TR^2}\ln\left(\frac{4R}{e^5 a}\right) \tag{18}
$$

and

$$
\frac{F}{N} \gg \frac{12D_t n_{\text{eq}}C}{k_{\text{B}}TR}\ln\left(\frac{4R}{e^5 a}\right) \tag{19}
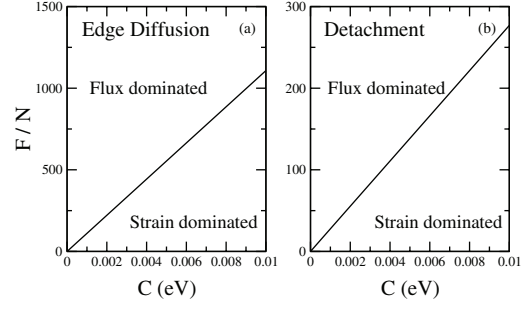$$

Fig. 2: Phase diagrams for the interplay between deposition and strain effect on the island shape instability. The shape is stabilized by edge diffusion (a) and attachment-detachment of atoms (b), respectively. Here $T = 300\,\text{K}$, $R = 50$, $\gamma = 0.1\,\text{eV}$, $D_e n_e = 10^5/\text{s}$, and $D_t n_{\text{eq}} = 10^4/\text{s}$ are chosen.

for edge diffusion and attachment-detachment processes as dominated stabilizing effects, respectively. The shape instability would be mainly caused by deposition in heteroepitaxial growth. In the other extreme, strain will dominate the shape instability.

Figure 2 shows the phase diagrams for the interplay between deposition and strain effect on the shape stability of an island with $R = 50$ (unit length = 1). The straight lines in fig. 2 divide the diagrams into two parts. In the region far above the lines where the deposition rate is relatively high, the island shape instability is mainly driven by deposition. In the region far below the lines, however, strain will dominate the shape instability, which corresponds to the regime of thermodynamic instability. But in the region close to the lines, both driving forces coexist, and may cause shape instability. For a given heteroepitaxial system, one can adjust the deposition rate to get stable compact islands, and further control the surface morphologies according to the interplay between growth kinetics and strain.

As mentioned above, however, for a specific system in certain growth conditions, islands may not grow unstable until their sizes exceed a critical value. According to eq. (17), in the deposition-dominated regime, a critical island radius can be derived as follows when edge diffusion dominates the stabilizing effect:

$$
R_1^c \approx \left(\frac{12D_e n_e \gamma}{k_{\text{B}}TF/N}\right)^{1/2}. \tag{20}
$$

Thus, the critical island size can be predicted based on the system properties and experimental conditions. On the other hand, according to $D_e \sim \exp(-E_e/k_BT)$, the edge diffusion barrier E_e can be estimated once the critical size is measured. We notice that a formula similar to eq. (20) was used to estimate the edge diffusion barriers in Ag/Pt(111) and Ag/Ag(111) systems [23]. However, eq. (20) contains more information which can be obtained from experiments. Thus, the prediction or estimate from eq. (20) should be more accurate [23,24]. This also confirms the validity and consistency of our model.
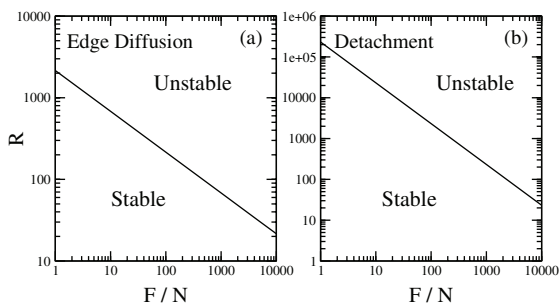
Fig. 3: Phase diagrams of the stable- and unstable-island shape plotted on the $(F/N, R)$-plane as deposition controls the shape instability. Parameters are the same as those in fig. 2.
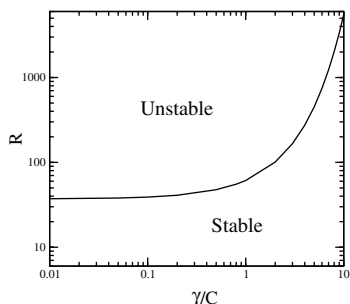

Fig. 4: Phase diagram of the stable- and unstable-island shape plotted on the (C, R)-plane as strain controls the shape instability.

When the attachment-detachment process dominates the stabilizing effect, the critical island radius is

$$R_2^c \approx \frac{6D_t n_{\mathrm{eq}}\gamma}{k_{\mathrm{B}}TF/N}. \tag{21}$$

One could also use the above expression to estimate the detachment barrier. But usually the edge diffusion barrier is much lower than the detachment barrier. Such growth mode might not be observed in epitaxial experiments.

Figure 3 shows the dependence of the critical island radius on the deposition flux when deposition dominates the island shape instability. In each phase diagram there are two growth regimes: unstable and stable. Below the lines, islands will grow stable and compact. Above the lines, however, islands will grow unstable. According to the phase diagrams of fig. 2 and fig. 3, one could adjust the growth kinetics in experiments to manipulate the surface morphologies.

When the deposition rate is very small and the deposition effect on the island shape instability can be negligible, the island growth goes back to the thermodynamic regime. Therefore, eq. (7) can be recovered from eq. (17). This also demonstrates the consistency and validity of our theoretical model. Figure 4 shows the phase diagram for shape instability in the thermodynamic regime.

From eqs. (20) and (21) we can predict the scaling behavior of shape instability in the submonolayer heteroepitaxy. With the relation $R \sim (D_t/F)^{-i/[2(i+2)]}$

(i is the critical nucleus) [1–3], scaling forms, $D_e/D_t \sim (D_t/F)^{(i-2)/(i+2)}$ and $D_{det}/D_t \sim (D_t/F)^{(i-4)/[2(i+2)]}$, can be derived for edge diffusion rate and detachment rate with deposition rate, respectively. The predicted scaling properties are consistent with previous results [21,25]. Here $D_{det} \approx D_t n_{\mathrm{eq}}\gamma/(k_{\mathrm{B}}T)$.

In summary, we developed a theoretical model to study the strain effect on shape instability of islands in both thermodynamic and kinetic regimes in heteroepitaxial growth. Our comprehensive model study provides new insight into the interplay between growth kinetics and strain effect in heteroepitaxy. The constructed phase diagrams and related derivations show how to obtain desired morphologies via proper manipulation of the growth kinetics in heteroepitaxy.

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