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Equilibrium morphologies of epitaxially strained islands

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An analytical expression of the free energy consisting of the strain energy, surface energy and interfacial energy for the coherent island/substrate system, as well as the evolving relations of aspect ratio against volume of the island, and misfit of the system, which provides a broad perspective on island behavior, is obtained, and used to study the equilibrium shapes of the systems. A two-dimensional model assuming linear elastic behavior is used to analyze an isolated island with elastic properties similar to those of the substrate. The results show that in order to minimize the total free energy, a coherent island will adopt a particular shape and height-to-width aspect ratio that are a function of only the island volume. The effect of a misfit dislocation on the equilibrium shape of an island is in passing examined. These can serve as a basis for interpretation of experiments.

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Models for coherent (dislocation-free) three-dimensional (3D) island (quantum dot) formation in heteroepitaxial thin film growth have recently been proposed [1–5]. Prediction and control of the islands' properties are essential for their technological applications, such as quantum dot lasers [6]. It is commonly accepted that depending on the magnitude of misfit compressive strain and the interfacial and surface energies, growth may proceed by layer-by-island (Stranski-Krastanov) mode, or at a higher misfit strain via direct islanding (Volmer-Weber) growth [7]. The islands tend to grow in fairly uniformly spaced arrays (for reasons that are not yet understood), and under given growth conditions they have well-defined sizes and shapes. In Ge/Si, essentially four forms of islands are observed: shallow mounds (prepyramids), square pyramids with {105} facets, "hut clusters" — elongated pyramids with {105} facets — and large domes with facets in several orientations [8,9]. In the first stage of growth, shallow prepyramids appear that later convert T pyramids to pyramids. Large domes form for Ge coverages above five monolayers [10]. The island cross-sectional profiles are commonly triangular shaped and arc shaped, as have clearly been shown by transmission electron microscopy dark-field images by several researchers [8,11]. Island characteristic lengths are on the order of 10 nm.

For a restricted class of island profiles, namely, those for which the island surface profile has uniform curvature was considered by Freund et al. [12] and Tersoff et al. [13]. With this restriction, the island configuration can be characterized by just two parameters, the height-to-width aspect ratio and the volume. Here, equilibrium morphologies of epitaxial islands are calculated under the assumption that free energy of the system is composed of the elastic strain energy and the surface/interfacial energy. Particular attention is focused on the relaxation of the elastic strain in a coherent island as the configuration evolves in a way which lowers its total free energy, and equilibrium island morphologies corresponding to a given value of film material deposited on the substrate surface are also determined. The effect of a misfit dislocation on the equilibrium shape of the island is simply examined.

Let us consider a heteroepitaxial system consisting of an isolated island and a semi-infinite substrate. The geometry of the island has the regular pyramid with the square base edge length l and the height h, or the dome-shaped form with the circular base diameter $L$and the height H. The island and substrate are assumed to be elastically isotropic materials with the same values of the shear modulus μ and the same values of the Poisson's ratio ν, which is a reasonable approximation for a number of strained-layer systems of technology interest, such as Ge/Si and InAs/GaAs. Misfit stresses occur in the system due to the misfit (geometric mismatch) between the crystal lattice parameters a_i and a_s of the island and the substrate, respectively. For simplicity, here and in the following we confine our consideration to the two-dimensional plane strain

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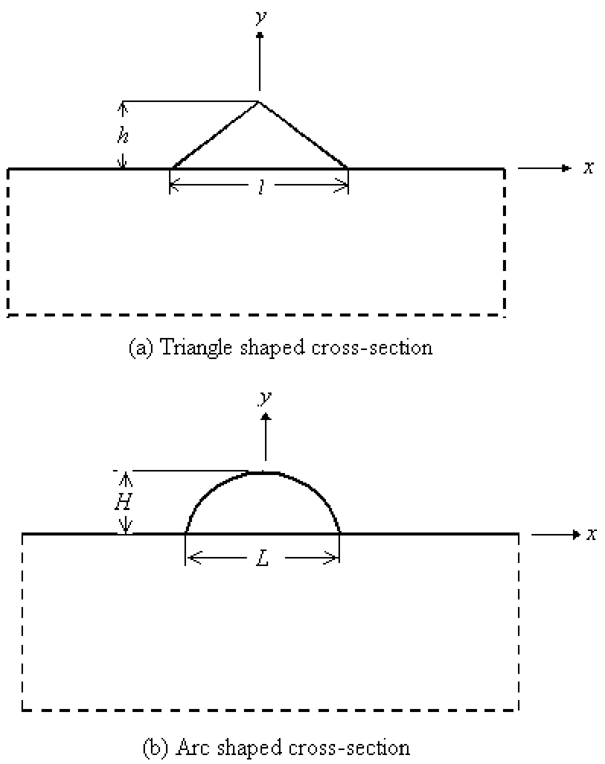

Fig. 1. Geometrical model of island/substrate system.

state model system with misfit strain $\varepsilon_0 = (a_s - a_i)/a_s$. The configurations are depicted in Figure 1. The shape of the island in the plane is taken to be a triangular shaped [14], or an arc shaped [8,11], which corresponds to the 'ridge' of the 3D pyramidal shape, or the 3D dome shape, respectively, but both the area of the island (volume per unit depth normal to the plane of Fig. 1) and the aspect ratio of the island are allowed to vary arbitrarily. The island we describe in two dimensions is equivalent to elongated island "ridge" in three dimensions [15]. If considered as a three-dimensional treatment of elongated island, we are assuming that there is no strain in the third dimension (along the island ridge). Including a misfit strain in this direction would modify only the length and energy scales without changing any qualitative features of the morphology [4], we expect all of the qualitative results here to carry over to the full three-dimensional case.

The total strains ε_{ij} of the system are the sum of the elastic strains ε_{ij}^e and the misfit strains $\varepsilon_{xx}^m = \varepsilon_0 H(y)$, $\varepsilon_{yy}^m = \varepsilon_{xy}^m = 0$ (in writing the formulas, it has been assumed that the film/substrate interface is perfectly sharp, i.e. there is no mixing of the film and substrate materials. Where $H(y)$ is Heaviside's unit step function with one in the island and zero in the substrate)

$$\varepsilon_{ij} = \varepsilon_{ij}^e + \varepsilon_{ij}^m. \tag{1}$$

Under the assumption of the plane strain state, total stresses σ_{ij} are related to the strains ε_{ij} by the constitutive equations

$$\sigma_{xx} = (\lambda + 2\mu)\varepsilon_{xx} + \lambda\varepsilon_{yy}, \tag{2}$$
$$\sigma_{yy} = (\lambda + 2\mu)\varepsilon_{yy} + \lambda\varepsilon_{xx}, \tag{3}$$
$$\sigma_{xy} = 2\mu\varepsilon_{xy}, \tag{4}$$

where λ, μ are Lame constants, $\lambda = 2\mu\nu/(1 - 2\nu)$. With equation (1) substituted into formulas (2)–(4), we get

$$\sigma_{ij} = \sigma_{ij}^e + \sigma_{ij}^m, \tag{5}$$

with the elastic stresses

$$\sigma_{xx}^e = (\lambda + 2\mu)\varepsilon_{xx}^e + \lambda\varepsilon_{yy}^e, \tag{6}$$
$$\sigma_{yy}^e = (\lambda + 2\mu)\varepsilon_{yy}^e + \lambda\varepsilon_{xx}^e, \tag{7}$$
$$\sigma_{xy}^e = 2\mu\varepsilon_{xy}^e, \tag{8}$$

and the misfit stresses

$$\sigma_{xx}^m = (\lambda + 2\mu)\varepsilon_0 H(y), \tag{9}$$
$$\sigma_{yy}^m = \lambda\varepsilon_0 H(y), \tag{10}$$
$$\sigma_{xy}^m = 0. \tag{11}$$

The equilibrium equations of elastic deformation of the system in the absence of body forces are

$$\frac{\partial \sigma_{ij}^e}{\partial x_j} = 0, \tag{12}$$

repeated indices are summed.

From equations (6), (9)–(12), the equilibrium equations can be written as [16]

$$\frac{\partial \sigma_{xx}}{\partial x} + \frac{\partial \sigma_{xy}}{\partial y} = 0, \frac{\partial \sigma_{xy}}{\partial x} + \frac{\partial \sigma_{yy}}{\partial y} - \lambda\varepsilon_0\delta(y) = 0, \tag{13}$$

with the boundary conditions $\sigma_{ij}n_j = 0$ expressing the fact that the surface of the system is stress free. Where σ_{ij} is stress tensor, $\boldsymbol{n} = (n_x, n_y)$ is the unit outward normal vector to the surface, $\delta(y)$ is Dirac's function.

From the mechanical effects of view, equations (13) indicate that the misfit strain is equivalent to applying a 'concentration body force' to the island/substrate interface in y-direction. The strain energy change of the system due to the formation of the island on the surface of the substrate can be expressed by

$$E^e = \frac{1}{2}\int_V -\lambda\varepsilon_0\delta(y)\, u_y(x, y)dV$$
$$= -\frac{1}{2}\lambda\varepsilon_0 \int_{-l(L)/2}^{l(L)/2} u_y(x, 0)dx, \tag{14}$$

e.g. the strain energy is equal to the work done by the external force (body force). Where V is the volume of the system, $u_y(x,y)$ the displacement of the interface in y-direction. It is energetically equivalent to the energy induced by applying stresses $\sigma_{yy}(x, 0) = -\lambda\varepsilon_0$,

$\sigma_{xy}(x, 0) = 0$ to the interface due to the island formation, while the body force is vanishing. Thus, the displacement of the interface induced by the interface stress can be obtained from the results of contact mechanics [17]

$$u_y(x,0) = \frac{1-\nu}{2\mu\pi}\lambda\varepsilon_0 \left[\left(x+\frac{l(L)}{2}\right)\ln\left(\frac{2x+l(L)}{l(L)}\right)^2 - \left(x-\frac{l(L)}{2}\right)\ln\left(\frac{2x-l(L)}{l(L)}\right)^2 - 2l(L)\ln 2\right], \quad -l(L)/2 < x < l(L)/2. \tag{15}$$

From equations (14) and (15), we obtain the strain energies for triangular shaped cross-sections and arc shaped cross-sections

$$E_1^e = \frac{2\mu\nu^2(1-\nu)}{(1-2\nu)^2\pi}\varepsilon_0^2\frac{A}{r},$$
$$E_2^e = \frac{\mu\nu^2(1-\nu)}{(1-2\nu)^2\pi}\varepsilon_0^2\frac{A}{f(R)}, \tag{16}$$

respectively. Where $f(R) = (\frac{1+4R^2}{8R})^2 \operatorname{arc\,cot}\frac{1-4R^2}{4R} - \frac{1-4R^2}{16R}$, A is the area of the island (volume per unit depth normal to the plane of Fig. 1), $r = h/l$ and $R = H/L$ are height-to-width aspect ratios of the two geometries, respectively.

The changes in the surface energy of the system due to formation of the island are

$$E_1^s = \sqrt{2A/r}(\sqrt{1+4r^2}\gamma_i - \gamma_s + \gamma_{is}) \tag{17}$$

and

$$E_2^s = \sqrt{A/f(R)}\left(\frac{1+4R^2}{4R}\operatorname{arc\,cot}\frac{1-4R^2}{4R}\cdot\gamma_i - \gamma_s + \gamma_{is}\right) \tag{18}$$

for triangular shaped and arc shaped cross-sections, respectively. Here γ_i and γ_s denote the surface energy densities of the island and the substrate materials, respectively, and γ_{is} denotes the energy density of the island/substrate interface, which are related to the contact angle α by the equilibrium Young's equation, $\gamma_i \cos\alpha = \gamma_s - \gamma_{is}$, where $\cos\alpha = (1+4r^2)^{-1/2}$ for triangular shaped and $\cos\alpha = (1-4R^2)/(1+4R^2)$ for arc shaped, respectively. It is noted that, the present assumptions including the form of the island and the validation of Young's equation is that the influence of the surface strain (stress) on the surface energies is neglected. N. Moll et al. [18] addressed the influence of surface stress on the equilibrium shapes of strained quantum dots using a hybrid approach that combines density functional theory calculations of microscopic parameters, surface energies, and surface stresses with elasticity theory for the long-range strain field and strain relaxation, their results fully confirm the reasonability of the present treatment, and that the quantitative differences between the present analysis and full physics are small.

From the formula (16), it is easily validated that the strain energy is a monotone decreasing function with the aspect ratio r, or R for a given volume of the island (i.e. a given value of material deposited on the surface of the substrate), in other words, the shapes of the island with a higher aspect ratio are strain-energetically favorable because that more and more of the elastic strain can be relaxed as the island becomes taller and narrower. This shows that the driving force for island formation is the reduction in the strain energy of a dislocation-free islanded morphology as compared to a flat film. On the other hand, with increasing the volume and the aspect ratio of the island, the total surface energy is increased with increasing area of the surface. The equilibrium shape of the island is therefore determined by the total free energy consisting of the strain energy and the surface energy.

The total free energies of the systems can be written as

$$E_1 = E_1^e + E_1^s = \frac{2\mu\nu^2(1-\nu)}{(1-2\nu)^2\pi}\varepsilon_0^2\frac{A}{r} + \sqrt{2A/r}[\sqrt{1+4r^2} - (1+4r^2)^{-1/2}]\gamma_i, \tag{19}$$

and

$$E_2 = E_2^e + E_2^s = \frac{\mu\nu^2(1-\nu)}{(1-2\nu)^2\pi}\varepsilon_0^2\frac{A}{f(R)} + \sqrt{A/f(R)}\left(\frac{1+4R^2}{4R}\operatorname{arc\,cot}\frac{1-4R^2}{4R} - \frac{1-4R^2}{1+4R^2}\right)\gamma_i, \tag{20}$$

for the two shapes, respectively.

The free energy is a balance of strain energy and surface energy terms, and a function of only the volume and the aspect ratio of the island for a given coherent island/substrate system. Equalizing its partial derivative with the aspect ratio to zero gives

$$\frac{\mu\nu^2(1-\nu)}{(1-2\nu)^2\pi}\varepsilon_0^2\sqrt{A/2} - \gamma_i\frac{3+4r^2}{(1+4r^2)^{3/2}}r^{5/2} = 0, \tag{21}$$

and

$$\frac{\mu\nu^2(1-\nu)}{(1-2\nu)^2\pi}\varepsilon_0^2\sqrt{A}\left(\frac{16R^4-1}{32R^3}\operatorname{arc\,cot}\frac{1-4R^2}{4R} + \frac{1+4R^2}{8R^2}\right) + \left[\frac{1}{2}f^{1/2}(R)\left(\frac{16R^4-1}{32R^3}\operatorname{arc\,cot}\frac{1-4R^2}{4R} + \frac{1+4R^2}{8R^2}\right)\cdot\left(\frac{1+4R^2}{4R}\operatorname{arc\,cot}\frac{1-4R^2}{4R} - \frac{1-4R^2}{1+4R^2}\right) + f^{3/2}(R)\left(\frac{1-4R^2}{4R^2}\operatorname{arc\,cot}\frac{1-4R^2}{4R} - \frac{1}{R} - \frac{16R}{(1+4R^2)^2}\right)\right]\gamma_i = 0, \tag{22}$$

for the two shapes, respectively. Which are functional relations between the aspect ratios and the areas of the islands when the epitaxial systems are at equilibrium state (minimum free energy) at fixed volume of the island.

For growth of Ge on Si (100), the system parameters have approximate values of $\varepsilon_0 = -0.04$, the Poisson's ratio $\nu = 0.25$, $\mu = 4\times10^{11}$ erg cm^{-3} and the surface energy

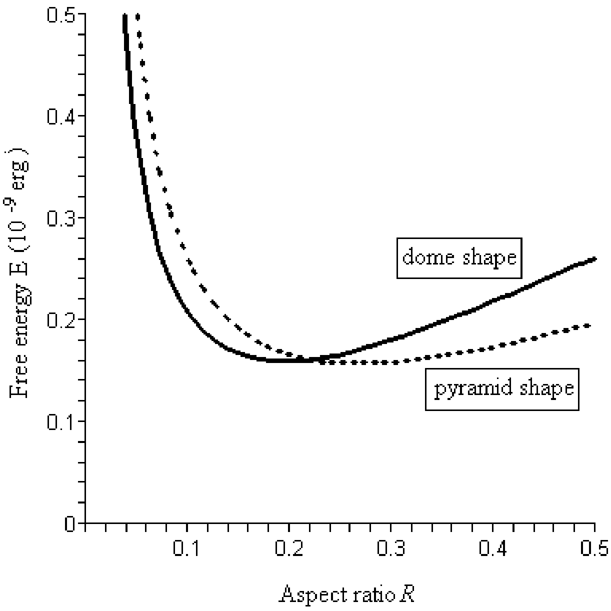

Fig. 2. Free energy E vs. island aspect ratio R for coherent island/substrate system with area of island $A = 160$ nm^2.

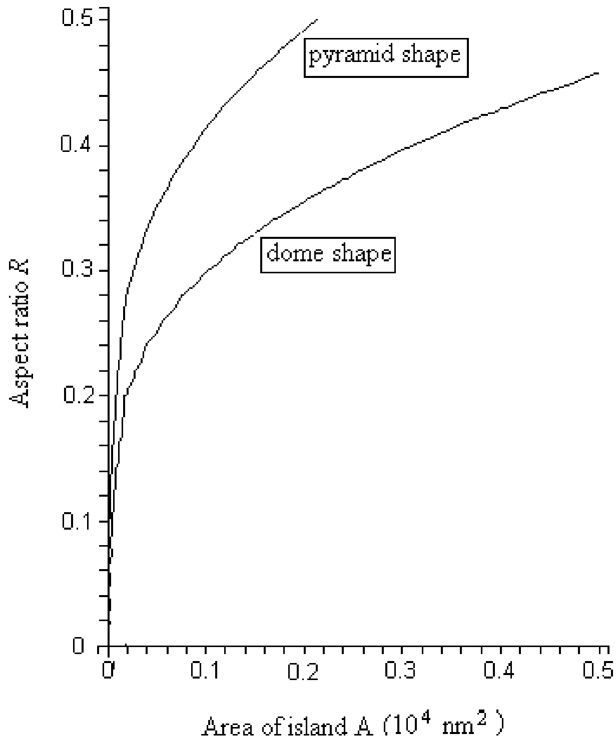

Fig. 3. Area of isaland A vs. island aspect ratio R for coherent island/substrate system with misfit $\varepsilon_0 = -0.04$.

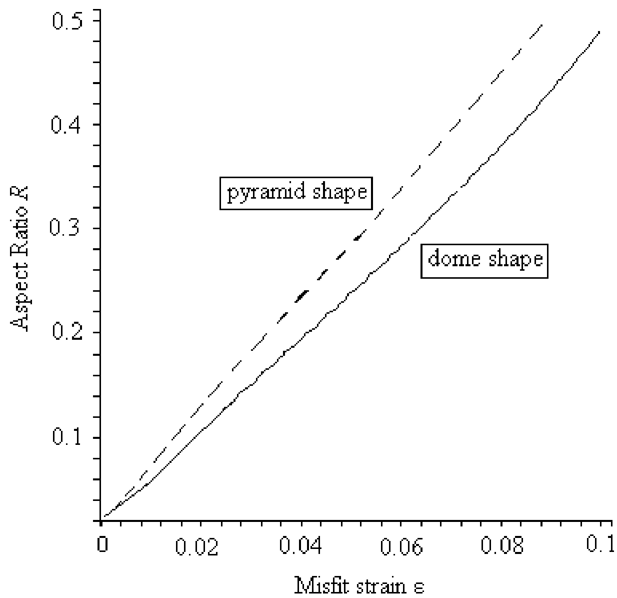

Fig. 4. The ε-R diagram for $A = 160$ nm^2.

density $\gamma_i = 2000$ erg cm^{-2} [19,20]. For the area of the island (volume per unit depth normal to the plane of Fig. 1) $A = 160$ nm^2 for which such size is a typicality observed experimentally [21], and equivalent to the equilibrium size of the island about 40 nm, the variations of the total free energy with aspect ratio is shown in Figure 2. The variations of the aspect ratio with area at fixed misfit, and with misfit at fixed volume are shown in Figures 3, 4, respectively. Several features relevant to coherent island growth are evident from equations (19)–(22) or Figures 2–4. First, the variation of the total free energy with aspect ratio shows that there is an equilibrium aspect ratio at which the free energy is a local minimum at fixed island volume (seen in Fig. 2). Second, That the equilibrium aspect ratio is increased for increasing island volume implies that islands become taller as they grow, consequently, for islands of small volume, surface energy effects are more important than elastic energy effects, the equilibrium aspect ratio is expected to be small. On the other hand, for island of larger volume, elastic effects are more important, the equilibrium aspect ratio is to be larger. These trends are illustrated quantitatively in Figure 3 and equations (21)–(22). Furthermore, that the aspect ratio is increased for increasing misfit strain between island and substrate implies that islands are taller for heteroepitaxial systems with larger misfit strain and same or similar mechanical properties at fixed islands volume (seen in Fig. 4). This, in turn, implies an aspect ratio of about 0.23 for triangular shaped cross-sections or 0.20 for arc shaped cross-sections, which are obtained from equations (21) and (22) and similar to what is observed experimentally [8].

An important problem connected with the growth of self-assembled islands is the change of their shape. Minimum free energies of the two geometries with the same volume are determined by equations (19) and (21), and equations (20) and (22), respectively. It follows that the free energy for triangular shaped is only slightly smaller than that for arc shaped when the area of the island is smaller than about 1600 nm^2, and the free energy for triangular shaped is larger than that for arc shaped when the area of the island is larger than about 1600 nm^2. It is shown in Figure 5. That is, in the first stage of growth, pyramids appear that later convert to domes form with increasing the volume of island, the critical area of island

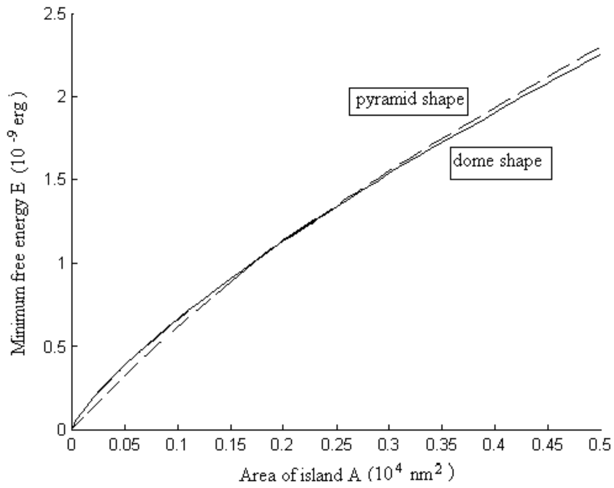

Fig. 5. Minimum free energy E vs. island area A for coherent island/substrate system with misfit $\varepsilon_0 = -0.04$.

for the conversion is about 1600 nm^2. Which is in good agreement with the experiments [8,9].

The explicit analytical forms such as equations (19)–(22), or Figures 2–4 are particularly valuable for identifying fundamental regimes of behavior for strained-layer growth. Besides the equilibrium shapes of coherent islands discussed here, they can be used to qualitatively investigate the equilibrium shapes of dislocated islands. Consider the introduction of a dislocation of Burgers displacement b parallel to the island/substrate interface. To estimate the effect of the dislocation, assume that the Burgers displacement is not confined at a single point, but is instead spread out along the interface. In this case the effective misfit strain is $\varepsilon_0^{eff} = \varepsilon_0 - b/l$ [22]. The equilibrium shape of the island is the same as that of an island with a misfit strain lower than ε_0 by an amount b/l. It is shown from the equations (19)–(22) and Figures 2–4 that a decrease in the misfit strain results in a decrease in the free energy and the island's aspect ratio, which is consistent with experiment observations [23]. Thus, a decrease in aspect ratio can be expected to follow the nucleation of a misfit dislocation in an island. For Burgers displacement $b = -0.4$ nm for which it is chosen to have the same sign as the misfit strain so that the dislocation acts to relax the elastic misfit strain, and $l(L) = 40$ nm, the equilibrium dislocated island has an aspect ratio of about 0.18 for triangular shaped, or 0.16 for arc shaped, which are obtained from the equations (21) and (22).

A two dimensional model consisting of an isolated island on a semi-infinite substrate with similar elastic properties is treated as a linearly elastic continuum, and is developed to study the mechanics of the island growth process in strained epitaxial systems. An analytical expression of free energy, which is a function of the island's volume, aspect ratio and film/substrate lattice misfit, as well as the evolving relations of aspect ratio against volume of the island and misfit of the system, which are new compared with the previous work, is achieved, and then used to model the growth process of a coherent island/substrate system. In a perfectly coherent system, for a given island volume, there is a particular island shape and height-to-width aspect ratio that result in a minimum system free energy, and the shape's transition occurs with increasing volume. That is, for a growing island, it is possible to predict the preferred equilibrium island shape and aspect ratio based on the minimization of the system free energy. Once a film/substrate system forms an interface dislocation, there is a new island height-to-width aspect ratio that minimizes the system free energy. The system reduces its free energy, low its height-to-width ratio.

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