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Morphologies of epitaxial islands on a lattice-misfitted substrate*

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Under certain growth conditions for systems with a film/substrate lattice misfit, the deposited material is known to aggregate into island-like shapes. We have obtained an analytical expression of the total free energy, which consists of strain energy, surface energy and interfacial energy of a coherent island/substrate system, and the change of equilibrium aspect ratio versus the volume of the island and the misfit of lattices in the system, which provides a broad perspective on island behaviour. These then were used to study the equilibrium shapes of the system. The results show that in order to minimize the total free energy, a coherent island will have a particular height-to-width aspect ratio, called equilibrium aspect ratio, that is a function of the island volume and misfit. The aspect ratio is increased with increasing island volume at a fixed misfit, and with increasing misfit strain between the island and substrate at a fixed island volume. Moreover, the effect of misfit dislocation on the equilibrium shape of the island is also examined. The results obtained are in good agreement with experiment of observations and thus can serve as a basis for interpreting the experiments.

Keywords: epitaxial island, free energy, equilibrium shape
PACC: 6220, 6150J, 6855, 8160C

1. Introduction

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 compressive misfit 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] When the film stays flat, growth occurs in a layer-by-layer (Frank–van der Merwe) fashion, which occurs in lattice-matched and closely lattice-mismatch systems. To map these complex behaviours, their phase diagram was obtained as a function of film height and misfit strain.[2]

Despite its complexity, the qualitative nature of the island formation is simple. When elastic strain energy is equal to the surface energy at some critical thickness,[8] the film cannot sustain a flat surface configuration, and undergoes a morphology transition by developing surface undulations, permitting strain relaxation to reduce the strain energy. This instability will then produce well-defined surface domains, i.e. 3D islands, as characterized by their nonzero local slopes or aspect ratio (height-to-width ratio). The islands tend to grow in fairly uniformly spaced arrays (for reasons that are not yet understood), and the islands themselves are usually dome-shaped, although cigar-shaped or elongated island have been observed in some higher temperature growth regimes.[9] The island cross-sectional profiles are commonly arc-shaped, as has been clearly shown by transmission electron microscopy and scanning electron microscopy dark-field images by several researchers.[10−12] Island characteristic lengths are on the order of 10nm.

For a restricted class of island profiles, namely, those for which the island surface profile has uniform curvature was discussed by Freund *et al* [13] and Ter-

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soff *et al.*[14] With this restriction, the island configuration can be characterized by just two parameters, the height-to-width aspect ratio and the volume. They showed that the free energy of an island of a certain volume assumes a minimum value for a certain value of aspect ratio, and that this value of aspect ratio depends on the island volume. In this article, equilibrium morphologies of epitaxial islands are calculated under the assumption that the free energy of the system is composed of elastic strain energy and surface/interfacial energy. An analytical expression of the free energy which is derived as a function of island's volume, aspect ratio and film/substrate lattice misfit; and the variation of aspect ratio with the volume of the island and misfit of the system is also obtained. These are new compared with the above mentioned work. Particular attention is focused on the relaxation of the elastic strain in a coherent island as the configuration evolves in a way to lower its total free energy, and the equilibrium island morphologies relevant 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.

2. Formulations

Let us consider a heteroepitaxial system consisting of an isolated nanoisland and a semi-infinite substrate. The geometry of the island has the dome-shaped form with the circular base diameter l and the height h. The nanoisland and substrate are assumed to be elastically isotropic materials with the same values of shear modulus μ and the same values of the Poisson ratio ν, which is a reasonable approximation for a number of strained-layer systems of technological 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 $a_{\rm i}$ and $a_{\rm s}$ of the island and the substrate, respectively. For simplicity, here and in the following we confine our consideration to the two-dimensional plane strain model system (i.e. $\varepsilon_{zz} = 0$, but the results can be extended to the case of a biaxial misfit) with uniaxial misfit strains $\varepsilon^{\rm m}_{xx} = \varepsilon_0 H(y)$, $\varepsilon^{\rm m}_{yy} = \varepsilon^{\rm m}_{xy} = 0$ (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 the misfit $\varepsilon_0 = (a_{\rm s} - a_{\rm i})/a_{\rm s}$, $H(y)$ is Heaviside's unit step function with unity in the island and zero in the substrate. The configuration is depicted in Fig.1. The shape of the island in the plane is taken to be an arc,[10−12] which corresponds to the 'ridge' of a 3D dome, 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 an elongated island "ridge" in three dimensions.[15] As a 3D 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] so we expect all of the qualitative results here to carry over to the full 3D case. The mechanical analysis is based on a linearly elastic continuum description of the system.

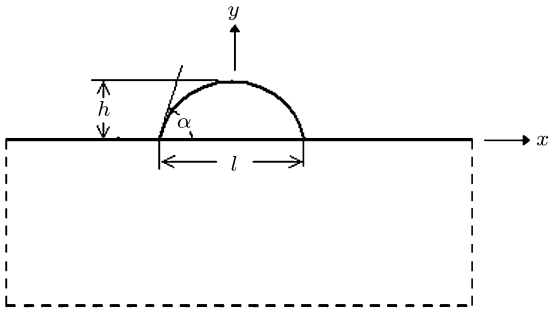


Fig.1. Geometrical model of island/substrate system.

The total strain ε_{ij} of the system is the sum of the elastic strain $\varepsilon^{\rm e}_{ij}$ and the misfit strain$\varepsilon^{\rm m}_{ij}$ $(i, j = x, y)$

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

The strain ε_{ij} is related to the displacements u_i by the relation

$$\varepsilon_{ij} = \frac{1}{2}\left(\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i}\right). \tag{2}$$

Under the assumption of plane strain state, the total stressσ_{ij} is related to the strain ε_{ij} by the constitutive equations

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

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

$$\sigma_{xy} = 2\mu\varepsilon_{xy}, \tag{5}$$

and the out-of-plane stress is $\sigma_{zz} = \nu(\sigma_{xx}+\sigma_{yy})$. Here λ, μ are Lame constants, $\lambda = 2\mu\nu/(1 - 2\nu)$. With Eq.(1) substituted into formulas (3)–(5), we have

$$\sigma_{ij} = \sigma^{e}_{ij} + \sigma^{\rm m}_{ij}, \tag{6}$$

with the elastic stresses

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

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

$$\sigma_{xy}^{\mathrm{e}} = 2\mu\varepsilon_{xy}^{\mathrm{e}}, \tag{9}$$

and the misfit stresses

$$\sigma_{xx}^{\mathrm{m}} = (\lambda + 2\mu)\varepsilon_0 H(y), \tag{10}$$

$$\sigma_{yy}^{\mathrm{m}} = \lambda\varepsilon_0 H(y), \tag{11}$$

$$\sigma_{xy}^{\mathrm{m}} = 0. \tag{12}$$

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

$$\frac{\partial\sigma_{ij}^{\mathrm{e}}}{\partial x_j} = 0. \tag{13}$$

Repeated indices denote summation.

The boundary conditions are the total stresses free on the surface of the system, i.e.

$$\sigma_{ij}n_j = 0, \tag{14}$$

where $\boldsymbol{n} = (n_x, n_y)$ is the unit outward vector normal to the surface. From Eqs.(6), (10)–(14), the equilibrium equations and the boundary conditions can be respectively written as

$$\frac{\partial\sigma_{xx}}{\partial x} + \frac{\partial\sigma_{xy}}{\partial y} = 0 \tag{15}$$

and

$$\frac{\partial\sigma_{xy}}{\partial x} + \frac{\partial\sigma_{yy}}{\partial y} - \lambda\varepsilon_0\delta(y) = 0, \tag{16}$$

where $\delta(y)$ is Dirac's function, and

$$\sigma_{xx}^{\mathrm{e}}n_x + \sigma_{xy}^{\mathrm{e}}n_y = -(\lambda + 2\mu)\varepsilon_0 H(y)n_x, \tag{17}$$

$$\sigma_{xy}^{\mathrm{e}}n_x + \sigma_{yy}^{\mathrm{e}}n_y = -\lambda\varepsilon_0 H(y)n_y. \tag{18}$$

From the viewpoint of mechanical effect, Eqs.(15) and (16) indicate that the misfit strain is equivalent to applying a 'concentration body force' to the island/substrate interface in y-direction, while Eqs.(17) and (18) indicate that the misfit strain is also equivalent to applying an elastic traction stress to the surface of the island. The stresses and strains can be determined by solving Eqs.(15) and (16) with boundary conditions (14), or solving Eqs.(13) with boundary conditions (17) and (18).

The total strain energy change of the system due to the formation of the island on the surface of the substrate can be expressed as[16]

$$\begin{aligned} E^{\mathrm{e}} &= \int_V -\lambda\varepsilon_0\delta(y)\, u_y(x,y)\mathrm{d}V \\ &= -\lambda\varepsilon_0 \int_{-l/2}^{l/2} u_y(x,0)\mathrm{d}x, \end{aligned} \tag{19}$$

where V is the volume of the system, e.g. the strain energy is equal to the work done by the external force (body force) under the stresses free conditions (14). 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 vanished. Thus, the displacements of the interface induced by the interface stresses are obtained from the results of contact mechanics[17]

$$u_x(x,0) = \frac{1-2\nu}{2\mu}\lambda\varepsilon_0 x, \tag{20}$$

$$\begin{aligned} u_y(x,0) =& \frac{1-\nu}{2\mu\pi}\lambda\varepsilon_0\left[\left(x+\frac{l}{2}\right)\ln\left(\frac{2x+l}{l}\right)^2\right. \\ &\left. -\left(x-\frac{l}{2}\right)\ln\left(\frac{2x-l}{l}\right)^2 - 2l\ln 2\right]. \end{aligned} \tag{21}$$

From Eq.(19), we obtain the strain energy

$$E^{\mathrm{e}} = \frac{2\mu\nu^2(1-\nu)}{(1-2\nu)^2\pi}\varepsilon_0^2\frac{A}{f(R)}, \tag{22}$$

where A is the area of the island (volume per unit depth normal to the plane of Fig.1) and $f(R)$ is a function of only the height-to-width aspect ratio $R = h/l$, described as

$$\begin{aligned} f(R) =& \left(\frac{1+4R^2}{8R}\right)^2 \operatorname{arc\,cot}\frac{1-4R^2}{4R} \\ &- \frac{1-4R^2}{16R}. \end{aligned} \tag{23}$$

While the strain distributions in the system cannot be obtained analytically (the strain distributions can be calculated by using numerical finite element method[18−20]); the analytical expression of the elastic strain energy for the system considered is achieved here.

The change in surface energy of the system due to the formation of an island is

$$\begin{aligned} E^{\mathrm{s}} =& l\left(\frac{1+4R^2}{4R}\operatorname{arc\,cot}\frac{1-4R^2}{4R}\cdot\gamma_{\mathrm{i}} - \gamma_{\mathrm{s}} + \gamma_{\mathrm{is}}\right) \\ =& \sqrt{A/f(R)}\left(\frac{1+4R^2}{4R}\operatorname{arc\,cot}\frac{1-4R^2}{4R}\right. \\ &\left.\cdot\gamma_{\mathrm{i}} - \gamma_{\mathrm{s}} + \gamma_{\mathrm{is}}\right), \end{aligned} \tag{24}$$

where γ_{i} and γ_{s} denote the surface energy densities of the island and the substrate material, 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,[21]

$$\begin{aligned} &\gamma_{\mathrm{i}}\cos\alpha = \gamma_{\mathrm{s}} - \gamma_{\mathrm{is}}, \\ &\cos\alpha = (1-4R^2)/(1+4R^2). \end{aligned} \tag{25}$$

3. Free energy and equilibrium shape

From formulae (22) and (23), it is easily shown that the strain energy is monotonically decreasing as a function of the aspect ratio 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 shape of the island with a higher aspect ratio is strain-energetically favourable because more and more fraction 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, when the volume and the aspect ratio of the island are increased, 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 energy of the system can be written as

$$E_{=}E^{\mathrm{e}}+E^{\mathrm{s}}=\frac{2\mu\nu^2(1-\nu)}{(1-2\nu)^2\pi}\varepsilon_0^2\frac{A}{f(R)}+\sqrt{A/f(R)}\left(\frac{1+4R^2}{4R}\operatorname{arc\,cot}\frac{1-4R^2}{4R}-\frac{1-4R^2}{1+4R^2}\right)\gamma_{\mathrm{i}}. \tag{26}$$

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

$$\begin{aligned}&\frac{2\mu\nu^2(1-\nu)}{(1-2\nu)^2\pi}\varepsilon_0^2\sqrt{A}\\&\times\left(\frac{16R^4-1}{32R^3}\operatorname{arc\,cot}\frac{1-4R^2}{4R}+\frac{1+4R^2}{8R^2}\right)\\&+\left[\frac{1}{2}f^{1/2}(R)\left(\frac{16R^4-1}{32R^3}\operatorname{arc\,cot}\frac{1-4R^2}{4R}+\frac{1+4R^2}{8R^2}\right)\right.\\&\times\left(\frac{1+4R^2}{4R}\operatorname{arc\,cot}\frac{1-4R^2}{4R}-\frac{1-4R^2}{1+4R^2}\right)\\&+f^{3/2}(R)\left(\frac{1-4R^2}{4R^2}\operatorname{arc\,cot}\frac{1-4R^2}{4R}-\frac{1}{R}\right.\\&\left.\left.-\frac{16R}{(1+4R^2)^2}\right)\right]\gamma_{\mathrm{i}}=0.\end{aligned} \tag{27}$$

This is a functional relation between the aspect ratio and the area of the island when the epitaxial system is in equilibrium state (minimum free energy) at a fixed volume of the island. For the growth of Ge on Si (100), the system parameters have approximate values of $\varepsilon_0=-0.04$, the Poisson's ratio $\nu=0.25$, $\mu=4\times10^{11}\mathrm{erg\,cm^{-3}}$ and the surface energy density $\gamma_{\mathrm{i}}=2000\mathrm{erg\ cm^{-2}}$.[18,22] The curves for Eqs.(26) and (27) are depicted in Figs.2–4.

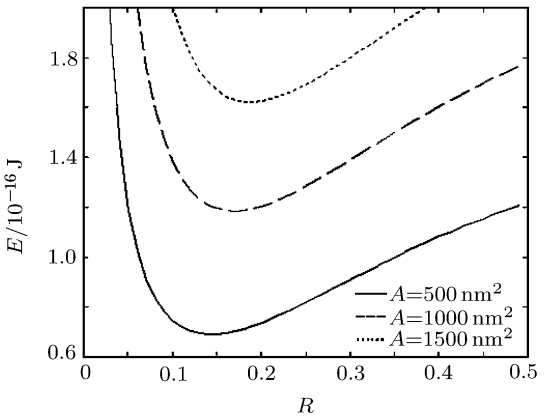


Fig.2. Free energy E vs island aspect ratio R for the coherent island/substrate system (Ge/Si).

Several features relevant to coherent island growth are evident from Eqs.(26) and (27). First, the variation of the total free energy with the aspect ratio shows that there is an equilibrium aspect ratio at which the free energy is a local minimum at a fixed island volume (see Fig.2). Second, that the minima occur at increasing values of aspect ratio for increasing island volume implies that the island becomes taller as it grows. Consequently, for an island with a small volume, surface energy effects are more important than elastic energy effects, hence the equilibrium aspect ratio is expected to be small. On the other hand, for an island with a larger volume, elastic effects are more important, hence the equilibrium aspect ratio will be larger. These trends are illustrated quantitatively in Figs.2 and 3 and Eq.(27). Furthermore, that the minima occur at increasing values of aspect ratio for increasing misfit strain between the island and substrate implies that the island is taller for a heteroepitaxial system with a larger misfit strain and the same or similar mechanical properties at fixed island volume (see Figs.3 and 4). For the area of the island (volume per unit depth normal to the plane of Fig.1) A=500nm^2. Such a size is a typical one observed experimentally[23] and equivalent to the base width of the equilibrium island being about 70nm, and height about 10nm. The variations of the total free energy with aspect ratio, and the aspect ratio with area and misfit of the island

are shown in Figs.2–4. This, in turn, implies an aspect ratio of about 0.14, which is similar to that observed experimentally.[10]

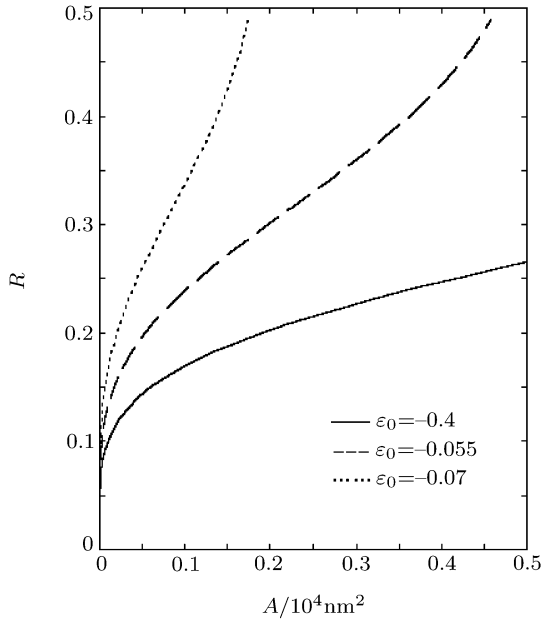


Fig.3. Island aspect ratio R vs area of island A for a coherent island/substrate system.

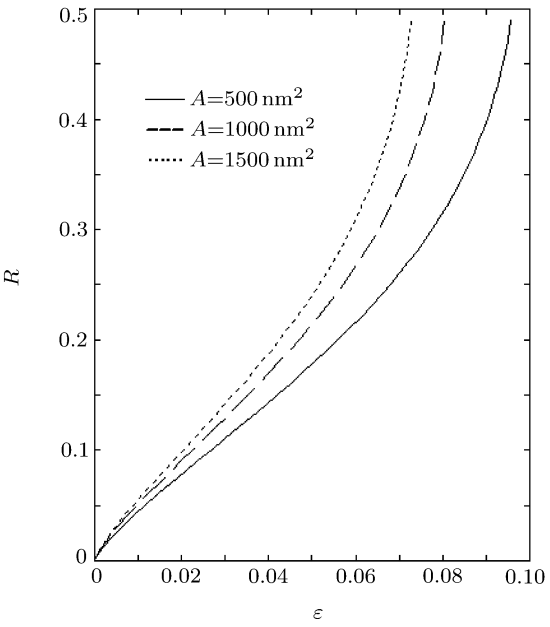


Fig.4. The $\varepsilon - R$ diagram.

The explicit analytical expressions, such as Eqs.(26) and (27), or diagrams shown in Figs.2–4 are particularly valuable for identifying the fundamental regimes of strained-layer growth. Besides the equilibrium shape of coherent island discussed here, they can be also used to qualitatively investigate the equilibrium shape of an island having dislocations. Consider the introduction of a dislocation of Burgers displacement b parallel to the island/substrate interface. To estimate the effect of the dislocation, we assume that the Burgers displacement is not confined at a single point, but instead spreads out along the interface. In this case the effective misfit strain is $\varepsilon_0^{\text{eff}} = \varepsilon_0 - b/l$.[24] The equilibrium shape of the island is the same as that of an island with a misfit strain lower than $\varepsilon_0$by an amount b/l. It is shown from Eqs.(26) and (27) and Figs.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 observations.[25] Thus, a decrease in aspect ratio can be expected to bring about the nucleation of a misfit dislocation in an island. For Burgers displacement b=–0.4nm, the sign of it is chosen to be the same as the misfit strain so that the dislocation acts to relax the elastic misfit strain, and for l=70nm, the equilibrium dislocated island has an aspect ratio of about 0.12.

4. Summary

A two-dimensional model has been developed to study the mechanism of the island growth process in strained epitaxial systems. A system consisting of an isolated arc-shaped island on a semi-infinite substrate with similar elastic properties is treated as a linear elastic continuum. An analytical expression of free energy, which consists of strain energy, surface energy and interfacial energy, is obtained and then used to model the growth process of a coherent island/substrate system. The island morphology has a lower free energy because it has a smaller strain energy; and the difference in strain energy between the islanded and flat films is large enough to offset the increasing surface energy. In a perfectly coherent system, for a given island volume, there is a particular island height-to-width aspect ratio that results in a minimum free energy of the system. That is, for a growing island, it is possible to predict the preferred equilibrium island aspect ratio based on the minimization of the free energy of the system. Once a film/substrate system forms an interface dislocation, there is a new island height-to-width aspect ratio that minimizes the free energy. The system reduces its free energy, and lowers its height-to-width ratio.

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