

Nano-islands on a composite substrate with misfit dislocations

I.A. Ovid'ko*, A.G. Sheinerman

Institute of Problems of Mechanical Engineering, Russian Academy of Sciences, Bolshoj 61, Vas. Ostrov, St. Petersburg 199 178, Russia

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Abstract. Spatial arrangements of nano-islands (quantum dots) on the free surface of a composite two-layer substrate containing misfit dislocations of edge type are theoretically examined. It is shown that the elastic interaction between misfit dislocations and nano-islands is capable of causing coagulation of nano-islands. The coagulation of nano-islands is shown to be favourable when the upper-layer thickness is smaller than a critical thickness, H_0 . An analytical form of H_0 is presented for the partial case with four-to-one correspondence between nano-islands and cells of the misfit dislocation network.

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The rapidly growing scientific and technological interest in nanostructured films and coatings has arisen from the unique properties associated with their nano-scale structure (see, e.g., [1–26]). In particular, the formation of spatially ordered ensembles of nano-islands on crystalline substrates is the subject of intensive theoretical and experimental study due to their high technological potential for device applications (see, e.g., [12–26]). Of special interest are applications of self-assembled semiconductor nano-islands (quantum dots) exhibiting unique optoelectronic properties for devices with reduced size and weight. In doing so, highly desired (from an applications viewpoint) functional characteristics of self-assembled semiconductor nano-islands crucially depend on their spatial arrangement and distributions in size and form. At present, in order to design and fabricate ordered ensembles of nano-islands, several technological parameters and methods of growth of nano-islands are used to influence their spatial position, size and form. Such parameters are, for instance, deposited material quantity, deposition rate, substrate temperature, chemical composition, and growth interruption time (see the reviews in [13, 14] and references therein). Recently, a very promising technological method for fabrication of spatially ordered ensembles of quantum dots has been

suggested and theoretically examined [25, 26]; it entails the formation of nano-islands (quantum dots) on free surfaces of substrates with internal interphase or low-angle grain boundaries. This method exploits the idea of controlling spatial organization of arrays of nano-islands by means of the elastic interaction between nano-islands and ordered networks of dislocations at the internal boundaries. Romanov et al. [25] and Bourret [26] examined the simplest situations with the one-to-one correspondence between nano-islands and cells of interfacial dislocation network. However, in many real situations the one-to-one correspondence is not realized; it is replaced by a general N -to- M correspondence, where N denotes the number of nano-islands that correspond to M cells of the interfacial dislocation net. In the general situation discussed, some new effects come into play which cannot be realized in the simplest situation with the one-to-one correspondence ($N = M = 1$) and which are interesting for technological applications. The main aim of this paper is to theoretically examine one of these effects occurring when $N > M$, namely the coagulation of nano-islands due to their elastic interaction with misfit dislocations located at an interfacial boundary in a composite (two-layer) substrate.

1 Nano-islands on a substrate with misfit dislocations (model)

Let us consider a model composite system consisting of a semi-infinite substrate 1 (phase 1), substrate 2 (phase 2) of finite thickness H , and nano-islands (phase 3) that grow on substrate 2 (Fig. 1). Substrates 1 and 2 as well as the nano-islands are assumed to be isotropic and characterized by the same shear moduli G and the same Poisson ratios ν . (All the formulae and conclusions of this paper are also valid in the situation where an additional wetting layer exists between substrate 2 and the nano-islands. In the situation discussed, thickness H plays the role of the total thickness of both substrate 2 and the additional wetting layer.) For definiteness, nano-islands are assumed to be regular pyramids with square bases having edges of length $2a$.

*Corresponding author.

(Fax: +7-812/3214771, E-mail: ovidko@def.ipme.ru)

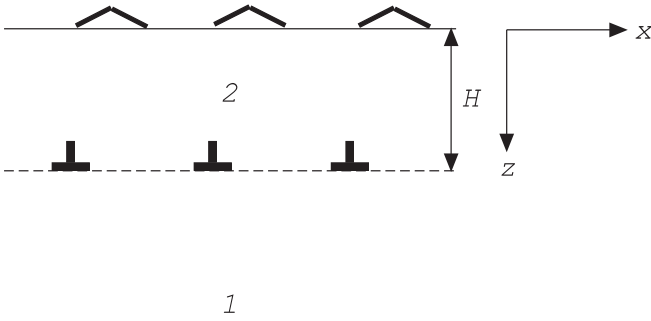


Fig. 1. Pyramid-like nano-islands on composite substrate (consisting of substrates 1 and 2) with misfit dislocations at internal interphase boundary

Substrate 2 and the nano-islands are elastically strained due to a misfit between the crystalline lattices of phases 1, 2 and 3. The misfit is assumed to be two-dimensional, in which case it is characterized by the parameters $f_i = (2a_1 - a_i)/(a_1 + a_i)$, where a_i is the crystal lattice parameter for the i th phase. If the thickness H of substrate 2 is smaller than some critical value H_c , the misfit between crystalline lattices of substrates 1 and 2 is accommodated completely by the elastic straining of substrate 2. For $H > H_c$, the formation of misfit dislocations (MDs) is energetically favourable, which effectively contribute to accommodation of the misfit (relaxation of the misfit stresses). For simplicity, in this paper we will focus on consideration of edge MDs located at the substrate-1–substrate-2 boundary and characterized by identical Burgers vectors \mathbf{b} that are parallel to the boundary plane. For an illustration of this, see Fig. 1, where MDs are shown which correspond to the misfit parameter $f_2 < 0$.

The elastic interaction between nano-islands and MDs causes nano-islands to have favorable spatial positions on the free surface of the composite substrate. In the particular situation with one-to-one correspondence between nano-islands and cells of the MD network ($N = M = 1$), arrays of nano-islands have the same spatial ordering as the MD network [26]. In the following sections, we will analyze the situation with the N -to- M correspondence between nano-islands and the MD network when $N > M$.

2 Nano-island-induced stresses in substrate

Now let us calculate the stresses created by a nano-island in the substrate, which will be used as input in a quantitative description of the elastic interaction between MDs and nano-islands (Fig. 1). Substrate 2 creates forces acting on the base of a nano-island that are distributed over the base and lead to either tension (if $f_3 > 0$) or compression (if $f_3 < 0$) of the nano-island. In turn, the nano-island creates forces acting on the substrate that lead to either compression (if $f_3 > 0$) or tension (if $f_3 < 0$) of the substrate region located under the nano-island and to either tension (if $f_3 > 0$) or compression (if $f_3 < 0$) of the rest of the substrate. In the following, for simplicity, we will confine our examination to the situation with symmetric strain of the nano-island, which comes into play when MDs and other nano-islands create axially symmetric strain fields in the substrate region under the nano-island considered. Such strain fields are created, for instance, by two orthogonal dislocations equidistant from the nano-

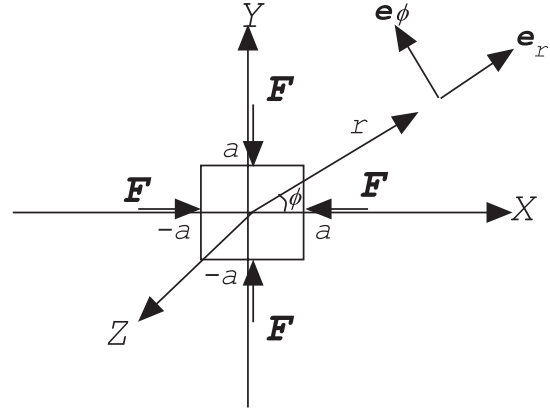


Fig. 2. Quadrupole of nano-island-induced forces \mathbf{F} acting on substrate

island or an ensemble of dislocation pairs, consisting of two orthogonal dislocation rows. In the situation discussed, we will model the nano-island-induced forces acting on substrate 2 by a quadrupole of concentrated forces. For an illustration of this, see Fig. 2 which corresponds to $f_3 > 0$.

Supposing that the side length $2a$ of the nano-island base is smaller than the thickness H of substrate 2, we can treat substrate 2 as semi-infinite in calculations of the displacement and stress fields created by the nano-island in substrate 2. In these circumstances, from general formulae for displacements induced in a semi-infinite medium by mechanical loads q_X , q_Y and q_Z distributed over a region of the plane that bounds the medium [27], we find the displacements induced by the quadrupole of the forces \mathbf{F} (Fig. 2) in substrates 1 and 2. In doing so, when the normal mechanical load q_Z is absent, we find the following formulae for the displacements, u_X , u_Y and u_Z , in the coordinate system (X, Y, Z) shown in Fig. 2:

$$u_X = \frac{1}{2\pi G} \left[\varphi_X - \frac{1}{2} Z \frac{\partial}{\partial X} \left(\frac{\partial \psi_X}{\partial X} + \frac{\partial \psi_Y}{\partial Y} \right) + \nu \frac{\partial}{\partial X} \left(\frac{\partial \chi_X}{\partial X} + \frac{\partial \chi_Y}{\partial Y} \right) \right], \quad (1)$$

$$u_Y = \frac{1}{2\pi G} \left[\varphi_Y - \frac{1}{2} Z \frac{\partial}{\partial Y} \left(\frac{\partial \psi_X}{\partial X} + \frac{\partial \psi_Y}{\partial Y} \right) + \nu \frac{\partial}{\partial Y} \left(\frac{\partial \chi_X}{\partial X} + \frac{\partial \chi_Y}{\partial Y} \right) \right], \quad (2)$$

$$u_Z = \frac{1}{2\pi G} \left[\frac{1}{2} (1-2\nu) \left(\frac{\partial \psi_X}{\partial X} + \frac{\partial \psi_Y}{\partial Y} \right) - \frac{1}{2} Z \left(\frac{\partial \varphi_X}{\partial X} + \frac{\partial \varphi_Y}{\partial Y} \right) \right], \quad (3)$$

where

$$\varphi_i(X, Y, Z) = \iint_{\vartheta} \frac{q_i(X', Y')}{R'} dX' dY', \quad (4)$$

$$\psi_i(X, Y, Z) = \iint_{\vartheta} q_i(X', Y') \ln(R' + Z) dX' dY', \quad (5)$$

$$\chi_i(X, Y, Z) = \iint_{\vartheta} q_i(X', Y') [Z \ln(R' + Z) - R'] dX' dY'. \quad (6)$$

Here ϑ is the integration region; $i = X, Y$; $R'^2 = (X - X')^2 + (Y - Y')^2 + Z^2$.

For the quadrupole of the forces F acting at the points $(X' = -a, Y' = 0)$, $(X' = a, Y' = 0)$, $(X' = 0, Y' = -a)$ and $(X' = 0, Y' = a)$, we have

$$q_X = F\delta(X' + a)\delta(Y') - \delta(X' - a)\delta(Y'), \quad (7)$$

$$q_Y = F\delta(X')\delta(Y' + a) - \delta(X')\delta(Y' - a), \quad (8)$$

where $\delta(t)$ denotes the Dirac delta function. From these formulae it follows that potentials φ_i , ψ_i and χ_i behave as follows:

$$\varphi_X = -\frac{2aX}{R^3}F, \quad \varphi_Y = -\frac{2aY}{R^3}F, \quad (9)$$

$$\psi_X = \frac{2aX}{R(R+Z)}F, \quad \psi_Y = \frac{2aY}{R(R+Z)}F, \quad (10)$$

$$\chi_X = -\frac{2aX}{R+Z}F, \quad \chi_Y = -\frac{2aY}{R+Z}F, \quad (11)$$

for $R \gg a$, where $R^2 = X^2 + Y^2 + Z^2$.

With (9)–(11), (1)–(3) can be presented as follows:

$$u_X = \frac{aF}{2\pi G} \frac{X}{R^3} \left(-2(1-\nu) + 3\frac{Z^2}{R^2} \right), \quad (12)$$

$$u_Y = \frac{aF}{2\pi G} \frac{Y}{R^3} \left(-2(1-\nu) + 3\frac{Z^2}{R^2} \right), \quad (13)$$

$$u_Z = \frac{aF}{2\pi G} \frac{Z}{R^3} \left(-2\nu + 3\frac{Z^2}{R^2} \right). \quad (14)$$

Then the displacements u_r and u_φ in the cylindrical coordinates (see Fig. 2) can be written, using (12) and (13), as follows:

$$u_r = \frac{aF}{2\pi G} \frac{r}{R^3} \left(-2(1-\nu) + 3\frac{Z^2}{R^2} \right), \quad (15)$$

$$u_\varphi = 0, \quad (16)$$

where $r^2 = X^2 + Y^2$. From (14)–(16) we find the components of the tensor ε^{isl} of strain created by the nano-island in substrate 2:

$$\varepsilon_{rr}^{\text{isl}} = \frac{\partial u_r}{\partial r} = \frac{aF}{2\pi G} \frac{1}{R^3} \left(4(1-\nu) - 6(3-\nu)\frac{Z^2}{R^2} + 15\frac{Z^4}{R^4} \right), \quad (17)$$

$$\varepsilon_{\varphi\varphi}^{\text{isl}} = \frac{u_r}{r} = \frac{aF}{2\pi G} \frac{1}{R^3} \left(-2(1-\nu) + 3\frac{Z^2}{R^2} \right), \quad (18)$$

$$\varepsilon_{ZZ}^{\text{isl}} = \frac{\partial u_Z}{\partial Z} = \frac{aF}{2\pi G} \frac{1}{R^3} \left(-2\nu + 3(1+2\nu)\frac{Z^2}{R^2} - 15\frac{Z^4}{R^4} \right), \quad (19)$$

$$\varepsilon_{rZ}^{\text{isl}} = \frac{1}{2} \left(\frac{\partial u_r}{\partial Z} + \frac{\partial u_Z}{\partial r} \right) = \frac{3aF}{2\pi G} \frac{rZ}{R^5} \left(2 - 5\frac{Z^2}{R^2} \right), \quad (20)$$

$$\varepsilon_{\varphi Z}^{\text{isl}} = \varepsilon_{\varphi Z}^{\text{isl}} = 0. \quad (21)$$

Thus, we have obtained expressions for the strain fields created by nano-islands in the substrate. These expressions will be used in the following sections for a consideration of the elastic interaction of nano-islands with MDs as well as the island–island interaction.

3 Coagulation of nano-islands due to their elastic interaction with misfit dislocations

Let us consider an ensemble of nano-islands on a composite (two-layer) substrate with MDs in the situation when $N > M$ (Fig. 3). In the situation discussed, the elastic interaction between nano-islands (creating stress fields which drop as R^{-3} , where R is the distance from a nano-island) should be taken into account, if several nano-islands are attracted to one point on the free surface of a composite substrate, and, as a corollary, the distance between them becomes comparatively small. In these circumstances, stable equilibrium positions of nano-islands are derived from the balance between attractive forces acting owing to elastic interaction of nano-islands and MDs and repulsive forces acting due to island–island interaction. If the attractive forces exceed the repulsive ones, the nano-islands come in contact with each other and then coagulate. This scenario of evolution of nano-islands comes into play when the thickness H of substrate 2 is smaller than some critical value H_0 , because the repulsive forces discussed increase with a decrease in H .

A detailed general analysis of island–island and MD–island interactions is beyond the scope of this paper. In the rest of this section, for definiteness, we will focus on a theoretical examination of a partial case, namely the case with a group consisting of four equal-size nano-islands on a composite substrate. This partial case is interesting as a good model of many real situations with arrays of nano-islands and can serve as a basis for the general analysis of behavior exhibited by groups of nano-islands.

Let us estimate both equilibrium distance $2l$ between nano-islands and critical thickness H_0 (below which the nano-islands coagulate) in the case with four nano-islands. Let the nano-islands be attracted to the point $x_0 = 0$ located above an intersection node of two orthogonal MDs due to MD–island interactions¹ (Fig. 3). Under these circumstances, in our first-approximation calculations of the energies characterizing island–island and MD–island interactions, we will take into consideration only the constituents related to interaction-induced changes of the elastic energy

¹ This situation is realized when $f_2 f_3 < 0$

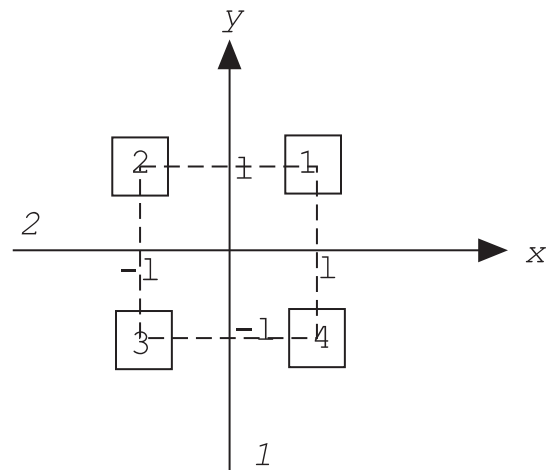


Fig. 3. Four nano-islands are attracted to a node of two orthogonal misfit dislocations due to elastic interaction

of nano-islands, while the changes of the elastic energy of the substrate will be neglected. With the short-range character of island–island interactions taken into account, we will describe them with the help of the “approximation of nearest neighbors”. For example, in calculations of the interaction energy characterizing nano-island 1, shown in Fig. 3, only its interactions with nano-islands 2 and 4 are considered.

In order to estimate the equilibrium distance $2l$ between the nano-islands, we will calculate the energy of interaction of nano-island 1 with nano-islands 2 and 4 and with MDs. To do so, first, let us calculate strains $\boldsymbol{\varepsilon}^{(2)}$ and $\boldsymbol{\varepsilon}^{(4)}$, created by nano-islands 2 and 4, respectively, in the substrate 2 fragment under nano-island 1. In our approximate calculations, strains $\boldsymbol{\varepsilon}^{(2)}$ and $\boldsymbol{\varepsilon}^{(4)}$ are assumed to be spatially homogeneous and equal to values of corresponding real (spatially inhomogeneous) strains in the center of the nano-island 1 base. In these circumstances, strains $\boldsymbol{\varepsilon}^{(2)}$ and $\boldsymbol{\varepsilon}^{(4)}$ are calculated with the help of (17) and (18) as follows:

$$\varepsilon_{xx}^{(2)} = \varepsilon_{yy}^{(4)} = \varepsilon_{rr}^{\text{isl}}(r = 2l, Z = 0) = \frac{aF(1-\nu)}{4\pi Gl^3}, \quad (22)$$

$$\varepsilon_{yy}^{(2)} = \varepsilon_{xx}^{(4)} = \varepsilon_{\varphi\varphi}^{\text{isl}}(r = 2l, Z = 0) = -\frac{aF(1-\nu)}{8\pi Gl^3}. \quad (23)$$

The total strains, $\varepsilon_{xx}^{\text{isl}} = \varepsilon_{xx}^{(2)} + \varepsilon_{xx}^{(4)}$ and $\varepsilon_{yy}^{\text{isl}} = \varepsilon_{yy}^{(2)} + \varepsilon_{yy}^{(4)}$, created by nano-islands 2 and 4 are identical and can be derived from the following formula:

$$\varepsilon_{xx}^{\text{isl}} = \varepsilon_{yy}^{\text{isl}} = \varepsilon^{\text{isl}} = \frac{aF(1-\nu)}{8\pi Gl^3}. \quad (24)$$

Now let us calculate the strains created by dislocations 1 and 2 under nano-island 1. In doing so, for simplicity, we assume that the characteristic size a of the nano-island base is essentially smaller than the thickness H of substrate 2: $a \ll H$. This assumption allows us to neglect spatial inhomogeneities of dislocation-induced strain on the interphase boundary between the nano-island base and substrate 2. That is, the strain in question is effectively treated as if it were independent of the spatial coordinates of the interphase boundary.

The stress fields $\boldsymbol{\sigma}^{\text{d1}}$ and $\boldsymbol{\sigma}^{\text{d2}}$ created by dislocations 1 and 2 can be derived from the stress function [28] for a dislocation near a free surface. From the symmetry of the system considered it follows that the nano-island is equidistant from dislocations 1 and 2. Therefore, the component $\sigma_{yy}^{\text{d2}}(l)$ of the stress field $\boldsymbol{\sigma}^{\text{d2}}$ in the region under the nano-island (having a center at point $x = y = l$) is equal to the component $\sigma_{xx}^{\text{d1}}(l)$ of the stress fields $\boldsymbol{\sigma}^{\text{d1}}$ in this region: $\sigma_{yy}^{\text{d2}}(l) = \sigma_{xx}^{\text{d1}}(l)$. From this equation, Hooke’s law [29], and the expressions for σ_{xx}^{d1} and σ_{yy}^{d2} (obtained as stated above), we find components of the total strain field $\boldsymbol{\varepsilon}^{\text{d}}$ created by the pair of dislocations in the region under the nano-island (Fig. 3):

$$\varepsilon_{xx}^{\text{d}}(l) = \varepsilon_{yy}^{\text{d}}(l) = \varepsilon^{\text{d}}(l) = \frac{8b_x H(1+\nu)}{\pi} \frac{l^2}{(l^2 + H^2)^2}, \quad (25)$$

where $\varepsilon^{\text{d}} = \varepsilon_{xx}^{\text{d}} = \varepsilon_{yy}^{\text{d}}$.

The strain energy ΔW^{isl} of the interaction of nano-island 1 with dislocations (1 and 2) and nano-islands (2 to 4)

is defined as the difference between the nano-island 1 energy W^{isl} in the presence of dislocations and other nano-islands and its energy W_0^{isl} in the absence of the dislocations and other nano-islands: $\Delta W^{\text{isl}} = W^{\text{isl}} - W_0^{\text{isl}}$. The energy W_0^{isl} can be written as

$$W_0^{\text{isl}} = 2G \frac{1+\nu}{1-\nu} \alpha f_3^2 V, \quad (26)$$

where $V (= \frac{4}{3}a^2 \tan \theta)$ denotes the volume of the nano-island, θ the tilt angle of lateral facets of the pyramid-like nano-island, and α the factor taking into account both a decrease in the mean strains of the nano-island due to the effects of strains of the substrate fragment under the nano-island and relaxation of stresses and strains due to the bending of the island. [It should be noted that (26) does not take into account the difference between the equilibrium interatomic distances on the free surface of the nano-island and in its bulk [14] that causes an additional bending of the nano-island. However, for the aim of this paper, dealing with the first-approximation examination of the effect of MDs on nano-islands, the difference in question can be neglected.]

The energy W^{isl} is obtained from (26) by the replacement of f_3 with $f_3 + \varepsilon^{\text{d}}(l) + \varepsilon^{\text{isl}}$. Hence the interaction energy ΔW^{isl} is

$$\Delta W^{\text{isl}} = 2G \frac{1+\nu}{1-\nu} \alpha \left\{ [f_3 + \varepsilon^{\text{d}}(l) + \varepsilon^{\text{isl}}]^2 - f_3^2 \right\} V. \quad (27)$$

In order to calculate the force F appearing in (24), we, following [30], represent the stresses acting in the nano-island as the sum of the stresses $\boldsymbol{\sigma}^0$ and $\boldsymbol{\sigma}'$. Here $\boldsymbol{\sigma}^0$ and $\boldsymbol{\sigma}'$ are respectively the stresses that would exist in a thin film and the stresses that are related to the relaxation of stresses in the nano-island, providing the validity of the condition $\sigma_{nn}^0 + \sigma'_{nn} = \sigma_{n\tau}^0 + \sigma'_{n\tau} = 0$ on the lateral facets of the nano-island, where \mathbf{n} is a normal to a lateral facet and the axis τ is located in the lateral facet plane (Fig. 4). The stresses $\boldsymbol{\sigma}'$ are

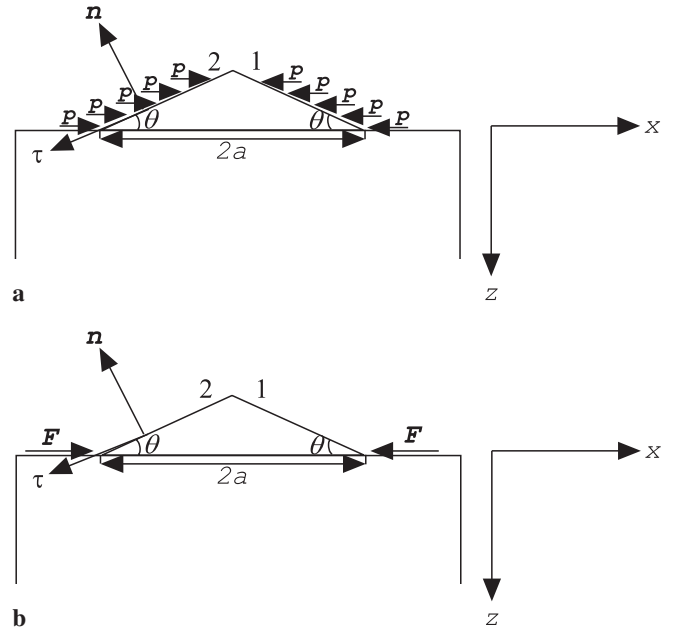


Fig. 4a,b. Nano-island-induced forces acting on substrate. **a** Forces p distributed over lateral facets of nano-island. **b** Concentrated forces F equivalent to forces p

equivalent to the image forces \mathbf{p} (Fig. 4a) distributed over the lateral facets of the nano-island, with the modulus being $p = \sigma_0 \sin \theta$, where

$$\sigma_0 \approx 2G \frac{1+\nu}{1-\nu} [f_3 + \varepsilon^d(l) + \varepsilon^{\text{isl}}]. \quad (28)$$

For our purposes, the forces \mathbf{p} distributed over the lateral facets can effectively be replaced by concentrated forces \mathbf{F} applied to the centers of the edges of the nano-island base (Fig. 4b) and characterized by modulus $F \approx S_{\text{lat}} p$, where $S_{\text{lat}} = a^2 / \cos \theta$ is the area of the lateral facet. As a corollary, the formula for the force F can be written as follows:

$$F = 2G \frac{1+\nu}{1-\nu} \beta a^2 \tan \theta [f_3 + \varepsilon^d(l) + \varepsilon^{\text{isl}}], \quad (29)$$

where $\beta \approx 1$.

By substituting (29) into (24), we have an equation for ε^{isl} , whose solution is given as

$$\varepsilon^{\text{isl}} = \frac{K}{1-K} [f_3 + \varepsilon^d(l)] \approx K [f_3 + \varepsilon^d(l)]. \quad (30)$$

Here $K = (\beta a^3 \tan \theta) / (4\pi l^3) \ll 1$.

By substituting (25) and (30) into (27) and by taking inequalities $l/H \ll 1$ and $K \ll 1$ into consideration, we find the solution of the equation (which characterizes the equilibrium of nano-island 1)

$$\partial \Delta W^{\text{isl}} / \partial l = 0 \quad (31)$$

to be as follows:

$$l^5 = \frac{3\beta a^3 \tan \theta f_3}{64b(1+\nu)} H^3. \quad (32)$$

From (32) and the condition $l = a$ (which corresponds to the coagulation of nano-islands), we find the maximum thickness H_0 of substrate 2, at which nano-islands coagulate, to be given as

$$H_0 = \left(\frac{64b(1+\nu)}{3\beta \tan \theta f_3} a^2 \right)^{1/3}. \quad (33)$$

According to (33), H_0 decreases with a decrease in the characteristic size of nano-islands and/or increase in the misfit parameter f_3 . If substrate 1 and the nano-islands are identical crystalline phases, we have $f_3 = 0$ and $H_0 \rightarrow \infty$, in which case coagulation of nano-islands is energetically favorable for any value of the thickness H of substrate 2.

4 Concluding remarks

In this paper we have theoretically described coagulation of nano-islands owing to their elastic interaction with MDs located at the internal interphase boundary in a composite (two-layer) substrate (Fig. 1). It has been shown that, for a group of nano-islands nucleating on the substrate surface above a node of the MD network, the stable equilibrium positions of islands are determined by a balance between attractive

forces, induced by interaction of islands and MDs, and repulsive forces, resulting from the island–island interaction. As a corollary, nano-islands can either converge or form stable configurations, each consisting of several nano-islands, in the vicinity of the points to which the islands are attracted by MDs. This phenomenon can potentially be used for fabrication of ordered arrays of quantum dots with desired spatial arrangements and distributions in size and form.

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