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The Asaro–Tiller–Grinfeld instability revisited

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Abstract

Deposition of a thin film on a solid substrate in the presence of a misfit leads to a growth instability that favors three-dimensional (3D) morphology of the free surface. The amount of the misfit and the conditions of the film deposition (molecular beam epitaxy) lead to an elastic problem, where surface energy has the same order of magnitude as the bulk energy. The instability occurs at a critical thickness of the film. The value of the critical thickness is shown to be given by the competition between the bulk and surface effects. We investigate (via a Fourier method) the Asaro–Tiller–Grinfeld instability for cubic materials and in the presence of an arbitrary misfit. We solve the problem in the general case and we specialize our results to recover values which are in good agreement with experimental data in the case of a $In_{1-x}Ga_xAs$ alloy. We consider in a 3D framework sinusoidal perturbations of the free boundary at arbitrary orientations with respect to crystallographic axes. Thus, we are able to minimize the sum of the bulk and surface energies with respect to the orientation and therefore to predict qualitative aspects of the surface morphology.

1. Introduction

For practical purposes growth of crystals is a common process today. However, the morphology of the free boundary, which is desired planar for most applications, is distorted by different types of *instabilities*. A self-contained review of this kind of phenomena can be found in Politi et al. (2000).

We shall focus in this paper on the analysis of one type of growth instability which is observed in a process called molecular beam epitaxy (MBE), and which is known as the Asaro–Grinfeld instability. The MBE is a process of deposition of an elastic layer on an elastic substrate. Since both crystals are anisotropic and the lattice parameters are slightly different from one material to another, there is a *misfit* at the interface between the layer and the substrate. The amount of this misfit may differ from one material to another, but typically the range of it is between 2% and 5%. The deposition process takes place in a chamber where the pressure is controlled to remain around 10^{-7} Pa. Thus, from a mechanical point of view the free boundary of the layer may be considered as being traction-free.

In practical applications the misfit between the layer and the substrate depends on the composition of both materials. We shall apply our results to a typical case which is the deposition of a layer of $In_{1-x}Ga_xAs$

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alloy on a InP alloy. It is known that both materials have cubic structure and the value of the misfit can be computed using the lattice parameters. The computation shows that the layer supports a compressive strain of 2% in the crystallographic directions.

Experiments show that the growth process evidences a transition between a two-dimensional (2D) morphology (i.e., a planar free surface), to a 3D morphology (i.e., a wavy surface) at a critical thickness, subsequently denoted h_c. Roughly speaking this process can be explained by the competition between the bulk and surface energy. A wavy boundary decreases the bulk energy (by stress relaxation) but increases the surface energy.

The paper is organized as follows: Section 2 presents several elastic problems necessary to model the elasticity of the layer, and establish some general estimates about the gap between the bulk energy in a planar configuration and a distorted one (a wavy boundary). As the estimates from this section are only qualitative, we develop in Section 3, for a small amplitude perturbation of the boundary, a quantitative estimate for the bulk energy. We develop the general solution of the *first order problem* in the general context, and then specialize it to the case of a *long wave limit* in Section 4. The result provides explicit formulas for the orthotropic case that we may further specialize to the cubic and isotropic case in the Section 4. In Section 5 we introduce the surface energy (considered here as being proportional to the free surface of the layer) and consider the competition between the bulk and surface energy. We give an explicit formula for the critical thickness and we specialize it for $In_{1-x}Ga_{1-x}As$ alloy at $x = 0.18$. Finally, we compare this estimate with experimental results and find a good agreement with experimental data.

The idea of the elastic relaxation goes back to Asaro and Tiller (1972), and was recently reconsidered by Grinfeld (1993). We point out in the following the main new features of our study: we deal with *orthotropic materials* and not only with isotropic materials as it was done in Asaro et al. (1972), Grinfeld (1993). We consider wavy boundaries which are not necessarily oriented according to the crystallographic directions, and consequently we are able to predict the direction of the wavy boundary with respect to crystallographic direction. We derive the general solution of the first order problem in the above mentioned context and we recover previous results obtained by Grinfeld (1986) in the isotropic case.

Several comments about the limits of our method and of its possible extensions are made in the final section. An interesting point is the fact that what can be obtained here with a constant surface energy density cannot be obtained for an orientational dependent surface energy density, because in that case *the force system at the surface* changes, i.e. the surface shear does not vanish.

2. Estimates concerning the bulk energy

2.1. Preliminaries

Consider an open bounded subset $\Sigma \subset \mathbb{R}^2$, and let $h > 0$. Denote $\Omega_0 = \Sigma \times (0, h)$, $\Sigma_h = \Sigma \times \{h\}$, $\Sigma_0 = \Sigma \times \{0\}$, $S_h = \partial\Sigma \times [0, h]$, thus $\partial\Omega_0 = \Sigma_h \cup \Sigma_0 \cup S_h$. Consider a smooth function Φ: $\Sigma \to \mathbb{R}$, such that $\Phi > -h$ and $\int_\Sigma \Phi \, d\Sigma = 0$. Denote further by

$$\Omega_\Phi = \{\mathbf{x} \in \mathbb{R}^3,\ (x_1, x_2) \in \Sigma,\ 0 < x_3 < h + \Phi(x_1, x_2)\},$$

$$\Sigma_{h+\Phi} = \{\mathbf{x} \in \mathbb{R}^3,\ (x_1, x_2) \in \Sigma,\ x_3 = h + \Phi(x_1, x_2)\},$$

$$S_{h+\Phi} = \{\mathbf{x} \in \mathbb{R}^3,\ (x_1, x_2) \in \partial\Sigma,\ 0 \leqslant x_3 \leqslant h + \Phi(x_1, x_2)\},$$

thus, $\partial\Omega_\Phi = \Sigma_{h+\Phi} \cup \Sigma_0 \cup S_{h+\Phi}$. Obviously, for $\Phi = 0$ we have $\Omega_\Phi = \Omega_0$, $\Sigma_{h+\Phi} = \Sigma_h$, and $S_{h+\Phi} = S_h$. Moreover, if μ denotes the 3D Lebesgue measure, because the mean-value of Φ over Σ vanishes, we have

$\mu(\Omega_0) = \mu(\Omega_\Phi)$.

We interpret Ω_0 as a (planar) layer of material whose height may vary during the deposition process. As usual, we let greek subscripts $\{\alpha, \beta, \ldots\} \in \{1, 2\}$, roman subscripts $\{i, j, \ldots\} \in \{1, 2, 3\}$, we use bold face letters to denote vectors and tensors (e.g. $\boldsymbol{\sigma}, \boldsymbol{\varepsilon}, \mathbf{u}, \ldots$) while components of tensors are denoted using subscripts, e.g., $\boldsymbol{\sigma} = (\sigma_{ij})$.

2.2. Elastic problems

In this paper we consider only linear elastic materials. From a constitutive point of view our analysis can be applied up to orthotropic materials (this means that if components are expressed with respect to the orthotropy axes, $\varepsilon_{ij} = 0 \Longleftrightarrow \sigma_{ij} = 0$ for $i \neq j$) and in particular to cubic materials. The first results in this direction were obtained for isotropic materials (see Asaro and Tiller, 1972; Grinfeld, 1986, 1993). For completeness, we present all details for the orthotropic case and then we specialize the results to the cubic and isotropic materials.

The misfit between the substrate and the layer is denoted by $\boldsymbol{\varepsilon}^0 = (\varepsilon_{\alpha\beta})$. Since the upper surface of the layer is stress free (in fact, in experiments the outside pressure is maintained around 10^{-2} bars) we may suppose $\sigma_{33} = 0$, and in the case of orthotropic materials with two orthotropy axes in the (x_1, x_2)-plane, we get

$$\varepsilon^0_{33} = -\frac{1}{H_{3333}} H_{33\alpha\beta}\varepsilon^0_{\alpha\beta}, \qquad \varepsilon^0_{13} = \varepsilon^0_{23} = 0. \tag{1}$$

We denote by $\mathbb{H}$, the (fourth order) Hooke tensor, and by H_{ijkl} its components with respect to a given basis.

For a given misfit $\varepsilon^0_{\alpha\beta}$, equilibrium equations in Ω_0, for a stress-free upper surface, provides a displacement field as the solution of the following boundary value problem:

$(\mathscr{P}_0)$ *Find* $\mathbf{u}$ *such that* :

$$\begin{aligned}
&\operatorname{div}\boldsymbol{\sigma} = 0 \quad \text{in } \Omega_0,\\
&u_\alpha = \varepsilon^0_{\alpha\beta} x_\beta \quad \text{on } \Sigma_0 \cup S_h,\\
&u_3 = \varepsilon^0_{33} x_3 \quad \text{on} \quad \Sigma_0 \cup S_h,\\
&\boldsymbol{\sigma}\mathbf{n} = 0 \quad \text{on } \Sigma_h.
\end{aligned}$$

The boundary condition on the lateral boundary S_h is not completely justified as long as the choice of Σ (as a representative domain in the (x_1, x_2)-plane) is not selected by the underlying physics of the problem. A more realistic approach is to impose, as in the homogeneization theory, periodic boundary conditions on S_h and, in this case, the problem $\mathscr{P}_0$ can be regarded as a particular case of the following problem:

$(\mathscr{P}^\#_\Phi)$ *For* Σ *and* Φ, (x_1, x_2)-*periodic, find* $\mathbf{u}^\#$ *such that:*

$$\begin{aligned}
&\operatorname{div}\boldsymbol{\sigma}^\# = 0 \quad \text{in } \Omega_\Phi,\\
&\mathbf{u}^\# = \boldsymbol{\varepsilon}^0\mathbf{x} \quad \text{on } \Sigma_0,\\
&\mathbf{u}^\# = \boldsymbol{\varepsilon}^0\mathbf{x} + \mathbf{u}^{\text{per}}. \quad \text{on } S_{h+\Phi},\\
&\boldsymbol{\sigma}^\#\mathbf{n}^\Phi = \mathbf{t}^{\text{aper}}. \quad \text{on } S_{h+\Phi},\\
&\boldsymbol{\sigma}^\#\mathbf{n}^\Phi = 0 \quad \text{on } \Sigma_{h+\Phi},
\end{aligned}$$

where $\mathbf{u}^{\text{per.}}$ and $\mathbf{t}^{\text{aper.}}$ are, respectively, a periodic displacement field and an antiperiodic stress vector field. Existence and uniqueness for $\mathscr{P}_0$ and $\mathscr{P}^\#_\Phi$ are standard in suitable functional spaces. We note that, obviously, $\mathscr{P}_0$ has an homogeneous solution: $\mathbf{u} = \boldsymbol{\varepsilon}^0\mathbf{x}$ (this is due to our choice of ε^0_{33}). The elastic energy for this problem is

$$W_0 = \frac{1}{2}\int_{\Omega_0} \boldsymbol{\sigma}^0 : \boldsymbol{\varepsilon}^0 \mathrm{d}v = \frac{1}{2}\boldsymbol{\sigma}^0 : \boldsymbol{\varepsilon}^0 \mu(\Omega_0). \tag{2}$$

In order to compute the elastic energy for the solution of $\mathscr{P}^{\#}_{\Phi}$, denoted $W^{\#}_{\Phi}$ we note that the homogeneous solution is an *admissible displacement field* for $\mathscr{P}^{\#}_{\Phi}$ and thus, we directly conclude that

$$W^{\#}_{\Phi} \leqslant \frac{1}{2}\int_{\Omega_\Phi} \boldsymbol{\sigma}^0 : \boldsymbol{\varepsilon}^0 \mathrm{d}v = \frac{1}{2}\boldsymbol{\sigma}^0 : \boldsymbol{\varepsilon}^0 \mu(\Omega_\Phi) = W_0. \tag{3}$$

As a second argument for the above inequality, we claim that the difference $\overline{W} = W_0 - W^{\#}_{\Phi}$ (the energy gap) is given by

$$\overline{W} = \frac{1}{2}\int_{\Omega_\Phi} (\boldsymbol{\sigma}^0 - \boldsymbol{\sigma}^{\#}) : (\boldsymbol{\varepsilon}^0 - \boldsymbol{\varepsilon}^{\#})\mathrm{d}v. \tag{4}$$

To check this we have only to show that

$$\int_{\Omega_\Phi} \boldsymbol{\sigma}^{\#} : \boldsymbol{\varepsilon}^0 \mathrm{d}v = \int_{\Omega_\Phi} \boldsymbol{\sigma}^{\#} : \boldsymbol{\varepsilon}^{\#} \mathrm{d}v, \tag{5}$$

which follows from the fact that:

$$\int_{\Omega_\Phi} \boldsymbol{\sigma}^{\#} : (\boldsymbol{\varepsilon}^0 - \boldsymbol{\varepsilon}^{\#})\mathrm{d}v = \int_{\partial\Omega_\Phi} \boldsymbol{\sigma}^{\#}\mathbf{n}^{\Phi} \cdot (\mathbf{u}^0 - \mathbf{u}^{\#}) = 0 \tag{6}$$

and the second equality holds because $\mathbf{u}^0 = \mathbf{u}^{\#}$ on Σ, $\boldsymbol{\sigma}^{\#}\mathbf{n} = 0$ on $\Sigma_{\Phi+h}$, and the periodic conditions on the lateral boundary. The result in Eq. (4) is a second argument for the inequality in Eq. (3) but it is useful for a quantitative estimate of $\overline{W}$. We note here that a classical result in linear elasticity proves that $\overline{W}$ depends only on the values of $\mathbf{u}^{\#}$ on $\Sigma_{\Phi+h}$. Indeed, a straightforward computation shows that

$$\overline{W} = \frac{1}{2}\int_{\partial\Omega_\Phi} (\boldsymbol{\sigma}^0 - \boldsymbol{\sigma}^{\#})\mathbf{n}^{\Phi} \cdot (\mathbf{u}^0 - \mathbf{u}^{\#})\mathrm{d}a = \frac{1}{2}\int_{\Sigma_{\Phi+h}} \boldsymbol{\sigma}^0\mathbf{n}^{\Phi} \cdot (\mathbf{u}^0 - \mathbf{u}^{\#})\mathrm{d}a. \tag{7}$$

2.3. Extensions of energy estimates

We introduced in the previous section the problem $\mathscr{P}^{\#}_{\Phi}$ for practical purposes. In fact, for this problem Fourier analysis provides explicit solutions. However, $\mathscr{P}_0$ can also be considered as a special case of one of the following boundary value problems

($\mathscr{P}_\Phi$) *Find* $\mathbf{u}^\Phi$: $\Omega_\Phi \to \mathbb{R}^3$ *such that:*

$$\operatorname{div}\boldsymbol{\sigma}^\Phi = 0 \quad \text{in } \Omega_\Phi,$$
$$\mathbf{u}^\Phi = \boldsymbol{\varepsilon}^0\mathbf{x} \quad \text{on } \Sigma_0 \cup S_{h+\Phi},$$
$$\boldsymbol{\sigma}^\Phi\mathbf{n}^\Phi = 0 \quad \text{on } \Sigma_{h+\Phi}.$$

($\mathscr{P}^0_\Phi$) *Find* $\mathbf{u}^\Phi$: $\Omega_\Phi \to \mathbb{R}^3$ *such that:*

$$\operatorname{div}\boldsymbol{\sigma}^\Phi = 0 \quad \text{in } \Omega_\Phi,$$
$$\mathbf{u}^\Phi = \boldsymbol{\varepsilon}^0\mathbf{x} \quad \text{on } \Sigma_0,$$
$$u^\Phi_\alpha = \varepsilon^0_{\alpha\beta}x_\beta \quad \text{on } S_{h+\Phi},$$
$$\sigma^\Phi_{3\alpha}n^\Phi_\alpha = 0 \quad \text{on } S_{h+\Phi},$$
$$\boldsymbol{\sigma}^{\#}\mathbf{n}^\Phi = 0 \quad \text{on } \Sigma_{h+\Phi}.$$

Once again existence and uniqueness of solutions for problems $(\mathscr{P}_\Phi)$ and $(\mathscr{P}^0_\Phi)$ respectively, is a classical issue and arguments similar with those employed in Section 2.2 can be used to obtain inequalities similar to Eq. (3). If we denote by $\mathbf{u}_\Phi$ and $\mathbf{u}^0_\Phi$ the solutions of the problems $(\mathscr{P}_\Phi)$ and $(\mathscr{P}^0_\Phi)$ respectively we have

$$W(\mathbf{u}^\#) \leqslant W(\mathbf{u}_\Phi) \leqslant W_0, \tag{8}$$

$$W(\mathbf{u}^0_\Phi) \leqslant W_0. \tag{9}$$

3. The first order approximation and its general solution

An idea going back to Asaro and Tiller (1972), Grinfeld (1993), Noziéres (1991) was to estimate $\overline{W}$ using Fourier series. This result holds under the hypothesis of small amplitude perturbations of the free surface of the layer.

Generally, the normal field to $\Sigma_{h+\Phi}$ is

$$\mathbf{n}^\Phi = \frac{1}{(1+\|\nabla\Phi\|)^{1/2}}\left(-\frac{\partial\Phi}{\partial x_1}, -\frac{\partial\Phi}{\partial x_2}, 1\right), \tag{10}$$

and if $|\Phi|$ is small, say $\mathscr{O}(\epsilon)$, then at the first order we get

$$\mathbf{n}^\Phi = \mathbf{n} + \epsilon\overline{\mathbf{n}} + \mathscr{O}(\epsilon^2), \tag{11}$$

where

$$\overline{\mathbf{n}} = \left(-\frac{\partial\Phi}{\partial x_1}, -\frac{\partial\Phi}{\partial x_2}, 1\right).$$

We look for the first order solution of the problem $\mathscr{P}^\#_\Phi$ in the form $\mathbf{u}^\# = \mathbf{u}^0 + \epsilon\overline{\mathbf{u}}$. If $\overline{\boldsymbol{\varepsilon}}$ denotes the symmetric part of the $\nabla\overline{\mathbf{u}}$, and $\overline{\boldsymbol{\sigma}} = \mathbb{H}[\overline{\boldsymbol{\varepsilon}}]$, the first order problem, denoted in the subsequent $\mathscr{P}_I$, reduces to:

$(\mathscr{P}_I)$ *Find* $\overline{\mathbf{u}}$ *such that* :

$$\begin{aligned}
&\operatorname{div}\overline{\boldsymbol{\sigma}} = 0 \quad \text{in } \Omega_\Phi,\\
&\overline{\mathbf{u}} = 0 \quad \text{on } \Sigma_0,\\
&\overline{\mathbf{u}} = \mathbf{u}^{\text{per.}} \quad \text{on } S_{h+\Phi},\\
&\overline{\boldsymbol{\sigma}}\mathbf{n}^\Phi = \mathbf{t}^{\text{aper.}} \quad \text{on } S_{h+\Phi},\\
&\overline{\boldsymbol{\sigma}}\mathbf{n}^\Phi = -\boldsymbol{\sigma}^0\overline{\mathbf{n}} \quad \text{on } \Sigma_{h+\Phi}.
\end{aligned}$$

3.1. Formulation for small amplitude sinusoidal boundary

As stated before the method used to solve the first order problem will be based on Fourier series. The special case of a small amplitude sinusoidal boundary, where $\Phi = \epsilon e \sin(k_\alpha x_\alpha)$, is of particular interest because one can completely solve the problem $(\mathscr{P}_I)$. In the general setting for orthotropic materials there is a major technical difficulty related to the fact that, a priori, the axes x_1 and x_2 *are not axes of orthotropy*. Nevertheless, the general problem $(\mathscr{P}_I)$ can be solved and its solution will provide qualitative information on the *optimal* orientation of the *wave vector* $\mathbf{k}$. Moreover, this case covers situations of practical interest. For example, in some applications deposition is made on a plane which is not a symmetry plane for the cubic crystal (e.g. the crystallographic direction (111)) but with respect to this the material is still orthotropic.

Before going into details we shall compute $\overline{W}$ in terms of $\overline{\mathbf{u}}$. Using Eq. (6) we obtain

$$\overline{W} = -\frac{\epsilon}{2}\int_{\Sigma_{h+\Phi}} \boldsymbol{\sigma}^0\mathbf{n}^{\Phi}\cdot\overline{\mathbf{u}}\,\mathrm{d}a = -\frac{\epsilon^2}{2}\boldsymbol{\sigma}^0 : \int_{\Sigma_{h+\Phi}} \overline{\mathbf{n}}\otimes\overline{\mathbf{u}}\,\mathrm{d}a, \tag{12}$$

which can be approximated by

$$\overline{W} = -\frac{\epsilon^2}{2}\sigma^0_{\alpha\beta}\int_{\Sigma_h} \overline{n}_\beta\overline{u}_\alpha\,\mathrm{d}a = \frac{\epsilon^2}{2}\sigma^0_{\alpha\beta}\int_{\Sigma_h}\frac{\partial\Phi}{\partial x_\beta}\overline{u}_\alpha\,\mathrm{d}a. \tag{13}$$

Let $\{\mathbf{x},\mathbf{y},\mathbf{z}\}$ be a frame such that $\Phi(x_1,x_2) = \epsilon e\sin(kx)$. There exists a rotation $\mathbf{Q}$ such that $\mathbf{Q}\mathbf{x}_1 = \mathbf{x}$, $\mathbf{Q}\mathbf{x}_2 = \mathbf{y}$. The components of $\mathbf{Q}$ with respect to the axes $\{\mathbf{x}_1,\mathbf{x}_2,\mathbf{x}_3\}$ can be written as

$$\mathbf{Q} = \begin{pmatrix}\cos\theta & -\sin\theta & 0\\ \sin\theta & -\cos\theta & 0\\ 0 & 0 & 0\end{pmatrix}.$$

We denote the elastic coefficients with respect to axes $\{\mathbf{x}_1,\mathbf{x}_2,\mathbf{x}_3\}$ (which are actually the symmetry axes) with C_1 $(i = 1,\dots,9)$ and the Hooke's law can be written as

$$\begin{pmatrix}\sigma_{11}\\ \sigma_{22}\\ \sigma_{33}\end{pmatrix} = \begin{pmatrix}C_1 & C_4 & C_5\\ C_4 & C_2 & C_6\\ C_5 & C_6 & C_3\end{pmatrix}\begin{pmatrix}\varepsilon_{11}\\ \varepsilon_{22}\\ \varepsilon_{33}\end{pmatrix}, \tag{14}$$

$$\begin{pmatrix}\sigma_{12}\\ \sigma_{13}\\ \sigma_{23}\end{pmatrix} = \begin{pmatrix}C_7 & 0 & 0\\ 0 & C_8 & 0\\ 0 & 0 & C_9\end{pmatrix}\begin{pmatrix}\varepsilon_{12}\\ \varepsilon_{13}\\ \varepsilon_{23}\end{pmatrix}. \tag{15}$$

We choose Σ_0 to be the rectangle $(-\pi/k,\pi/k)\times(-1,1)$ (with respect to $(x,\ y,\ z)$ axes). Let $\Omega_0 = \Sigma_0\times[0,h]$; the normal field to $\overline{\mathbf{n}}$ is,

$$\overline{\mathbf{n}} = (-ke\cos(kx),0,0),$$

and the first order problem may be written as:

$\mathscr{P}_1$ *Find* $\overline{\mathbf{u}}$ *such that:*
(a) $\mathrm{div}\,\overline{\boldsymbol{\sigma}} = 0$ in Ω_0,
(b) $\overline{\mathbf{u}}(x,-1,z) = \overline{\mathbf{u}}(x,1,z),$ for $(x,z)\in(-\pi/k,\pi/k)\times(0,h)$,
(c) $\overline{\mathbf{u}}(-\pi/k,y,z) = \overline{\mathbf{u}}(\pi/k,y,z),$ for $(y,z)\in(-\pi/k,\pi/k)\times(0,h)$,
(d) $\overline{\mathbf{u}}(x,y,0) = 0,$ for $(x,y)\in(-\pi/k,\pi/k)\times(-1,1)$,
(e) $\overline{\boldsymbol{\sigma}}\mathbf{n}(x,-1,z) = -\overline{\boldsymbol{\sigma}}\mathbf{n}(x,1,z),$ for $(x,z)\in(-\pi/k,\pi/k)\times(0,h)$,
(f) $\overline{\boldsymbol{\sigma}}\mathbf{n}(-\pi/k,y,z) = -\overline{\boldsymbol{\sigma}}\mathbf{n}(\pi/k,y,z),$ for $(y,z)\in(-1,1)\times(0,h)$,
(g) $\overline{\boldsymbol{\sigma}}\mathbf{z} = -\boldsymbol{\sigma}^0\overline{\mathbf{n}},$ for $(x,y)\in(-\pi/k,\pi/k)\times(-1,1)$,
(h) $\overline{\boldsymbol{\sigma}} = \mathbb{H}[\overline{\boldsymbol{\varepsilon}}]$.

The isotropic case previously studied in Grinfeld (1993) is covered by relations (14) and (15) with $C_1 = C_2 = C_3 = \lambda + 2\mu,\ C_4 = C_5 = C_6 = \lambda$, and $C_7 = C_8 = C_9 = 2\mu$, while the cubic case is obtained for $C_1 = C_2 = C_3 = \lambda + 2\mu + \eta,\ C_4 = C_5 = C_6 = \lambda$, and $C_7 = C_8 = C_9 = 2\mu$.

3.2. Fourier series method

We look for solutions to $\mathscr{P}_1$ in the form

$$\overline{\mathbf{u}} = (eU(kz)\cos(kx), eV(kz)\cos(kx), eW(kz)\sin(kx)). \tag{16}$$

Subsequently, we denote $Z = kz$ and we use the prime to denote the derivative with respect to Z. We obtain for the strain tensor the following components:

$$\begin{pmatrix} \varepsilon_{xx} & \varepsilon_{xy} & \varepsilon_{xz} \\ & \varepsilon_{yy} & \varepsilon_{yz} \\ & & \varepsilon_{zz} \end{pmatrix} = \frac{ek}{2} \begin{pmatrix} -2\sin(kx)U & -\sin(kx)V & \cos(kx)(W + U') \\ & 0 & \cos(kx)V' \\ \text{sym.} & & 2\sin(kx)W' \end{pmatrix}, \tag{17}$$

and with respect to axes $\{\mathbf{x}_1, \mathbf{x}_2, \mathbf{x}_3\}$ we have generally

$$\varepsilon_{11} = c^2\varepsilon_{xx} - 2cs\varepsilon_{xy} + s^2\varepsilon_{yy},$$

$$\varepsilon_{12} = cs(\varepsilon_{xx} - \varepsilon_{yy}) + (c^2 - s^2)\varepsilon_{xy},$$

$$\varepsilon_{22} = s^2\varepsilon_{xx} + 2cs\varepsilon_{xy} + c^2\varepsilon_{yy},$$

$$\varepsilon_{13} = c\varepsilon_{xz} - s\varepsilon_{yz},$$

$$\varepsilon_{23} = s\varepsilon_{xz} + c\varepsilon_{yz},$$

$$\varepsilon_{33} = \varepsilon_{zz}.$$

The constitutive relations give

$$\begin{aligned}\sigma_{xx} = {} & \varepsilon_{zz}(C_5c^2 + C_6s^2) + \varepsilon_{xy}(-2C_1c^3s + 2C_4c^3s + 2C_7c^3s + 2C_2cs^3 - 2C_4cs^3 - 2C_7cs^3) \\ & + \varepsilon_{xx}(C_1c^4 + 2C_4c^2s^2 + 2C_7c^2s^2 + C_2s^4) + \varepsilon_{yy}(C_4c^4 + C_1c^2s^2 + C_2c^2s^2 - 2C_7c^2s^2 + C_4s^4),\end{aligned}$$

$$\begin{aligned}\sigma_{xy} = {} & \varepsilon_{zz}(-(C_5cs) + C_6cs) + \varepsilon_{xx}(-(C_1c^3s) + C_4c^3s + C_7c^3s + C_2cs^3 - C_4cs^3 - C_7cs^3) \\ & + \varepsilon_{yy}(C_2c^3s - C_4c^3s - C_7c^3s - C_1cs^3 + C_4cs^3 + C_7cs^3) + \varepsilon_{xy}(C_7c^4 + 2C_1c^2s^2 + 2C_2c^2s^2 \\ & - 4C_4c^2s^2 - 2C_7c^2s^2 + C_7s^4),\end{aligned}$$

$$\sigma_{xz} = \varepsilon_{yz}(-(C_8cs) + C_9cs) + \varepsilon_{xz}(C_8c^2 + C_9s^2),$$

$$\begin{aligned}\sigma_{yy} = {} & \varepsilon_{zz}(C_6c^2 + C_5s^2) + \varepsilon_{xy}(2C_2c^3s - 2C_4c^3s - 2C_7c^3s - 2C_1cs^3 + 2C_4cs^3 + 2C_7cs^3) \\ & + \varepsilon_{yy}(C_2c^4 + 2C_4c^2s^2 + 2C_7c^2s^2 + C_1s^4) + \varepsilon_{xx}(C_4c^4 + C_1c^2s^2 + C_2c^2s^2 - 2C_7c^2s^2 + C_4s^4),\end{aligned}$$

$$\sigma_{yz} = \varepsilon_{xz}(-(C_8cs) + C_9cs) + \varepsilon_{yz}(C_9c^2 + C_8s^2),$$

$$\sigma_{zz} = C_3\varepsilon_{zz} + \varepsilon_{xy}(-2C_5cs + 2C_6cs) + \varepsilon_{yy}(C_6c^2 + C_5s^2) + \varepsilon_{xx}(C_5c^2 + C_6s^2).$$

Using the above relations the equilibrium equations can be written as

$$\mathbf{PU}'' + \mathbf{NU}' + \mathbf{MU} = 0, \tag{18}$$

where $\mathbf{U}$ denotes the vector $(U(Z), V(Z), W(Z))$ and the *symmetric* matrices $\mathbf{M}$, $\mathbf{N}$ and $\mathbf{P}$ are given by

$$M_{xx} = -(s^2C_2 + c^2C_1) + s^2c^2(C_1 + C_2 - 2C_4 - 2C_7),$$

$$M_{xy} = sc^3(C_1 - C_4 - C_7) + cs^3(C_4 + C_7 - C_2),$$

$$M_{yy} = -C_7/2 + c^2s^2(2C_7 + 2C_4 - C_1 - C_2),$$

$$M_{zz} = (c^2C_8 + s^2C_9)/2, \quad M_{xz} = M_{yz} = 0,$$

$$N_{xz} = c^2C_5 + s^2C_6 + (c^2C_8 + s^2C_9)/2,$$
$$N_{yz} = +cs(C_6 - C_5) + cs(C_9 - C_8)/2,$$
$$N_{xx} = N_{xy} = N_{yy} = N_{zz} = 0,$$

$$P_{xx} = (c^2C_8 + s^2C_9)/2, \qquad P_{xy} = cs(C_9 - C_8)/2,$$
$$P_{yy} = (c^2C_9 + s^2C_8)/2, \qquad P_{zz} = -C_3, \quad P_{xz} = P_{yz} = 0.$$

There is a classical way to solve Eq. (18). If we denote $\mathbf{D}(Z) = (\mathbf{U}(Z), \mathbf{U}'(Z)) = (U(Z), V(Z), W(Z), U'(Z), V'(Z), W'(Z))$, system (18) is equivalent to

$$\frac{\mathrm{d}\mathbf{D}}{\mathrm{d}Z} = \mathbb{M}\mathbf{D}. \tag{19}$$

where the matrix $\mathbb{M}$ is defined as

$$\mathbb{M} = \begin{pmatrix} 0 & \mathbf{I} \\ -\mathbf{P}^{-1}\mathbf{M} & -\mathbf{P}^{-1}\mathbf{N} \end{pmatrix}.$$

The general solution for Eq. (19) is

$$\mathbf{D}(Z) = \exp(\mathbb{M}Z)\mathbf{D}(0), \tag{20}$$

and the (3×3) blocks of $(\mathbb{M}Z)$ will be denoted in the following:

$$\exp(\mathbb{M}Z) = \begin{pmatrix} \mathbf{E}(Z) & \mathbf{F}(Z) \\ \mathbf{G}(Z) & \mathbf{H}(Z) \end{pmatrix}. \tag{21}$$

3.3. The boundary conditions

The general form of the solution (16) satisfies obviously $\mathscr{P}_1$(b), $\mathscr{P}_1$(c), $\mathscr{P}_1$(e), $\mathscr{P}_1$(f) and the remaining boundary conditions to be satisfied are $\mathscr{P}_1$(d) and $\mathscr{P}_1$(g). Condition $\mathscr{P}_1$(d) gives

$$U(0) = V(0) = W(0) = 0, \tag{22}$$

and $\mathscr{P}_1$(g) is equivalent to

$$\overline{\sigma}_{xz} = ek\cos(kx)\sigma^0_{xx}, \qquad \overline{\sigma}_{yz} = ek\cos(kx)\sigma^0_{xx}. \tag{23}$$

With respect to the axes $\{\mathbf{x}_1, \mathbf{x}_2, \mathbf{x}_3\}$ condition (23) becomes

$$cC_8\varepsilon_{33} + sC_9\varepsilon_{23} = -ek\cos(kx)(c^2\sigma^0_{11} + 2sc\sigma^0_{12} + s^2\sigma^0_{22}),$$
$$-sC_8\varepsilon_{33} + cC_9\varepsilon_{23} = -ek\cos(kx)(-cs\sigma^0_{11} + (s^2 - c^2)\sigma^0_{12} + sc\sigma^0_{22}),$$
$$C_5\varepsilon_{11} + C_6\varepsilon_{22} + C_3\varepsilon_{33} = 0,$$

which is equivalent to

$$C_8\varepsilon_{13} = -ek\cos(kx)(c\sigma^0_{11} + s\sigma^0_{12}),$$
$$C_9\varepsilon_{23} = -ek\cos(kx)(c\sigma^0_{12} + s\sigma^0_{22}),$$
$$C_5\varepsilon_{11} + C_6\varepsilon_{22} + C_3\varepsilon_{33} = 0.$$

Taking into account Eq. (17) we can write the above relations as

$$c(W(kh) + U'(kh)) - cV'(kh) = -\frac{2}{C_8}(c\sigma^0_{11} + s\sigma^0_{12}),$$

$$s(W(kh) + U'(kh)) + cV'(kh) = -\frac{2}{C_9}(c\sigma^0_{12} + s\sigma^0_{22}), \tag{24}$$

$$C_5(csV(kh) - c^2U(kh)) + C_6(csV(kh) - s^2U(kh)) + C_3W'(kh) = 0.$$

In terms of $\mathbf{U}(Z)$ Eq. (24) can be written as

$$\mathbf{A}\mathbf{U}(kh) + \mathbf{B}\mathbf{U}'(kh) = -\mathbf{\Lambda}, \tag{25}$$

where

$$\mathbf{A} = \begin{pmatrix} 0 & 0 & \frac{1}{2}(C_8c^2 + C_9s^2) \\ 0 & 0 & \frac{1}{2}cs(C_9 - C_8) \\ -(C_5c^2 + C_6s^2) & cs(C_5 - C_6) & 0 \end{pmatrix},$$

$$\mathbf{B} = \begin{pmatrix} \frac{1}{2}(C_8c^2 + C_9s^2) & \frac{1}{2}cs(C_9 - C_8) & 0 \\ \frac{1}{2}cs(C_9 - C_8) & \frac{1}{2}(C_9c^2 + C_8s^2) & 0 \\ 0 & 0 & C_3 \end{pmatrix}, \quad \mathbf{\Lambda} = \begin{pmatrix} \sigma^0_{xx} \\ \sigma^0_{xy} \\ 0 \end{pmatrix}. \tag{26}$$

3.4. Explicit estimate for the energy gap $\overline{W}$

By using Eq. (20) and taking into account condition (22) we get

$$\mathbf{U}(Z) = \mathbf{F}(Z)\mathbf{U}'(0), \quad \mathbf{U}'(Z) = \mathbf{H}(Z)\mathbf{U}'(0) = \mathbf{F}'(Z)\mathbf{U}'(0).$$

Eq. (25) gives

$$\mathbf{U}'(0) = -(\mathbf{A}\mathbf{F}(kh) + \mathbf{B}\mathbf{F}'(kh))^{-1}\mathbf{\Lambda}, \tag{27}$$

and the displacement is obtained as

$$\mathbf{U}(Z) = -\mathbf{F}(Z)(\mathbf{A}\mathbf{F}(kh) + \mathbf{B}\mathbf{F}'(kh))^{-1}\mathbf{\Lambda}. \tag{28}$$

which is a linear expression in $\boldsymbol{\sigma}^0$. We note that Eq. (13) shows that to compute $\overline{W}$ we need only $U(kh)$ and $V(kh)$. Indeed, formula (13) gives for $\overline{W}$

$$\overline{W} = -\frac{e^2\epsilon^2}{2}\int_{-\pi/k}^{\pi/k}\int_{-1}^{1}\Big[k\cos^2(kx)(\sigma^0_{xx}U(kh) + \sigma^0_{xy}V(kh))\Big]\mathrm{d}x\,\mathrm{d}y = -\pi e^2\epsilon^2(\sigma^0_{xx}U(kh) + \sigma^0_{xy}V(kh)). \tag{29}$$

Eq. (28) gives

$$\mathbf{U}(kh) = -\mathbf{F}(kh)(\mathbf{A}\mathbf{F}(kh) + \mathbf{B}\mathbf{F}'(kh))^{-1}\mathbf{\Lambda} = -(\mathbf{A} + \mathbf{B}\mathbf{F}'(kh)\mathbf{F}^{-1}(kh))^{-1}\mathbf{\Lambda}, \tag{30}$$

and it is obvious from Eqs. (29) and (30) that $\overline{W}$ is a quadratic form in $\sigma^0_{\alpha\beta}$.

4. The long wave-length limit

4.1. The general case

In this section using the form (30) of the general solution for the first order problem we study the behavior of $\overline{W}$ for kh small. We recall here that the height of the layer is denoted h and the wave length of the perturbation of the upper boundary is π/k. Thus, kh small means *magnitude of the thickness of the layer much smaller than magnitude of the wave length of the perturbation.*

Using the definition of $\mathbf{F}$ one obtains easily

$$\mathbf{F}(Z) = Z\mathbf{I} - \frac{Z^2}{2}\mathbf{P}\mathbf{N}^{-1} + \frac{Z^3}{6}\left(\mathbf{P}\mathbf{N}^{-1}\mathbf{P}\mathbf{N}^{-1} - \mathbf{P}\mathbf{M}^{-1}\right) + \mathcal{O}(Z^4). \tag{31}$$

If we denote $\mathbf{K}(Z) = \mathbf{F}(Z)(\mathbf{A}\mathbf{F}(Z) + \mathbf{B}\mathbf{F}'(Z))^{-1}$ and use relation (30), we obtain

$$\mathbf{K}(0) = 0, \qquad \frac{\mathrm{d}\mathbf{K}}{\mathrm{d}Z}(0) = \mathbf{B}^{-1}, \tag{32}$$

$$\frac{\mathrm{d}^2\mathbf{K}}{\mathrm{d}Z^2}(0) = \left(\mathbf{P}^{-1}\mathbf{N} - 2\mathbf{B}^{-1}\mathbf{A}\right)\mathbf{B}^{-1}, \tag{33}$$

$$\frac{\mathrm{d}^3\mathbf{K}}{\mathrm{d}Z^3}(0) = \left[\left(\mathbf{P}^{-1}\mathbf{N} - 3\mathbf{B}^{-1}\mathbf{A}\right)^2 - 3\mathbf{B}^{-1}\mathbf{A}\mathbf{B}^{-1}\mathbf{A} + 2\mathbf{P}^{-1}\mathbf{M}\right]\mathbf{B}^{-1}. \tag{34}$$

We note that Eq. (29) show that only components $\mathbf{K}_{\alpha\beta}$ are involved in the computation of $\mathbf{U}(kh)$ and using relations (26) and (33) one can show that

$$\left[\frac{\mathrm{d}^2\mathbf{K}}{\mathrm{d}Z^2}(0)\right]_{\alpha\beta} = 0. \tag{35}$$

It follows that Taylor expansion of $\mathbf{K}(Z)$ contains only the first and the third order terms given by Eqs. (32) and (34). In the general case the expression of the third order term is to complicated to be relevant but one can have an explicit form for the first order term. Using the above results we obtain for the orthotropic case

$$U(kh) = -\frac{2kh}{C_9C_8}\left[(\cos^2(\theta)C_9 + \sin^2(\theta)C_8)\sigma^0_{xx} + (C_8 - C_9)\sin(\theta)\cos(\theta)\sigma^0_{xy}\right], \tag{36}$$

$$V(kh) = -\frac{2kh}{C_9C_8}\left[(\cos^2(\theta)C_8 + \sin^2(\theta)C_9)\sigma^0_{yy} + (C_8 - C_9)\sin(\theta)\cos(\theta)\sigma^0_{xy}\right] \tag{37}$$

and using Eqs. (36) and (37) in Eq. (29) we obtain for $\overline{W}$ a quadratic form in σ^0_{xx} and σ^0_{xy} whose coefficients depend on θ. Obviously, one can find θ in order to maximize $\overline{W}$. This case, which is actually the more general setting, is not so important in applications. Rather one have in practical circumstances for the shear stress $\sigma^0_{xy} = 0$ and in this case the expression for $\overline{W}$ is

$$\overline{W} = 2\pi\epsilon^2 e^2 kh\left[\frac{\cos^2(\theta)}{C_8} + \frac{\sin^2(\theta)}{C_9}\right](\sigma^0_{xx})^2, \tag{38}$$

which is maximized for $\theta = 0$ if $C_8 < C_9$ and $\theta = \pi/2$ if $C_9 < C_8$ and is independent of θ if $C_8 = C_9$ which is the case for cubic (and in particular isotropic) materials.

4.2. The cubic case

This is the most important case for applications. We have: $C_1 = C_2 = C_3 = \lambda + 2\mu + \eta$, $C_4 = C_5 = C_6 = \lambda$, $C_7 = C_8 = C_9 = 2\mu$, and we obtain using relations (36) and (37)

$$\overline{W} = \frac{\pi\epsilon^2 e^2 kh}{\mu}\left[(\sigma^0_{xx})^2 + \left(\sigma^0_{xy}\right)^2\right] \tag{39}$$

so that the first order term is independent on θ. Moreover, in almost all applications $\sigma^0_{xy} = 0$, thus one obtains in Eq. (39)

$$\overline{W} = \frac{\pi\epsilon^2 e^2 kh}{\mu}\left(\sigma^0_{xx}\right)^2. \tag{40}$$

In the cubic case the third order term can be computed explicitly and Eq. (34) becomes

$$\overline{W} = \frac{\pi\epsilon^2 e^2}{\mu}\left(\sigma^0_{xx}\right)^2\left[kh + Ak^3h^3\right], \tag{41}$$

where

$$A = \frac{-1}{3\mu(\lambda + 2\mu + \eta)}\left[\eta(\lambda + 2\mu + \eta)\frac{1 + \cos^2(2\theta)}{2} + 3\mu^2 + 5\lambda\mu + \lambda\eta + 2\mu\eta\right]. \tag{42}$$

4.3. Exact solution in the isotropic case

The isotropic case is interesting because in this case the equations for U and W are independent from that of V and one can obtain an explicit expression for $\mathbf{U}(Z)$. This procedure was partially developed in Grinfeld (1993). From relations (18) and (24) and using the Poisson ratio, $v = \lambda/2(\lambda + \mu)$, $b = -\sigma^0_{xx}/\mu$ the system of ODE for U and W is

$$(1 - 2v)U'' + W' + 2(v - 1)U = 0,$$

$$2(v - 1)W'' + U' + (1 - 2v)W = 0,$$

$$U(0) = W(0) = 0,$$

$$(1 - v)W'(kh) - vU(kh) = 0, \quad W(kh) + U'(kh) = b.$$

One finds $U(kh)$ as

$$U(kh) = b\left[\frac{(4v - 3)(v - 1)\cosh(kh)\sinh(kh) - kh(v - 1)}{(3 - 4v)\sinh(kh)\sinh(kh) + k^2h^2 + 4(v - 1)^2}\right], \tag{43}$$

while the equation and boundary conditions for V give

$$V(kh) = -\frac{\sigma^0_{xy}}{\mu}\tanh(kh). \tag{44}$$

The Taylor expansion in Eq. (43) give

$$U(kh) = -\frac{\sigma^0_{xx}}{\mu}\left[kh + \frac{4v + 3}{6(v - 1)}k^3h^3 + \mathcal{O}(k^4h^4)\right]. \tag{45}$$

Obviously the relation (45) can be obtained directly via the cubic case with $\eta = 0$ and using the Poisson ratio. We conclude that *in the isotropic case and without misfit shear* the energy gap

$$\overline{W} = \frac{\pi\epsilon^2 e^2}{\mu}\left[kh + \frac{4v + 3}{6(v - 1)}k^3h^3 + \mathcal{O}(k^4h^4)\right]\left(\sigma^0_{xx}\right)^2. \tag{46}$$

5. The surface energy

5.1. The critical thickness, h_c

It follows from the previous section (see e.g., Eq. (46)) that $\overline{W}$ increases with k and thus the bulk energy is lowered by rapid oscillations of the free surface. To prevent this situation, one usually adds (Grinfeld, 1993; Nozières, 1991) to the bulk term, W, a surface energy contribution which is, in the general case, expressed as

$$\int_{\Sigma_{h+\Phi}} \psi(\mathbf{n}_\phi)\mathrm{d}a. \tag{47}$$

In the particular case of a constant surface energy density, i.e. when $\psi(\mathbf{n}) = \psi_0$ the integral in Eq. (47) equals area $(\Sigma_{h+\Phi})$. The case of a constant surface energy density is of particular interest since, in general, when the surface energy density depends on the orientation of the free surface the force system at the surface changes. In the following we shall suppose that the surface energy density is constant and thus, for the wavy boundary the surface contribution is

$$\int_{-k/\pi}^{k/\pi}\int_{-1}^{1} \psi_0\sqrt{1+(\nabla\Phi)^2}\,\mathrm{d}x\,\mathrm{d}y = \psi_0 \text{ area } (\Sigma_{h+\Phi}). \tag{48}$$

As in the bulk, for small amplitude perturbations, when we restrict attention to the terms of second order in ϵ, we find that the right-hand side in Eq. (48) becomes

$$\psi_0 \text{ area } (\Sigma_{h+\Phi}) \sim \frac{4\pi}{k}\psi_0\left[1+\frac{\epsilon^2 e^2}{4}\right]. \tag{49}$$

If we denote further

$$W_0^{\text{total}} = W_0 + \frac{4\pi}{k}\psi_0, \tag{50}$$

and

$$W_\phi^{\text{total}} = W_\phi + \frac{4\pi}{k}\psi_0\left[1+\frac{\epsilon^2 e^2}{4}\right], \tag{51}$$

the total energy difference is given by $\Delta W^{\text{total}} = W_0^{\text{total}} - W_\phi^{\text{total}}$. Up to second order terms in ϵ, relations (50) and (51) lead to

$$\Delta W^{\text{total}} = \overline{W} - \psi_0\frac{\epsilon^2 e^2 \pi}{k}. \tag{52}$$

Let us consider the simple case of a cubic material without misfit shear. Using Eq. (40) in Eq. (52) we obtain

$$\Delta W^{\text{total}} = \pi\epsilon^2 e^2 k\left[\frac{h(\sigma_{xx}^0)^2}{\mu} - \psi_0\right]. \tag{53}$$

If the thickness of the layer is big enough then the configuration with a wavy boundary has less energy than the one with a flat boundary. This fact evidences a critical thickness of the layer, denoted in the following h_c, such that the planar boundary is stable (locally) if $h < h_\mathrm{c}$, for

$$h_\mathrm{c} = \frac{\mu\psi_0}{(\sigma_{xx}^0)^2}. \tag{54}$$

Alternatively, for small h the configuration with a flat boundary is stable to small amplitude sinusoidal perturbations. In order estimate the magnitude of h_c we discuss in the following the cubic case without misfit shear.

By using the lattice parameters, for $In_{1-x}Ga_xAs$ alloy at $x = 0.18$ deposed on InP substrate the misfit equals $\varepsilon_{xx} = 0.02$. The value of surface energy, ψ_0 is $\sim$0.336 N/m (which corresponds to 21 meV/$\mathring{A}^2$, see Gendry et al., 1997) and the elastic moduli ($In_{1-x}Ga_xAs$ alloy is a cubic material) are $E \sim 57 \times 10^9$ N/m^2, $\mu \sim 40 \times 10^9$ N/m^2, $\nu \sim 0.31$. In this case formula (54) gives a critical thickness

$$h_{cr} \sim 5.6 \times 10^{-9} \text{ m} \sim 56 \text{ Å}. \tag{55}$$

This result seems to be in good agreement with experiments reported in Gendry et al. (1997), for strained epitaxial structures obtained by molecular beam epitaxy and observed via scanning tunneling microscopy. It must be noted that the experience is very sensitive to experimental conditions not considered here such as the temperature, the orientation of the growth plane with respect to the crystal symmetry, etc.

6. Conclusions

We point out in the following several remarks concerning the results obtained, the range of the method employed, its limitations and perspectives.

1. The case we have considered here is much larger than previously discussed situations (Asaro and Tiller, 1972, Grinfeld, 1993, 1991). We treat the orthotropic case which covers, as already mentioned, epitaxy on planes other the crystallographic ones in cubic materials and thus is of major interest in applications. We obtained a general energy estimate that holds in the case of an orthotropic materials and then we specialize it to the case of cubic and isotropic materials. Results obtained using the bulk elastic energy and the contribution of the surface energy provide a relation for the critical thickness that seems to be (quantitatively) in good agreement with experimental results.

2. A straightforward analysis of the above approach leads to the conclusion that Fourier method employed in this problem was necessary in order to obtain an explicit estimate for $\overline{W}$. The tools used for a sinusoidal boundary apply also to the analysis of an arbitrary boundary with one translational symmetry (by decomposition in Fourier series of the boundary and by summing up the energy contributions of each mode). We may conclude that as long as small perturbation analysis is concerned one will obtain (for an arbitrary perturbation ϕ) an estimate concerning for $\overline{W}$ given by

$$\overline{W} = K_1 h \|\phi\|_{H^1(\Sigma_0)} \tag{56}$$

for some constant K_1 *quadratic* in the misfit. If we consider a constant free energy density on the surface, and once again, taking into account only the second order terms, we get an estimate of the surface energy contribution in the form

$$K_2 \|\phi\|_{H^1(\Sigma_0)}. \tag{57}$$

Thus the *conclusion obtained in* Eq. (54) *holds for an arbitrary geometry of the perturbation*. There is no major interest to present the analysis of the general case here, since intricate computations will minimize the physical interest of the problem.

3. There is experimental evidence of the fact that the sign of the misfit stress (compressive stress or extensive stress) does not affect in the same way the growth of the free surface. However, by inspection of Eqs. (29), (36), (37) (or previously cited work, Grinfeld (1993), one can note that the critical thickness obtained with our method, does not depends on the sign of the misfit stress. This is due to the fact that in this work the surface energy is supposed constant. However, if the surface energy is not considered as

constant the boundary condition at the free-surface does not involve only the bulk stress but also the superficial divergence of surface stress. That problem seems much much more difficult than the situation discussed here, and such an approach is the subject of work in progress.

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