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Coherency stresses in multilayers

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Abstract

The determination of the elastic state of coherently matched layers is of importance in a wide range of domains: epitaxial films on a substrate with different crystal structures, deformation of a lamella welded on a substrate, lamellar crystals.
It is shown that the elastic state of coherently matched multilayers depends on two coupled field quantities: the stress (or equivalently the elastic strain), and the curvature. A general method, is derived to determine these fields, and the contribution of curvature on stress relaxation is emphasized. Detailed applications are given for the case of stress-free dilatation, and pure shear.

1. Introduction

Consider two parallel slices of crystals S, and F, (figure 1a) and let ($\mathbf{a}^s$, $\mathbf{b}^s$) and ($\mathbf{a}^f$, $\mathbf{b}^f$) be the two-dimensional lattice vectors defining the orientation of the free-standing substrate and of the free standing film respectively (figure 1b). Coherent match is achieved by imposing an adequate elastic strain on both slices. In the case of a multilayer (figure 1c), one must impose an elastic strain on each slice.
The elastic state of such a coherent multilayer depends on two coupled field quantities: the stress (or equivalently the elastic strain) and the curvature, depending themselves on the geometric relation between the crystal lattices of the layers, and of their elastic modulii. Explicit calculations have been made in the particular case where the geometric relation between the crystal lattices of the layers is a pure dilatation [1-3]. The purpose of this paper is to give a unified solution to the problem whatever this geometric relation: dilatation, shear, or combined shear and dilatation.
As shown by Eshelby [4], and Kröner [5], the elastic state is completely determined, for a given geometry, by the knowledge of the stress-free strain, or equivalently the transformation strain relating the various crystal lattices before matching, and by the usual elasticity equations and boundary conditions. A rigorous definiton of the stress-free strain relevant to our problem is given in section 2. The corresponding procedures allowing the elastic state of a coherent multilayer to be determined from the knowledge of the stress-free strain are described in section 3.
Taking elastic anisotropy into account is possible at the price of standard but heavy calculations that blur the physical meaning of the results. Useful equations are therefore given in the appendix, but they will not be discussed. The general explicit expressions of the elastic strain, the stress, and the curvature, in elastically isotropic materials are developped in section 4. In section 5, these expressions are applied to two-phases multilayers whose geometries are represented figure 2. Section 6 investigates the case where the stress-free strain is a pure shear, in relation with lamellar TiAl. We conclude in section 7.

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2. Geometry: stress-free strain and stress-free distortion

2.1. Bilayers

Consider a substrate S and a film F, with thicknesses $w^s$and w^f, respectively (figure 1a). In what follows quantities related to S (resp F) will be denoted by the subscript, or superscript, *s* (resp *f*). Let *w* be the total thickness of the plate, and *f* the volume fraction of the film:

$$w = w^s + w^f \quad (1a)$$
$$w^f = fw \quad (1b)$$

Let $\delta\mathbf{a}$, $\delta\mathbf{b}$, be defined by (figure 1b):

$$\delta\mathbf{a} \equiv \mathbf{a}^s - \mathbf{a}^f = \mathbf{B}^s\,\mathbf{a}^f \quad (2a)$$
$$\delta\mathbf{b} \equiv \mathbf{b}^s - \mathbf{b}^f = \mathbf{B}^s\,\mathbf{b}^f \quad (2b)$$

The homogeneous second-order tensor $\mathbf{B}^s$, is the *stress-free distortion* tensor. Itdefines the *geometrical transformation* of the *stress-free film* lattice, into the *stress-free substrate* lattice. The *stress-free strain* $\mathbf{E}^s$ is the symmetric part of $\mathbf{B}^s$. By construction $\mathbf{B}^f$ and $\mathbf{E}^f$ are nil.
With the axis Ox_1 chosen normal to the interface (figure 1a), the non-zero components of $\mathbf{B}^s$ are derived by solving equations (2) :

$$\begin{pmatrix} B^s_{22} & B^s_{23} \\ B^s_{32} & B^s_{33} \end{pmatrix} = \frac{\begin{pmatrix} b^f_3\delta a_2 - a^f_3\delta b_2 & a^f_2\delta b_2 - b^f_2\delta a_2 \\ b^f_3\delta a_3 - a^f_3\delta b_3 & a^f_2\delta b_3 - b^f_2\delta a_3 \end{pmatrix}}{a^f_2 b^f_3 - a^f_3 b^f_2} \quad (3)$$

It is useful to have in mind the correspondence between the stress-free distortion tensor and the geometrical transformation in some particular cases:

Pure dilatation: $B^s_{23} = B^s_{32} = B^s_{22} - B^s_{33} = 0$
Pure shear parallel to the axis Ox_2: $B^s_{22} = B^s_{33} = B^s_{32} = 0$
Pure shear parallel to the axis Ox_3: $B^s_{22} = B^s_{33} = B^s_{23} = 0$

2.2. Multilayers

Consider a plate of thickness *w* comprising *n* lamellae coherently matched along their planar interfaces, each of thickness w^q (q=1,2,...,n) and with volume fraction $f^q = \frac{w^q}{w}$ (figure 1c). Let ($\mathbf{a}^q$, $\mathbf{b}^q$) be the lattice vectors of lamella *q*. The transformation of the lattice of plate *n* into the lattice of plate *q* in a plane parallel to the interface is defined by the stress-free distortion tensor $\mathbf{B}^q$ as:

$$\mathbf{a}^q - \mathbf{a}^n = \mathbf{B}^q\,\mathbf{a}^n \quad (4a)$$
$$\mathbf{b}^q - \mathbf{b}^n = \mathbf{B}^q\,\mathbf{b}^n \quad (4a)$$

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The *stress-free strain* $\mathbf{E}^q$ is the symmetric part of $\mathbf{B}^q$. By construction, $\mathbf{B}^n$ and $\mathbf{E}^n$ are nil.

3. Statics

3.1. The Eshelby procedure [4]

This procedure is, in some sense, the most natural one:
a) Consider a stack made of *n* lamellae all with the lattice vectors of lamella *n*. The lamellae match perfectly.
b) Make cuts along all each interface and apply to each lamella *q* a *stress-free* distortion $\mathbf{B}^q$ defined by equation (4). The lamellae no longer match.
c) Restore coherent match by giving each lamella, an *elastic deformation* - $\mathbf{E}^q$ by means of a system of a system of stresses $\boldsymbol{\sigma}^{Eq}$ applied at the surface of each lamella.
d) Stick all the coherently matched lamellae together.
e) Relax the stresses $\boldsymbol{\sigma}^{Eq}$ by application of a system of stress $-\boldsymbol{\sigma}^{Eq}$.
The elastic state of the multilayer, i. e. the internal stress, the elastic strain and the curvature, is completely determined by these conditions. The internal stress is a consequence of the varying stress-free distortion in the multilayer.
Two points are worth noticing:

- the elastic state of the system does not depend on the reference lamella used to define the set of $\mathbf{B}^q$, since superimposing a homogeneous stress-free distortion $\mathbf{B}^*$ to the multilayer transforms each $\mathbf{B}^q$ into $\mathbf{B}^q+\mathbf{B}^*$ without changing its elastic state.
- one may impose an arbitrary rotation to each lamella without changing the elastic state. Therefore the above conditions are not sufficient to determine the *crystallographic* state. The latter is thus determined up to *n* rotations which are controlled by local interactions at the atomic level. For example, epitaxial films of Al on Al_20_3 exhibit a $\{111\}\pm\langle 110\rangle \mathrm{Al}\left\|(0001)\langle 10\bar{1}0\rangle\alpha-\mathrm{Al_2O_3}\right.$ orientation relationship, while epitaxial films of Cu on αAl_20_3 exhibit a $\{111\}\pm\langle 112\rangle \mathrm{Cu}\left\|(0001)\langle 10\bar{1}0\rangle\alpha-\mathrm{Al_2O_3}\right.$ orientation relationship [6].

3.2. The Kröner procedure [5]

a) Consider a stack made of *n* lamellae again all with the lattice vectors in P identical to those of lamella *n*.
b) Give each lamella *q* a stress-free distortion $\mathbf{B}^q$ defined by equation (4).
c) Recover the matching by a homogeneous surface dislocation distribution on each interface.
These dislocations, named *quasi-dislocations*, *are not* crystal dislocations but a *mathematical device* allowing the calculation of the coherency stresses. Their stress field, and their curvature field are calculated by standard methods.
This approach is particularly useful for the analysis of the stress relaxation by interfacial dislocation networks. The method consists in superimposing to the quasi-dislocations network, a periodic planar dislocation array made of crystal dislocations:

- Given the Miller indices of the interface, and the available Burgers vectors of the crystal dislocations of the structure it is possible to determine periodic planar dislocations arrays that relax the coherency stresses completely. There is not in general a unique solution to the problem [7].

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- Given the coherency stress and the Miller indices of the interface, the dislocation network determines the amount of stress relaxation [8].

Another way to state that the crystallographic state is determined up to *n* arbitrary rotations is to notice that the superposition of a Nye wall [9] to the quasi-dislocation distribution at one interface modifies the rotation field without changing the stress field.

3.3. Constraints imposed to the elastic field

Let $\boldsymbol{\sigma}^q$, $\boldsymbol{\varepsilon}^q$, $\mathbf{S}^q$, be the (internal) stress, the elastic strain, and the compliance tensor in lamella *q*. We have the general relation:

$$\boldsymbol{\varepsilon}^q = \mathbf{S}^q.\boldsymbol{\sigma}^q \qquad (5a)$$

$$\varepsilon_{ij}^q = \sum_{k,l=1}^{k,l=3} S_{ijkl}^q \sigma_{kl}^q \qquad (5b)$$

For clarity sake, we do not use the Einstein summation rule in this paper.
The total strain $\mathbf{E}^{Tq}$ in each lamella *q* is the sum of the stress-free strain $\mathbf{E}^q$, and of the elastic strain $\boldsymbol{\varepsilon}^q$:

$$\mathbf{E}^{Tq} = \mathbf{E}^q + \boldsymbol{\varepsilon}^q \qquad (6)$$

The elastic field must satisfy two kinds of conditions: continuity of the strain and elastic equilibrium:
1. Coherency imposes the continuity and the compatibility of the total strain in the multilayer.
2. The stress must satisfy equilibrium conditions in the bulk, at the interfaces, and at the free surface. These conditions are simply written as:
a) *in the bulk*:
Equilibrium requires that:

$$\sum_{j=1}^{j=3} \frac{\partial \sigma_{ij}}{\partial x_j} = 0 \qquad (7)$$

Besides, from Albenga's theorem the average of the internal stress must be zero.
b) *at the interfaces*:
The stress must not present discontinuities. Using the coordinate system with the reference axis Ox_1 normal to plane P, pictured in figure 1a, these conditions are simply that σ_{1j} must be continuous.
c) *at the free surface normal to* Ox_1:
The force must be nil at all points of the surface, which imposes:

$$\sigma_{1j} = 0 \qquad (8)$$

d) *at the free surface parallel to* Ox_1 *(lateral external surfaces)*
Since the thickness of the multilayer is small compared to its lateral dimensions, it is relevant to make use of the Saint Venant principle, and to impose simply that the average force, and the average moment be zero on these surfaces.
For simplicity sake, the equations corresponding to the Albenga's theorem, and to the conditions on the lateral surfaces, will be written after simplifying the expression of the elastic field (§411).

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4. Isotropic elasticity

We now demonstrate that the stress field in each lamella is the sum of two terms: one term is homogeneous and the second varies linearly with the distance to the interface. With the stress gradient is coupled a curvature which is homogeneous in the whole multilayer. This solution will be referred to as the "general solution". In section 4-2, we develop a solution in which the curvature and the stress are decoupled [10, 11] referred to as the "approximate solution".

4.1. General solution

4.1.1. Stress

One simple way to satisfy the above equilibrium equations at the interfaces and at the external surfaces perpendicular to Ox_1, is to impose a state of planar stress in the multilayer, by setting:

$$\sigma_{1j} = 0 \tag{9}$$

We define:

$$(\sigma_1, \sigma_2, \sigma_3) \equiv \left(\frac{\sigma_{22} + \sigma_{33}}{2}, \frac{\sigma_{22} - \sigma_{33}}{2}, \sigma_{23} \right) \tag{10a}$$

$$(\varepsilon_1, \varepsilon_2, \varepsilon_3) \equiv \left(\frac{\varepsilon_{22} + \varepsilon_{33}}{2}, \varepsilon_{22} - \varepsilon_{33}, 2\varepsilon_{23} \right) \tag{10b}$$

$$\left(E_1^q, E_2^q, E_3^q\right) \equiv \left(\frac{E_{22}^q + E_{33}^q}{2}, E_{22}^q - E_{33}^q, 2E_{23}^q \right) \tag{10c}$$

$$M = 2\mu \frac{1+\nu}{1-\nu} \tag{10d}$$

The stress, the elastic strain and the stress-free strain are completely determined by the quantities σ_i, ε_i, E_i^q (i=1,2,3). M is known as the biaxial modulus, μ, ν are the shear modulus and the Poisson ratio, respectively.
With these notations, the relation between the stress and the elastic strain is simply:

$$\sigma_i = m_i \, \varepsilon_i \qquad (i=1, 2, 3) \tag{11a}$$

$$(m_1, m_2, m_3) = (M, \mu, \mu) \tag{11b}$$

The elastic energy density is:

$$u = u_1 + \frac{u_2 + u_3}{2} \tag{12a}$$

$$u_i = m_i \varepsilon_i^2 \tag{12b}$$

Let E_i^T and χ_i be 6 dimensionless quantities, that are constant in the *whole* multilayer, we show now that the general solution to the problem is:

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$$\sigma_i^q = m_i^q \varepsilon_i^q = m_i^q \left(E_i^T - E_i^q + \chi_i \frac{x_1}{w} \right) \tag{13}$$

We notice at first that the elastic field defined by equation (13) satisfies the conditions of continuity and compatibility everywhere, as well as the conditions of equilibrium in the bulk, at the external surfaces parallel to P, and at the interfaces. It depends on the 6 unknown dimensionless parameters E_i^T and χ_i, which are entirely determined by the following 6 conditions:

a) The Albenga theorem is written:

$$\int_0^w \sigma_i dx_1 = 0 \tag{14}$$

Since the stress depends only on the coordinate x_1 (equation 13), condition 14 also expresses the fact that the total force is nil on the lateral surface.

b) the average moment on the lateral surface must be nil. Hence:

$$\int_0^w \sigma_i x_1 dx_1 = 0 \tag{15}$$

The 6 unknown constants are expressed as linear functions of the E_i^q, by substituting equations (10, 11, 13) into equations (14, 15):

$$E_i^T = \sum_{q=1}^{q=n} A_i^q E_i^q \tag{16a}$$

$$\chi_i = \sum_{q=1}^{q=n} B_i^q E_i^q \tag{16b}$$

The coefficients A_i^q and B_i^q are calculated in the appendix (formulae A-5). They depend on the thickness and elastic modulii of the various plates *and* on their stacking order (formula A-3c).

4.1.2. Curvature

The curvature tensor **K** is defined by [5]:

$$K_{ij} \equiv \frac{\partial \omega_i}{\partial x_j} = \sum_{k,r} \varepsilon_{ikr} \frac{\partial \varepsilon_{jr}}{\partial x_k} = \sum_l \varepsilon_{i1r} \frac{\partial \varepsilon_{jr}}{\partial x_1} \tag{17}$$

ω is the lattice rotation, and ε_{ikl} is the Levi-Civitta symbol, whose non-zero elements are:

$$\varepsilon_{123} = \varepsilon_{231} = \varepsilon_{312} = -\varepsilon_{321} = -\varepsilon_{213} = -\varepsilon_{132} = 1 \tag{18}$$

By substituting equation (13) into equation (17), the non-zero components of the curvature tensor are:

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$$K_{22} = -K_{33} = \frac{-\chi_3}{2w} \qquad K_{23} = \frac{\chi_2 - 2\chi_1}{2w} \qquad K_{32} = \frac{2\chi_1 + \chi_2}{2w} \tag{19}$$

The tilt components K_{23} and K_{32} of the local lattice curvature around Ox_2, and Ox_3 depend on χ_1 and χ_2, while the twist components K_{22} and K_{33} around the same axes depend on χ_3.

4.1.3. Remarks

In elastically isotropic media, there is no coupling between the shear and the dilatation components of the stress-free strain tensor, which yields simple results in the following cases:

1. *All the stress-free strains* $\mathbf{E}^q$ *are pure dilatations*
The non-zero component of the stress free strain and of the stress in lamella q are respectively E_1^q, and σ_1^q. The non-zero components of the curvature tensor are:

$$K_{23} = -K_{32} = -\frac{\chi_1}{w} \tag{20}$$

the elastic strain field is a pure dilatation everywhere, and the resulting rotation is a pure tilt around the Ox_2 and Ox_3 axes.

2. *All the stress-free strains* $\mathbf{E}^q$ *are pure shears*
Their non-zero component of the stress free strain and of the stress in lamella q are respectively E_2^q, E_3^q, σ_2^q, σ_3^q. The elastic strain field is a pure shear, while the induced rotation combines twist and tilt. The situation further simplifies if all the shear vectors are parallel to the coordinate axes. Then their non-zero component is E_3^q, the non-zero component of the stress is σ_3^q and the non zero components of the curvature tensor are:

$$K_{22} = -K_{33} = -\frac{\chi_3}{w} \tag{21}$$

4.2. Approximate solution neglecting the coupling between curvature and stress

A simplified, and approximate, solution is obtained by neglecting the force on the lateral surfaces, *i. e.* by ignoring the condition (15). In order to avoid confusion in the following, quantities related to this solution are upperlined with a ~.
A satisfactory solution to equations (14) is [10, 11]:

$$\tilde{\sigma}_i^q = m_i^q \tilde{\varepsilon}_i^q = m_i^q \left(\tilde{E}_i^T - E_i^q \right) \tag{22a}$$

$$\tilde{\chi}_i = 0 \tag{22b}$$

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This solution is however approximate since equation (15) is not satisfied, that is, the average moment on the lateral faces is not zero. Indeed condition (22b) is obviously in contradiction with the real situation. This issue shall be further discussed in § 5.
Substituting equation 22a in equation (14):

$$\tilde{E}_i^T = \sum_{q=1}^{q=n} \tilde{A}_i^q E_i^q \tag{23a}$$

$$\tilde{A}_i^q = \frac{f_q m_i^q}{\sum_{q=1}^{q=n} f_q m_i^q} \tag{23b}$$

From equations (22) and (23):

- *the state of stress is independent of the stacking order*. For example, the state of stress of a multilayer comprising *p* different crystalline components distributed in *n* slices of various width w^n, is completely determined by the volume fraction f_p of each component, whatever the distribution of the *p* variants in the multilayer [10, 11].
- the smaller the volume fraction of a component, the larger its internal stress.

Notice that $\tilde{E}_i^T$ is the volume average of the E_i^q, while E_i^T combines a volume average on the various stress-free strains E_i^q *and* of the average strain induced by the curvature.
Let us consider now a bilayer made of a substrate *s*, and a film *f* (figure 1a, 2a) whose elastic modulii are m_{is} and m_{if}. E_i^f is nil by construction.
We define r_i:

$$r_i = \frac{m_{is} - m_{if}}{m_{is}} \qquad r_i < 1 \tag{24}$$

For a homogeneous two-phase multilayer, $r_i = 0$.
From equations (23, 24):

$$\left(\tilde{A}_i^s, \tilde{E}_i^T\right) = \frac{1-f}{1-fr_i}\left(1, E_i^s\right) \tag{25}$$

$$\left(\tilde{\sigma}_i^s, \tilde{\sigma}_i^f\right) = (-f, 1-f)\frac{1-r_i}{1-fr_i} m_i^s E_i^s \tag{26}$$

the stress concentrates into the thinnest plate in a bilayer.
The result is the same for a sandwhich *sfs* where the film *f* is matched between two identical layers *s* as indicated in figure 2b, notwhistanding the position of the film. Formulae (25, 26) apply equally for a multilayer *sfsfs...f* made of *n* alternating layers *s* and *f* of the same thickness (figure 2c), with $f = 1/2$, which gives:

$$\left(\tilde{\sigma}_i^s, \tilde{\sigma}_i^f\right) = (-1, 1)\frac{1-r_i}{2-r_i} m_i^s E_i^s \tag{27}$$

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5. Two-phases multilayer

5.1. General equations

In this section, we consider a multilayer made of two components s and f whose elastic modulii are m_{is} and m_{if}. By definition, the stress free strain keeps the same value E_i^s in all the s layers, while it is nil in all the f layers, then formulae 16 can be written:

$$\left(E_i^T, \chi_i\right) = \left(A_i^s, B_i^s\right) E_i^s \tag{28}$$

The general expression of A_i^s and B_i^s can be calculated by standard algebra, in terms of r_i and of 3 numerical coefficients C_1^s, C_2^s, C_3^s, as indicated in § A-1-3 of the appendix. Then the stress is:

$$\sigma_i^s = m_i^s \left(A_i^s - 1 + B_i^s \frac{x_1}{w} \right) E_i^s \tag{29a}$$

$$\sigma_i^f = (1 - r_i) m_i^s \left(A_i^s + B_i^s \frac{x_1}{w} \right) E_i^s \tag{29b}$$

The curvature tensor is defined by equations (19,28).
From equations (12), the total energy per unit surface is:

$$U = U_1 + \frac{U_2 + U_3}{2} \tag{30a}$$

$$U_i = \int_0^w u_i dx_1 \tag{30b}$$

Substituting equations (29):

$$\frac{U_i}{m_i^s E_i^{s2} w} = \left[C_1^s \left(A_i^s - 1\right)^2 + 2 B_i^s \left(A_i^s - 1\right) C_2^s + B_i^{s2} C_3^s + (1 - r_i) \left(A_i^{s2} C_1^b + 2 A_i^s B_i^s C_2^b + B_i^{s2} C_3^b \right) \right] \tag{31}$$

From equations (26, 31):

$$\frac{\tilde{U}_i}{m_i^s E_i^{s2} w} = \left(\frac{1 - r_i}{1 - f r_i} \right)^2 f(1 - f) \tag{32}$$

The above formulae generalize results which have been obtained for the case where the stress-free strain is a pure dilatation [1-3] to any stress-free transformation: dilatation, shear or any combination of both.
Although combining standard algebra the general expressions are tedious, and not prone to simple physical interpretation. Therefore we concentrate in the following to a situation where the multilayer is homogeneous, and the stress-free strain has only one component. Applying formulae (A-9, A-14, 28, 29, 31) to the three homogeneous two phase multilayers defined in figure 2. we have:

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1. *Bilayer (figure 2a)*

$$\sigma_i^s = f\left[2-3f-6(1-f)\frac{x_1}{w}\right]m_i^s E_i^s \quad (33a)$$

$$\sigma_i^f = (1-f)\left[1+3f-6f\frac{x_1}{w}\right]m_i^s E_i^s \quad (33b)$$

$$\chi_i = 6f(1-f)E_i^s \quad (33c)$$

$$U_i = f(1-f)\left(1-3f+3f^2\right)m_i^s E_i^{s\,2} w \quad (33d)$$

2. *Sandwich (figure 2b)*

$$\sigma_i^s = f\left[-4+3f+6f_1+6(1-f-2f_1)\frac{x_1}{w}\right]m_i^s E_i^s \quad (34a)$$

$$\sigma_i^f = \left[(1-f)(1-3f)+6ff_1+6f(1-f-2f_1)\frac{x_1}{w}\right]m_i^s E_i^s \quad (34b)$$

$$\chi_i = 6f(1-f-2f_1)E_i^s \quad (34c)$$

$$U_i = f\left[1-4f+6f^2-3f^3+12ff_1\left(1-f-f_1^2\right)\right]m_i^s E_i^{s\,2} w \quad (34d)$$

3. *Multilayer sfsfs...f (figure 2c)*

$$\sigma_i^s = \left[-\frac{1}{2}+\frac{3}{2n}-\frac{3}{n}\frac{x_1}{w}\right]m_i^s E_i^s \quad (35a)$$

$$\sigma_i^f = \left[\frac{1}{2}+\frac{3}{2n}-\frac{3}{n}\frac{x_1}{w}\right]m_i^s E_i^s \quad (35b)$$

$$\chi_i = -\frac{3}{n}E_i^s \quad (35c)$$

$$U_i = \frac{1}{4}\left(1-\frac{3}{n^2}\right)m_i^s E_i^{s\,2} w \quad (35d)$$

5.2. Discussion

For simplicity sake we restrict at first the discussion to the case where the stress-free strain has only one non zero component and compare the results given by formulae (33-35) to those obtained through the approximate solution (26, 27, 32).

a) *homogeneous bilayer (figure 2a)*.

Figure 3 represents the variation of the stress in a homogeneous bilayer, calculated after formulae (33), as a function of x_1, compared to that calculated from formula (26). The discrepancy is obvious. Notice for example that the stress changes sign in the substrate.

The plot of the ratio $\frac{\tilde{U}_i}{U_i}$ of the average elastic energy stored in the bilayer calculated from formulae (31) and (32) represented figure 4 emphasizes the importance of the stress relaxation due to the curvature. As soon as the volume fraction of the film is larger than a few %, the approximate solution overestimates the stored elastic energy quite significantly. This stems from the fact that the *approximate solution does not take into account* the elastic relaxation

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due to the curvature. This should be taken into account when modelling the growth of epitaxial multilayers used in electronic devices.
Whenever $f << 1$, formula (A-7) shows that the stress in the film, and the curvature can be calculated to a good approximation by using formulae (33b, 33c), even in heterogeneous multilayers.
It has been known for a long time that if the stress free strain is a pure dilatation, the bilayer undergoes bending. The radius of curvature has been calculated by superimposing to the elastic field calculated by equation 27 a bending **G** which cancels that moment. The corresponding bending radius $\tilde{R}$ is [12]:

$$\tilde{R} = -\frac{(1-f)^2 w}{6 f E_i^s} \tag{36}$$

This result has been widely used to determine the stress in the film through the measurement of the curvature of the bilayer [13-15].
In our formulation, a stress-free strain which is a pure dilatation corresponds to $i = 1$ in formulae 33, which corresponds to a bending with a curvature radius R_1:

$$R_1 = -\frac{w}{6 f (1-f) E_i^s} \tag{37}$$

The relative error is of the order of $3f$, which is appreciable except if $f << 0.1$.

b) *Sandwich*

In this case:

$$U_i = f E_i^s w \left\langle \sigma_i^f \right\rangle \tag{38}$$

$\left\langle \sigma_i^f \right\rangle$ is the average stress in the f layer. (Be careful, this equation is valid only for a homogeneous sandwich).
Figure 5a is a plot of the elastic energy of the sandwich as a function of its position f_1. It is seen that the energy is maximum when the f layer is in the midle of the film, which corresponds to zero curvature. Figure 5b is the plot of the ratio h_b of the average energy per unit surface of the approximate solution to that given by equation 34d, as a function of f_1, which shows the same trend.

c) *aba..b* multilayer

Formulae 35 show explicitly that the curvature is a decreasing function of the number n of layers. As a consequence the stress, and the energy are an increasing function of n. When n tends to infinity, the approximate solution (formula 27) is recovered.

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6. The stress-free strain is a pure shear

6.1. The structure of lamellar TiAl

A relevant practical example of this situation is found in nearly stoïchiometric Titanium-Aluminium alloys, which, under appropriate thermal treatments, is made of coherently matched lamellae of the α_2 Ti_3Al and the γ TiAl phases [16, 17].
The lattice of the α_2 phase is DO19, with parameters $c_\alpha = 0.462$nm, and $a_\alpha = 0.577$nm, in the stoïchiometric composition. The lattice of the γ phase is centred tetragonal (figure 6a), with parameters $c_\gamma = 0.406$nm, and $a_\gamma = 0.400$nm, in the stoïchiometric composition. It is a stacking *ABCABC*, or *CBACBA* of {111}planes. The {111} planes are equivalent but the various <110> directions are not: the row $[1\bar{1}0]$ is composed of one kind (Ti or Al) of atom, the rows $[\bar{1}01]$ and $[0\bar{1}1]$ contain both kinds of atoms (figure 6b). The relation between the α_2 and γ lamellae is $\{111\}[110]_\gamma \| \{0001\}[11\bar{2}0]_{\alpha_2}$.

There are 6 variants, hence 6 ways to perform lattice match between a given α_2 and a γ lamella (Figure 7a). The stacking *ABCABC* (resp *CBACBA*) gives 3 variants obtained by successive rotations of $2\pi/3$, which we denote γ_1, γ_2, γ_3 (resp γ'_1, γ'_2, γ'_3) (Figure 7b). There is no mismatch between the twinned lamellae γ_i, γ'_i.

$$c_\gamma = a_\gamma(1+\alpha) \qquad a_{\alpha 2} = a_\gamma(1+\alpha')\sqrt{2} \tag{39}$$

Typical values are $\alpha = 1.5\ 10^{-2}$, and $\alpha' = 2\ 10^{-2}$. They vary slightly with composition as the parameters of both phases do.
The transformation strain between a γ_i and a γ_j is a pure shear (figure 7c). Since the terms of second order in α can be neglected, denoting $\mathbf{B}^{i/j}$ the transformation strain for a film γ_i on a substrate γ_j, we have very generally:

$$\mathbf{B}^{i/j} + \mathbf{B}^{j/i} = 0 \tag{40a}$$

$$\mathbf{B}^{i/j} + \mathbf{B}^{j/k} + \mathbf{B}^{k/i} = 0 \tag{40b}$$

It is not our purpose in this paper to give a full discussion of the various microstructures related to lamellar TiAl, and we shall restrict at first to the so called polysynthetically twinned (PST) crystals which exhibit a single lamellar orientation over the entire crystal [18].

6.2. General equations for a γ_3/γ_2 bilayer

Using the reference frame of figure 7a, we obtain, from formulae (3), and (39):

$$\mathbf{B}^{3/2} = \alpha \begin{pmatrix} 0 & \frac{2}{\sqrt{3}} \\ 0 & 0 \end{pmatrix} \qquad \mathbf{E}^{3/2} = \frac{\alpha}{\sqrt{3}} \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} \tag{41}$$

Therefore:

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$$E_3^s = \alpha \frac{\sqrt{3}}{3} \tag{42}$$

In the approximation of elastic isotropy, the elastic modulii are identical; formulae 33 apply with i =3. The curvature is a pure torsion with:

$$K_{22} = K_{33} = \chi_3 \tag{43}$$

6.3. Discussion

It is well known, that volume fractions of the various γ phases larger than 20% are currently observed. Besides the mismatch is rather large, of the order of 10^{-2}. The relaxation due to the stress-curvature coupling should be very important. Unfortunately, there are no precise experimental observations of free-standing bilayers.

The transposition to γ multilayers is obvious, although one must consider the 3 different types of matching (γ_1/γ_2, γ_2/γ_3, γ_3/γ_1).

Since the γ lamellae develop from the disordered α_2 phase, a complete description should in addition include an analysis of the various α_2/γ_i bilayers. There is no difficulty in writing the corresponding equations. The only difference is the existence of a dilatation component. Since in the observed multilayers the volume fraction of the α_2 phase is rather small, of the order of a few %, one observes a concentration of the compression stress in this phase. The curvature has both tilt and twist components.

One important consequence is that it is not *a priori* correct, to consider that the elastic state of a multilayer is defined by the volume fraction of each component, whatever the size and order distribution of the layers. This imposes constraints on the order of growth of the γ_i lamellae on one α_2 lamella.

The situation is more complex in polycrystalline fully lamellar TiAl. The observation of low angle grain boundaries in as grown crystals is very likely the result of the coupling between the stress and the curvature. The experimental observations on lamellar TiAl should be revisited taking into account this coupling.

7. Concluding remarks

We have shown that the elastic state of a coherent multilayer consists in a coupled stress (or elastic strain), curvature field, and we have developped a general method to calculate these fields. The method applies to every stress-free transformations. The importance of the stress relaxation due to the curvature has been emphasized on specific examples. The calculations have been developped under the approximation of isotropic elasticity, but they can easily be generalized to elastically anisotropic multilayers, as shown in appendix 2.

The stress-curvature coupling should be taken into account when considering the coherency stress relaxation by dislocations nucleation in multilayers, as well as the growth mechanisms of the latter.

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Appendix

A1. Calculation of E_i^T and c_i for a multilayer

A-1-1 General definitions

Figure 1c describes the geometry of a multilayer of total thickness w, made of n lamellae of thickness w^q, lamella q being defined as the domain corresponding to:

$$W^{q-1} \le x_1 \le W^q \qquad \text{(A-1)}$$

$$W^q = \sum_{r=1}^{r=q} w^r \qquad \text{(A-2)}$$

Here q and r are superscripts, not exponents.
Let the dimensionless quantities C_p^q, and the composite modulii M_{ip}, be defined by:

$$C_p^q = \frac{1}{(w)^p} \int_{W^{q-1}}^{W^q} x_1^{p-1} dx_1 = \frac{1}{p(w)^p} \left[\left(W^q\right)^p - \left(W^{q-1}\right)^p \right] \qquad p=1,2,3,... \qquad \text{(A-3a)}$$

$$M_{ip} = \frac{1}{(w)^p} \sum_{q=1}^{q=n} m_i^q \int_{W^{q-1}}^{W^q} x_1^{p-1} dx_1 = \sum_{q=1}^{q=n} C_p^q m_i^q \qquad \text{(A-3b)}$$

$$M_{i1} = \frac{1}{w} \sum_{q=1}^{q=n} m_i^q w^q \qquad \text{(A-3c)}$$

Notice that M_{ip} depends on the stacking order for $p>1$.

A-1-2 General multilayer

Substituting equations (13) into equations (14, 15), and using formulae (A-3), one obtains:

$$M_{i1} E_i^T + M_{i2} \chi_i = \sum_{q=1}^{q=n} C_1^q m_i^q E_i^q \qquad i=1,2,3 \qquad \text{(A-4a)}$$

$$M_{i2} E_i^T + M_{i3} \chi_i = \sum_{q=1}^{q=n} C_2^q m_i^q E_i^q \qquad \text{(A-4b)}$$

Solving equations A-4 gives equations (16), with:

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$$A_i^q = \frac{\left(C_1^q M_{i3} - C_2^q M_{i2}\right) m_i^q}{M_{i1} M_{i3} - \left(M_{i2}\right)^2} \quad \text{(A-5a)}$$

$$B_i^q = \frac{\left(C_2^q M_{i1} - C_1^q M_{i2}\right) m_i^q}{M_{i1} M_{i3} - \left(M_{i2}\right)^2} \quad \text{(A-5b)}$$

A-1-3 Two phased multilayer

Let C_p^s (resp C_p^f) be the sums defined by formulae (3a) restricted to the layers *s* (resp *f*), we have very generally:

$$C_p^s + C_p^f = \frac{1}{p} \quad \text{(A-6)}$$

From formula (A3b):

$$M_{ip} = \left(\frac{1}{p} - r_i C_p^f\right) m_{is} \quad \text{(A-7)}$$

r_i is defined by equation (24).

Let us define:

$$A_i^s = \frac{\left(C_1^s M_{i3} - C_2^s M_{i2}\right) m_i^s}{M_{i1} M_{i3} - \left(M_{i2}\right)^2} \quad \text{(A-8a)}$$

$$B_i^s = \frac{\left(C_2^s M_{i1} - C_1^s M_{i2}\right) m_i^s}{M_{i1} M_{i3} - \left(M_{i2}\right)^2} \quad \text{(A-8b)}$$

A_i^s and B_i^s are calculated by substituting equations (A-6,A-7), in equations (A-8). Let us just notice that A_i^s and B_i^s are completely defined by the knowledge of r_i, C_1^s, C_2^s, C_3^s. The general expression is however tedious and of little use.

Since E_i^f is nil, equations (16) can be written:

$$E_i^T = A_i^s E_i^s \quad \text{(A-9a)}$$

$$\chi_i = B_i^s E_i^s \quad \text{(A-9b)}$$

We now calculate C_1^s, C_2^s, C_3^s for the multilayers represented figure 2:

a) bilayer *sf* of figure 2a:

$$C_p^s = \frac{(1-f)^p}{p} \quad \text{(A-10)}$$

b) For the sandwich *sfs* (figure 2b):

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$$C_p^s = \frac{1+f_1^p-(f+f_1)^p}{p} \qquad \text{(A-11)}$$

c) For the alternate *sps..p* multilayer (figure 2c):

$$C_p^s = \frac{1}{np}\left[\sum_{h=1}^{h=n/2}(2h-1)^p - \sum_{h=1}^{h=-1+n/2}(2h)^p\right] \qquad \text{(A-12a)}$$

$$\left(C_1^s, C_2^s, C_3^s\right) = \left(\frac{1}{2}, \frac{n-1}{4n}, \frac{2n-1}{24n}\right) \qquad \text{(A-12b)}$$

Finally, if both phases have the same elastic modulii m_i^s (homogeneous two phases multilayer), from equation (A-6), r_i is nil, and:

$$M_{ip} = \frac{m_{is}}{p} \qquad \text{(A-13)}$$

Then:

$$A_i^s = 2\left(2C_1^s - 3C_2^s\right) \qquad \text{(A-14a)}$$

$$B_i^s = 6\left(2C_2^s - C_1^s\right) \qquad \text{(A-14b)}$$

A-2 Equations for a bilayer taking into account anisotropy

Let $\mathbf{C}^s$ (resp $\mathbf{C}^f$) and $\mathbf{S}^s$ (res $\mathbf{S}^s$) be the elastic modulii and elastic compliances tensor of the substrate (resp the film) in the axes of figure 1a. Using the matrix representation, we may write Hooke's law:

$$\sigma_m = \sum_{n=1}^{n=6} C_{mn}\gamma_n \qquad \text{(A-15a)}$$

$$\gamma_n = \varepsilon_n \qquad n = 1, 2, 3 \qquad \text{(A-15b)}$$

$$\gamma_n = 2\varepsilon_n \qquad n = 4, 5, 6 \qquad \text{(A-15c)}$$

With these notations equation (9), is written:

$$\sigma_1^r = \sigma_5^r = \sigma_6^r = 0 \qquad \text{(A-16)}$$

Here *r* stands for *s* or *f*.

By substituting equations (A-15a) in equations (A-16), one may express $\gamma_1^r, \gamma_5^r, \gamma_6^r$, and $\sigma_2^r, \sigma_3^r, \sigma_4^r$ as linear functions of $\gamma_2^r, \gamma_3^r, \gamma_4^r$, which we write:

$$\sigma_m^r = Q_{m2}^r\gamma_2^r + Q_{m3}^r\gamma_3^r + Q_{m4}^r\gamma_4^r \qquad m = 2, 3, 4 \qquad \text{(A-17)}$$

We define:

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$$\left(\Gamma_2^q, \Gamma_3^q, \Gamma_4^q\right) = \left(E_2^q, E_3^q, 2E_4^q\right) \qquad \text{(A-18)}$$

Here q stands for s, f, T.
Therefore the solution can be written as:

$$\gamma_n^s = \Gamma_n^T - \Gamma_n^s + \chi_n \frac{x_1}{w} \qquad n = 2, 3, 4 \qquad \text{(A-19a)}$$

$$\gamma_n^f = \Gamma_n^T + \chi_n \frac{x_1}{w} \qquad n = 2, 3, 4 \qquad \text{(A-19b)}$$

The 6 dimensionless constants (Γ_n^T, χ_n) are determined by substituting equations A-19 into equations A-17 and solving equations 14, and 15.

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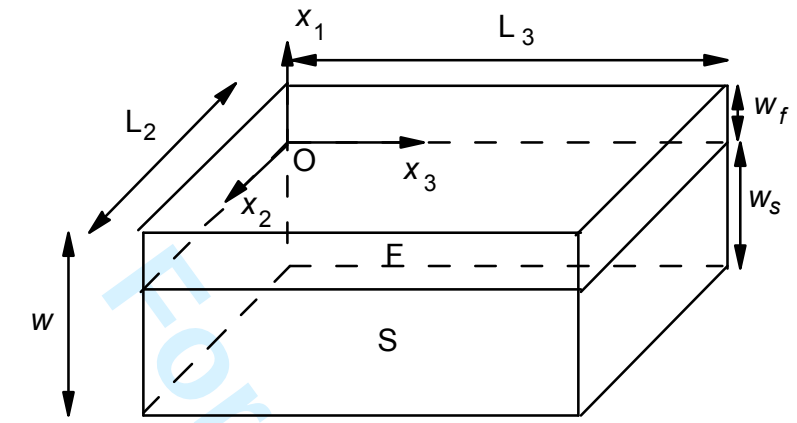
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Figures Captions

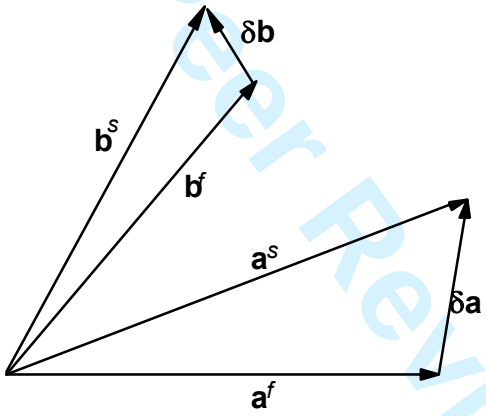
1. Coherently matched multilayers.
   a. Bilayer made of a substrate S and a film F. The axis Ox_1 is normal to the interface. The coordinate axes Ox_2, Ox_3, are in the interface plane.
   b. The lattice of the substrate is defined by the two vectors ($\mathbf{a}^s$, $\mathbf{b}^s$); the lattice of the film by the two vectors ($\mathbf{a}^f$, $\mathbf{b}^f$).
   c. Multilayer made of 5 layers of thickness w^q, q=1,...,5.
2. Three examples of two phase multilayer
   a. Bilayer *sf*.
   b. Sandwich *sfs*.
   c. *sfsfsf* multilayer (n = 6).
3. Plot of the variation of $\dfrac{\sigma_i^s}{m_{is}E_i^s}$ as a function of x_1/w for f=0.1, and $m_{if} = m_{is}$. Full line: general solution. Dotted line: approximate solution.
4. Plot of the ratio $h_a = \dfrac{\tilde{U}_i}{U_i}$ of the stored energy in a bilayer as a function of the volume fraction f.
5. a. Plot of U_i as a function of f_1 for f=0.1.
   b. Plot of the ratio $h_b = \dfrac{\tilde{U}_i}{\tilde{U}_i}$ as a function of f_1 for f=0.1.
6. a. Tetragonal TiAl
   b. Representation of the triangle $A_1A_2A_3$.
   $$a_1 = a_2 \approx a_\gamma\sqrt{2}\left(1+\frac{\alpha}{2}\right) \qquad a_3 = a_\gamma\sqrt{2}$$
7. a. Left:the elementary triangle in the basal plane of the α_2 lamella.
   Right: the reference axes.
   b. Projection of the (111) plane of the three possible arrangements γ_1, γ_2, γ_3 of tetragonal TiAl.
   c. The transformation strain for γ_3/γ_2, is a pure shear.

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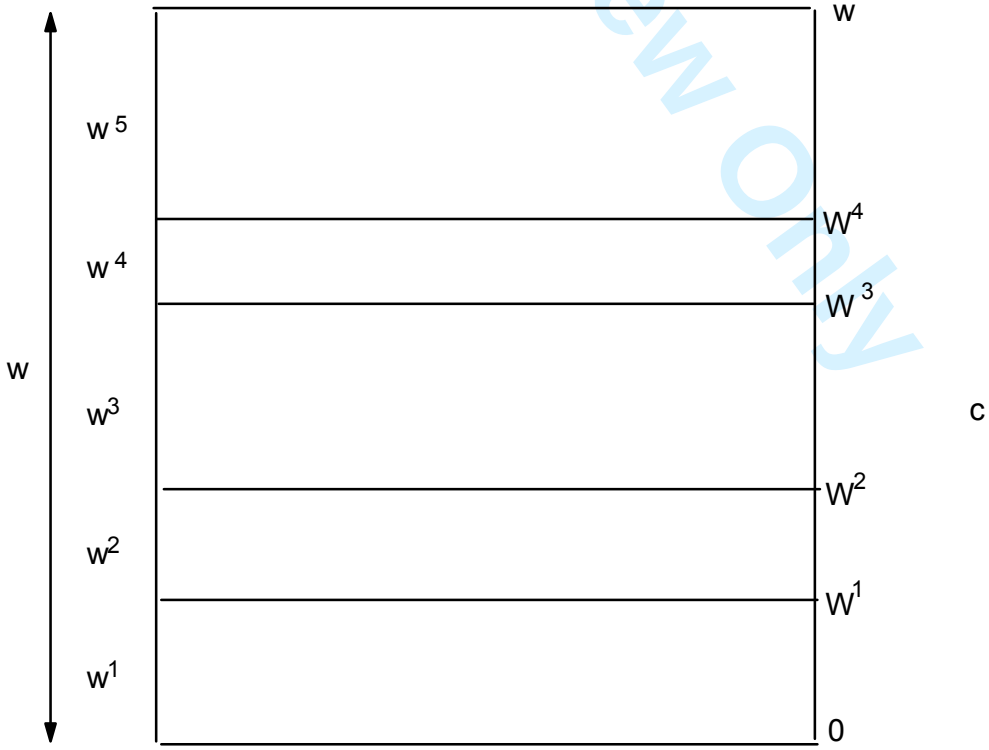
Figure 1



a



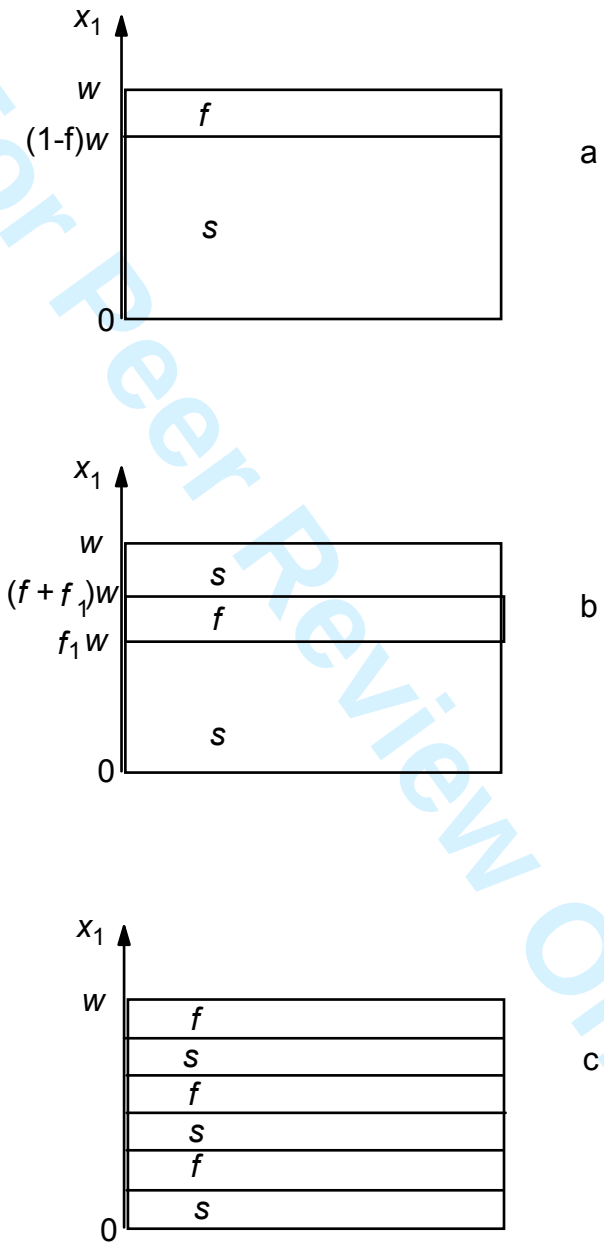
b



c

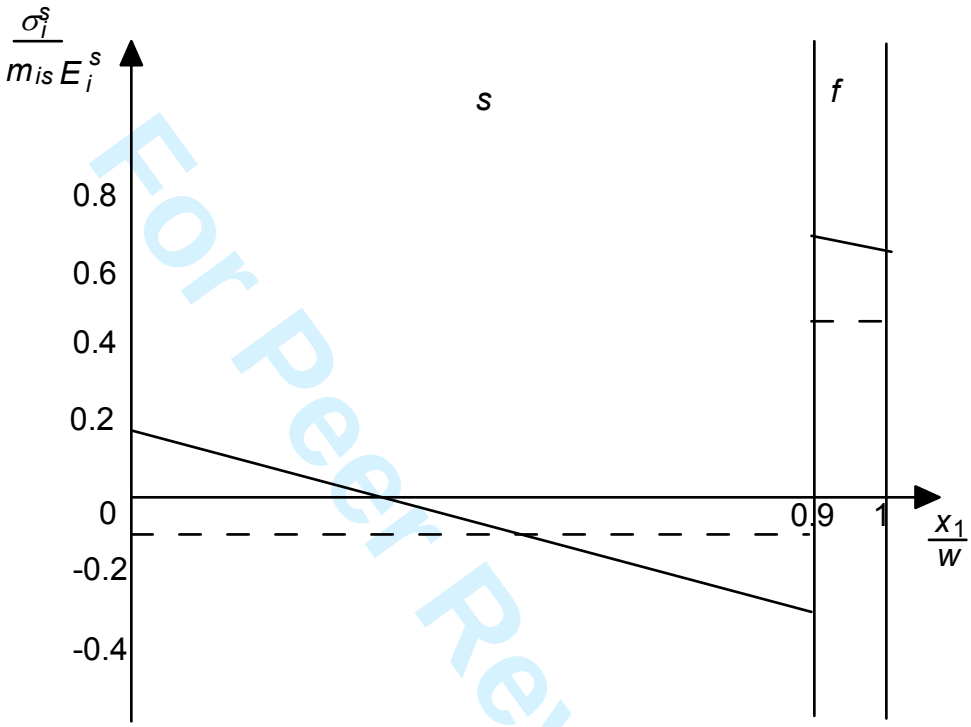
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Figure 2



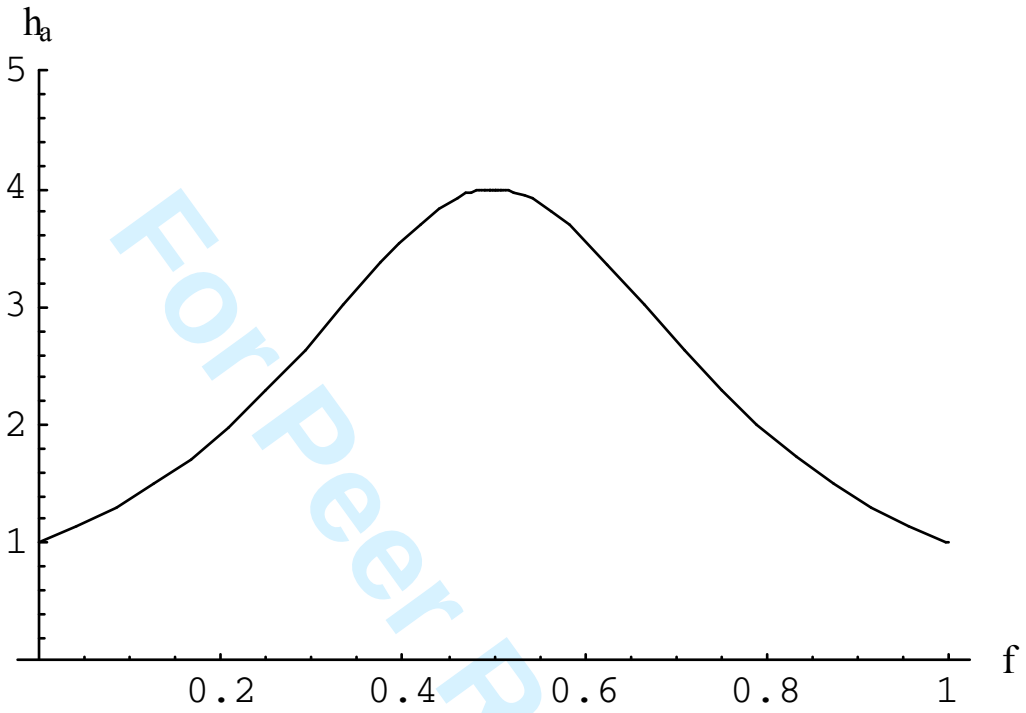
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Figure 3



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Figure 4



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Figure 5a

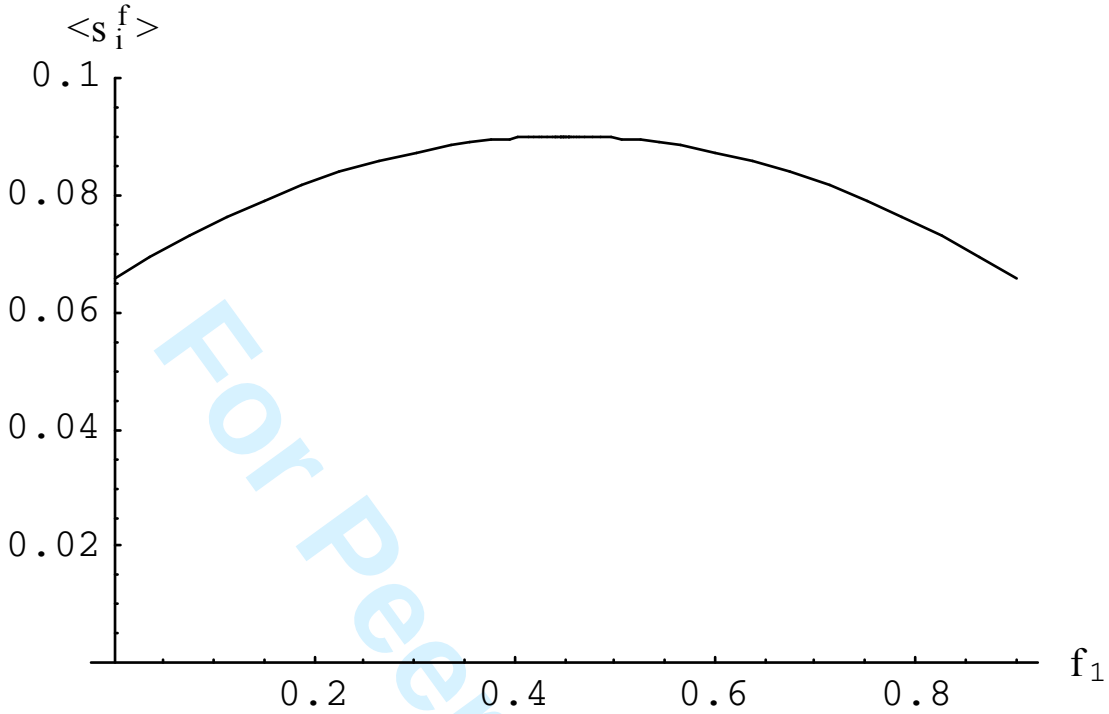
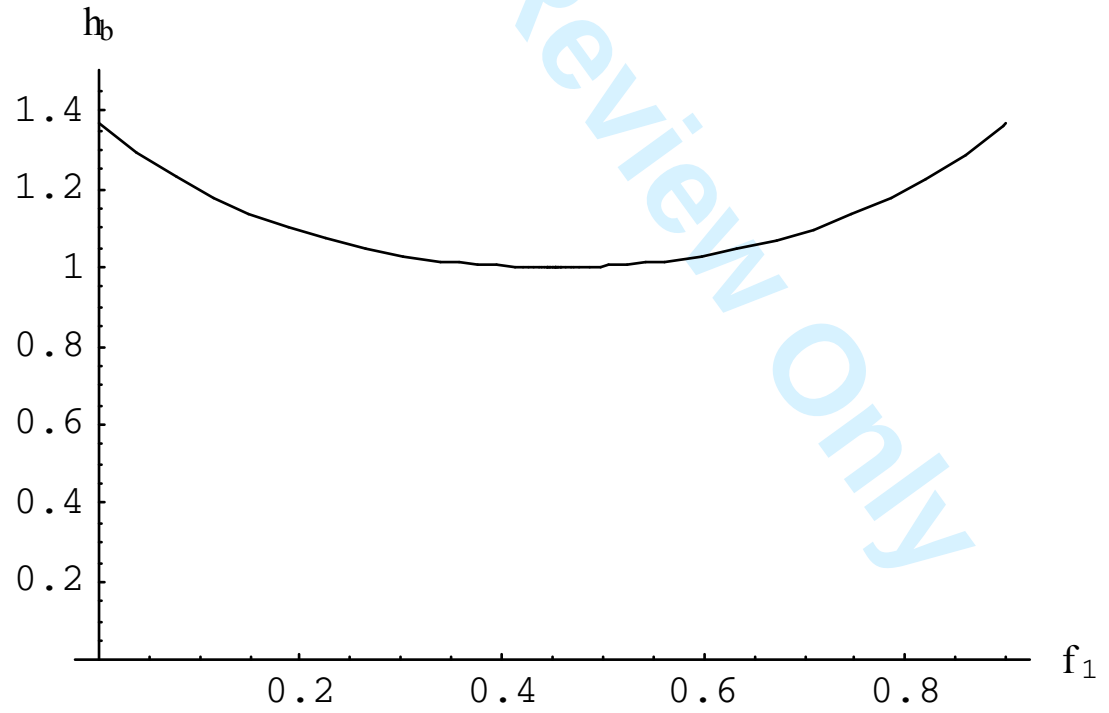
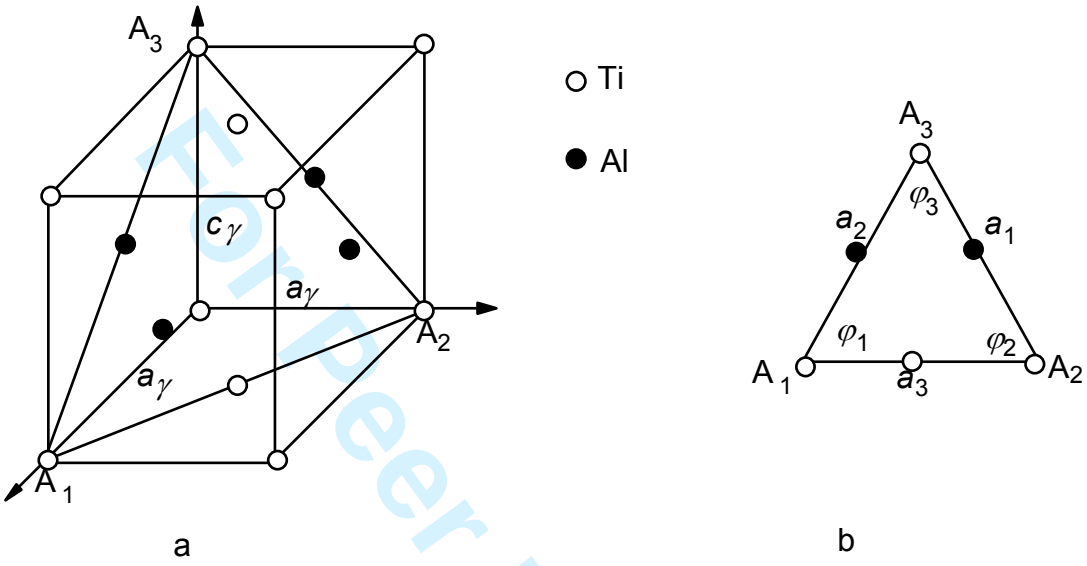


Figure 5b



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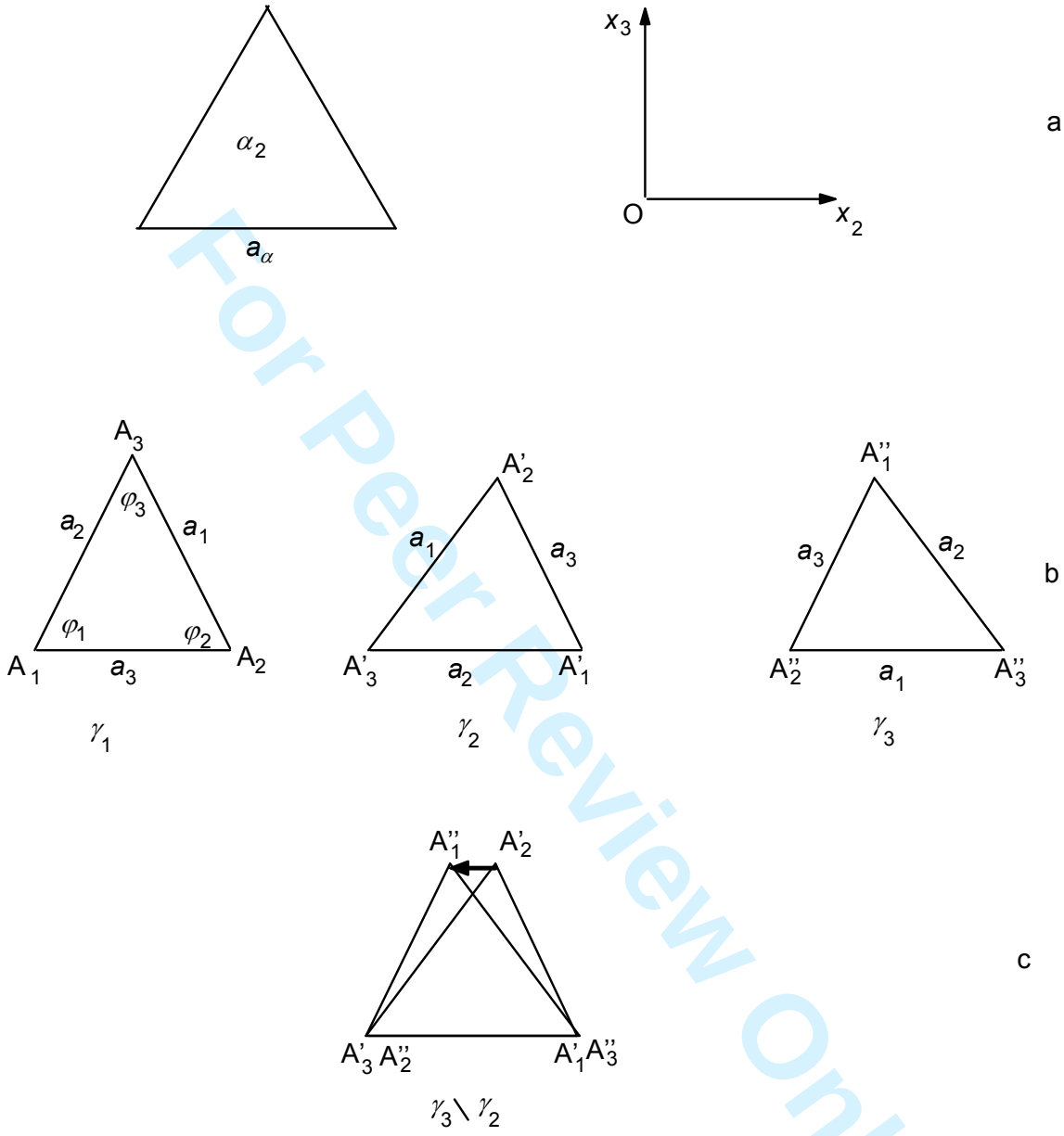
Figure 6



a b

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Figure 7



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