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Dislocation dipoles in nanoscale films with compositional inhomogeneities

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ABSTRACT

A theoretical model of misfit dislocations and their dipole configurations in nanoscale films with compositional inhomogeneities is suggested. Energy characteristics of misfit dislocation dipoles are calculated. The critical film thickness is estimated which characterizes the energetically favourable generation of misfit dislocation dipoles in films with compositional inhomogeneities. The results of the suggested model account for experimental data obtained by Wang *et al.* on observation of dislocation dipoles in nanoscale $Ga_{0.5}In_{0.5}P$ films.

§ 1. INTRODUCTION

Nanoscale films have become increasingly important in both fundamental and applied research because of their outstanding functional properties widely exploited in many contemporary high technologies (for example, Edelstain and Camarata (1996), Siegel *et al.* (1997), Nalwa (1999), Chow *et al.* (2000), Gleiter (2000), Ovid'ko (2000) and Roco *et al.* (2000)). The stability of both the structure and the properties of nanoscale films, which is crucial for their application, is strongly influenced by misfit stresses occurring owing to a misfit between crystal lattice parameters of films and substrates. In particular, relaxation of misfit stresses in single-crystal and nanocrystalline films effectively occurs via generation of misfit dislocations (MDs) (for example, Jain *et al.* (1990, 1997), Rocket and Kiely (1991), Atkinson and Jain (1992), Gosling *et al.* (1992, 1993), Gosling and Willis (1994), Ovid'ko (1999, 2000, 2001), Gutkin *et al.* (2000), Hirth (2000), Mahajan (2000) and Spaepen (2000)), which are capable of giving rise to degradation of the functional properties of films. Thus, the formation of MD rows at film–substrate boundaries (the standard micromechanism for relaxation of misfit stresses in single-crystal films) leads to degradation of coherency and associated functional characteristics of film–substrate boundaries (Jain *et al.* 1990, 1997, Mahajan 2000). Theoretical examinations in the area discussed commonly deal with MDs in compositionally homogeneous films. Recently, however, much attention has been attracted to nanoscale films with compositional inhomogeneities (Rich *et al.* 1997, Ipatova *et al.* 1998, Ledentsov *et al.* 1998, Mattila *et al.* 1999, Brunner *et al.* 2000). (This interest is motivated, in particular, by the possibility of fabricating semiconductor quantum

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wires in compositionally inhomogeneous films.) Such films exhibit the specific structural peculiarities capable of giving rise to the specific behaviour of MDs, different from that in conventional, compositionally homogeneous films. Thus, recently, MD dipoles (MD configurations of a new type) have been experimentally observed in nanoscale $Ga_{0.5}In_{0.5}P$ films with compositional inhomogeneities in the situation where the mean crystal lattice parameter of the film is the same as the crystal lattice parameter of the substrate (Wang *et al.* 2000). Wang *et al.* have observed dislocation dipoles of 60° and 90° type (as well as individual Lomer dislocations) lying in the interphase layer 3.5 nm thick of the $Ga_{0.5}In_{0.5}P$ film. Their appearance has been attributed to the existence of spatial inhomogeneities of the film composition. The main aim of this paper is to elaborate a theoretical model which describes MDs and their dipole configurations in films with periodically modulated compositional inhomogeneities. Special attention will be paid to consideration of the critical film thickness that characterizes the energetically favourable generation of MD dipoles in compositionally inhomogeneous films.

§ 2. Films with compositional inhomogeneities: model

In the general case of a film with spatial inhomogeneities of the chemical composition, critical parameters that characterize the generation of MDs are expected to depend crucially on both the misfit between crystal lattice parameters of the adjacent phases and the character of spatial inhomogeneities of the film composition. For definiteness, in this paper we shall confine our consideration to the situation where the mean lattice parameter of the film is equal to the lattice parameter of the substrate (this corresponds to the conditions of experimental observation (Wang *et al.* 2000) of MD dipoles in nanoscale $Ga_{0.5}In_{0.5}P$ films), and the film composition is periodically modulated. In the situation discussed, alternate regions with tensile and compressed stresses are formed in the film. Relaxation of misfit associated with the formation of the alternate regions with tensile and compressive stresses can effectively occur via the generation of MD dipoles (figure 1). In particular, such a MD dipole can be formed as a result of the generation of two dislocation semiloops with opposite Burgers vectors at the film free surface and their consequent expansion (figure 2).

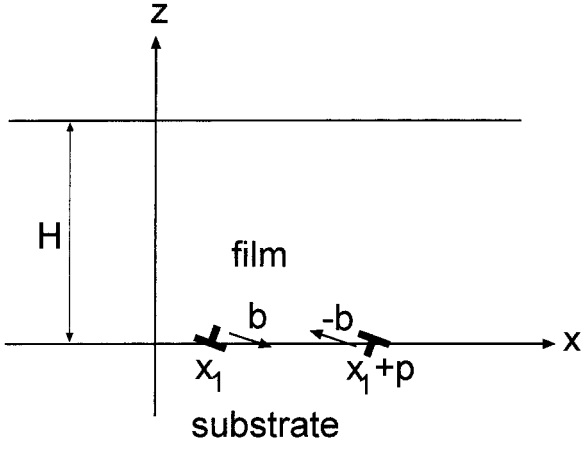

Figure 1. Dipole of MDs at interphase (film–substrate) boundary.

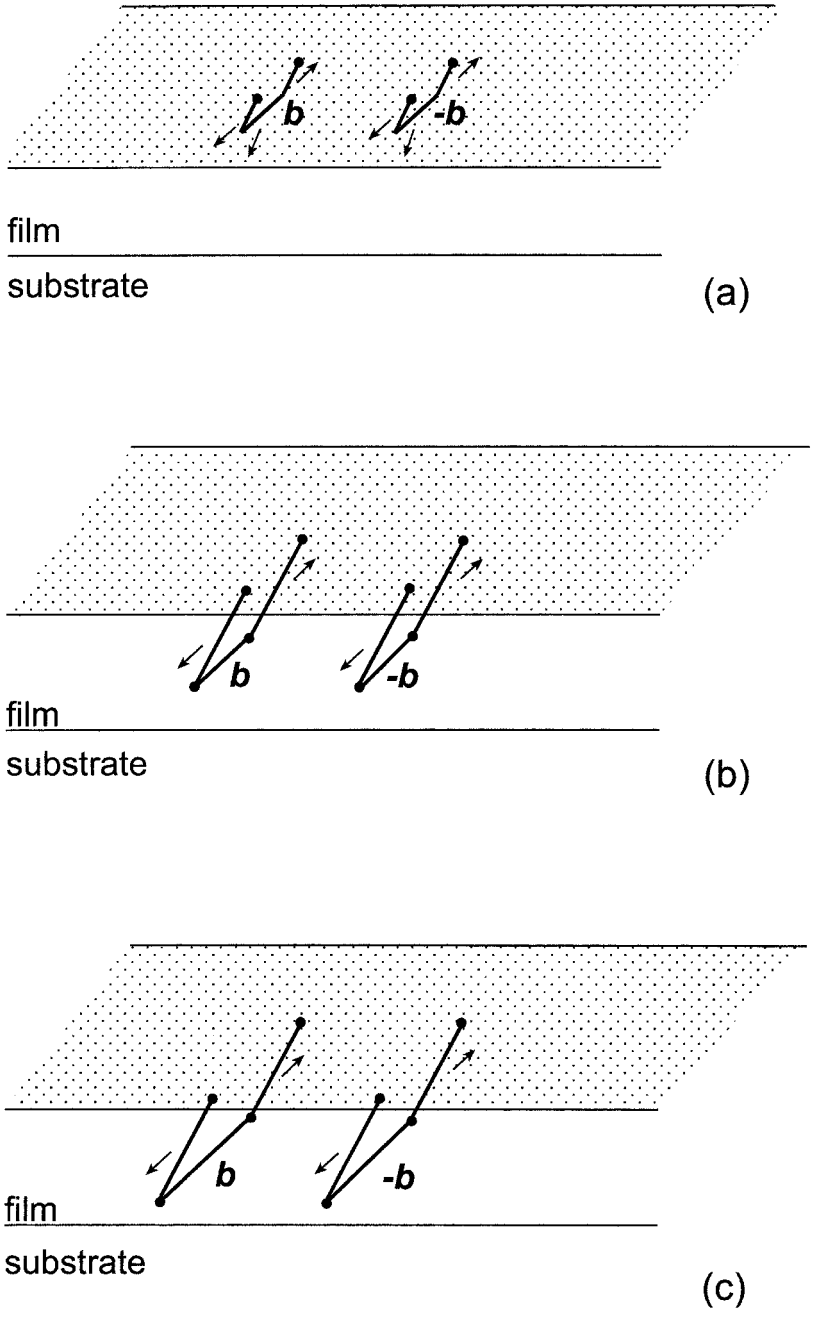

Figure 2. Generation and evolution of semiloops of dislocations with opposite Burgers vectors **b** and $-\mathbf{b}$. Dislocation semiloops are shown as solid semiloops. Arrows indicate the directions of expansion of dislocation semiloops. (*a*) Generation of dislocation semiloops at the free surface of the film. (*b*) Horizontal dislocation semiloop segments (parallel with the free surface of the film shown as a dotted surface) reach the film–substrate boundary where they are stopped. (*c*) Lateral dislocation semiloop segments expand, resulting in elongation of horizontal semiloop segments that form a MD dipole.

In order to calculate the energy characteristics of isolated MDs and MD dipoles in films with compositional inhomogeneities, in this paper we shall use representations of the model by Glas (1987) that describes compositional inhomogeneities in polyatomic films that are free of MDs. As with the Glas model, the following assumptions will be used in our consideration of MDs in compositionally inhomogeneous films.

(i) Both the thin film and the thick substrate have cubic crystal lattices and are isotropic solids characterized by the same values of the shear modulus G and the same values of Poisson's ratio ν.
(ii) The lattice parameter of the substrate coincides with the mean lattice parameter of the film.
(iii) Spatial modulation of the film composition does not vary along the direction normal to the film–substrate boundary.
(iv) The film lattice parameter depends on the cosine of the spatial coordinate x along the film–substrate boundary. More precisely, the modulation of the film lattice parameter a (which corresponds to modulation of the film composition) along x is described as

$$a = a_0[1 - \varepsilon_0 \cos(\alpha x)], \tag{1}$$

where a_0, ε_0 and α are the constant parameters.

With the assumptions (i)–(iv), following Glas (1987), the period that characterizes spatial modulation of the film can be derived from the condition that the elastic energy of the film/substrate system is minimized. In the next two sections, this modulation period will be used in our theoretical analysis of the conditions at which the generation of MD dipoles is energetically favourable in compositionally inhomogeneous films.

§ 3. Energy characteristics of misfit dislocation dipoles in films with compositional inhomogeneities

Let us consider an isolated dipole of edge MDs with Burgers vectors $\mathbf{b}$ and $-\mathbf{b}$ in the model heteroepitaxial system under the assumptions (i)–(iv). The MDs are located at the interphase (film–substrate) boundary and the distance between them is p (figure 1). For definiteness, in the following we shall consider dipoles of MDs of the 60° and 90° types, which have been experimentally observed in $Ga_{0.5}In_{0.5}P$ films (Wang *et al.* 2000).

Let us analyse the conditions in which the generation of MD dipole (figure 1) is energetically favourable. To do this, we shall compare energy characteristics of the two physical states realized in the heteroepitaxial system, namely the state with the MD-free interphase boundary and the state with the interphase boundary containing the MD dipole. The heteroepitaxial system in the MD-free (coherent) state is characterized by the total elastic energy which is equal to the misfit strain energy that arises from the mismatch at the interphase boundary. The misfit strain energy density (per unit volume) is given as follows (Glas 1987):

$$w^0 = 2\pi D(1+\nu)\varepsilon_0^2\left(1 - \frac{(1+\nu)[1-\exp(\alpha H)]^2}{\alpha H}\right), \tag{2}$$

where $D = G/2\pi(1-\nu)$ and H is the film thickness.

When the MD dipole (figure 1) is generated at the interphase boundary in the heteroepitaxial system, its total energy W (per unit of MD length) consists of four terms:

$$W = W^{\mathrm{f}} + W^{\mathrm{dip}} + W^{\mathrm{dip\text{–}f}} + 2W^{\mathrm{c}}, \tag{3}$$

where W^{f} denotes the misfit strain energy related to misfitting at the interphase boundary, W^{dip} the proper elastic energy of the MD dipole, W^{c} the energy of the MD core, and $W^{\mathrm{dip\text{–}f}}$ the elastic energy associated with the elastic interaction between the MDs and the misfit stresses. The generation of the MD dipole (figure 1) is energetically favourable if it leads to a decrease in the total energy, that is if $W - W^{\mathrm{f}} < 0$. From equation (3), we arrive at the following criterion for the generation of the MD dipole to be energetically favourable:

$$W^{\mathrm{dip}} + W^{\mathrm{dip\text{–}f}} + 2W^{\mathrm{c}} < 0. \tag{4}$$

In order to calculate the ranges of values of heteroepitaxial system parameters in which inequality (3) is valid, in the following we shall calculate the misfit stresses and terms W^{dip}, W^{c}, and $W^{\mathrm{dip\text{–}f}}$.

The energy $W^{\mathrm{dip\text{–}f}}$ that characterizes the interaction between the MD dipole (figure 1) and misfit stresses generated at the interphase boundary is given in its general form by the following formula (Mura 1968):

$$W^{\mathrm{dip\text{–}f}} = -b_x \int_0^H [\sigma^{\mathrm{f}}_{xx}(x_1, z) - \sigma^{\mathrm{f}}_{xx}(x_1 + p, z)]\,\mathrm{d}z - b_z \int_0^H [\sigma^{\mathrm{f}}_{xz}(x_1, z) - \sigma^{\mathrm{f}}_{xz}(x_1 + p, z)]\,\mathrm{d}z. \tag{5}$$

Here x_1 and $x_1 + p$ are the coordinates of the MDs with Burgers vectors $\mathbf{b}$ and $-\mathbf{b}$ respectively; $\sigma^{\mathrm{f}}_{xx}(x, z)$ and $\sigma^{\mathrm{f}}_{xz}(x, z)$ denote the components of the misfit stress tensor in the film with compositional inhomogeneities characterized by the spatial dependence (1) of the film lattice parameter.

The misfit stresses $\sigma^{\mathrm{f}}_{xx}(x, z)$ and $\sigma^{\mathrm{f}}_{xz}(x, z)$ can be derived from formulae (Glas 1987) for the corresponding displacements. In doing this, we obtain

$$\sigma^{\mathrm{f}}_{xx}(x, z) = C \cos(\alpha x) \exp(\alpha z)\,[S_1 + (2 + \alpha z)S_2 + S_3], \tag{6}$$

$$\sigma^{\mathrm{f}}_{xz}(x, z) = C \cos(\alpha x) \exp(\alpha z)\,[S_1 + (1 + \alpha z)S_2 - S_4], \tag{7}$$

where $C = 4\pi(1 + \nu)D\varepsilon_0$ and

$$S_1 = \tfrac{1}{2}[-(1 + 2\alpha H)\exp(-2\alpha H) + 2(1 + \alpha H)\exp(-\alpha H) + 1], \tag{8}$$

$$S_2 = \exp(-\alpha H)\,[\exp(-\alpha H) - 1], \tag{9}$$

$$S_3 = \tfrac{1}{2}[\exp(-2\alpha z) - 1], \tag{10}$$

$$S_4 = \tfrac{1}{2}[\exp(-2\alpha z) + 1]. \tag{11}$$

With equations (6)–(11) substituted into equation (5), we have

$$W^{\mathrm{dip\text{–}f}} = -\frac{C\{\cos(\alpha x_1) - \cos[\alpha(x_1 + p)]\}}{2\alpha} \exp(-2\alpha H)\,[\exp(\alpha H) - 1] \times \{b_x[1 + \exp(\alpha H) - 2\alpha H] + b_z[-1 + \exp(\alpha H) - 2\alpha H]\}. \tag{12}$$

In the following, for definiteness, we shall consider 'equilibrium MD dipoles', that is dipoles with parameter x_1 corresponding to minimum of the interaction energy $W^{\text{dip–f}}$. In this situation, as follows from equation (12), the parameter x_1 satisfies the condition $\alpha x_1 = \pi/2 - \alpha p/2$. With the above taken into account, the interaction energy $W^{\text{dip–f}}$ is represented as follows:

$$W^{\text{dip–f}} = -4\pi(1+\nu)Db_xH\sin\left(\frac{\alpha p}{2}\right)f_e, \tag{13}$$

where

$$f_e = \frac{\exp(-2\alpha H)[\exp(\alpha H)-1]\{1+\exp(\alpha H)-2\alpha H+(b_z/b_x)[-1+\exp(\alpha H)-2\alpha H]\}}{\alpha H}. \tag{14}$$

Now let us consider the terms W^{c} and W^{dip} on the left-hand side of inequality (4), the criterion for the formation of the MD dipole to be energetically favourable. The energy W_{c} of the MD core is about $Db^2/2$ (Hirth and Lothe 1982). The proper elastic energy W^{dip} of the MD dipole (per unit MD length) shown in figure 1 can be derived from the stress function of an edge dislocation located near a free surface. In turn, this stress function can be derived from the stress function of an edge dislocation in a cylinder (A. E. Romanov 1992, private communication) in the limit with the cylinder radius being infinite. In doing this, we have

$$W^{\text{dip}} = \frac{D}{2}\left\{(b_x^2+b_z^2)\left[2\ln\left(\frac{2H}{b}\right)-\ln\left(\frac{4H^2+p^2}{p^2}\right)-1\right] + \frac{4H^2[b_z^2(12H^2+p^2)-b_x^2(4H^2+3p^2)]}{(4H^2+p^2)^2}\right\}. \tag{15}$$

Equations (13) and (15) and the estimate of $W^{\text{c}} \approx Db^2/2$ will be used in the next section to estimate the critical film thickness that characterizes the energetically favourable generation of MD dipoles in compositionally inhomogeneous films.

§4. Critical thickness of films with compositional inhomogeneities

In the context of this paper, the critical film thickness H_{c}' is defined as follows. The formation of the MD dipole is energetically favourable (or unfavourable respectively) when the thickness of the film with compositional inhomogeneities $H > H_{\text{c}}'$ (or $H < H_{\text{c}}'$ respectively). The equation for the critical thickness H_{c}' is derived from the condition $W^{\text{dip}} + W^{\text{dip–f}} + 2W^{\text{c}} = 0$. From this condition, the estimate of $W^{\text{c}} \approx Db^2/2$ and equations (13) and (15) we find the following equation for the critical thickness H_{c}':

$$4\pi(1+\nu)f_e = \frac{1}{2b_xH_{\text{c}}'\sin(\pi p/T)}\left\{(b_x^2+b_z^2)\left[2\ln\left(\frac{2H_{\text{c}}'}{b}\right)-\ln\left(\frac{4H_{\text{c}}'^2+p^2}{p^2}\right)+1\right] + \frac{4H_{\text{c}}'^2[b_z^2(12H_{\text{c}}'^2+p^2)-b_x^2(4H_{\text{c}}'^2+3p^2)]}{(4H_{\text{c}}'^2+p^2)^2}\right\}. \tag{16}$$

Here T $(=2\pi/\alpha)$ is the period of spatial modulation of the film lattice parameter. Analysis of equation (16) shows that the critical thickness H_{c}' is minimal at $p = T/2$.

The equation for the critical thickness H_{c} that characterizes the energetically favourable generation of an isolated MD in a compositionally modulated film can

be found in the same way as equation (16) for H_c'. In doing this, after some algebra, we have

$$4\pi(1+\nu)f_e = \frac{b_x^2 + b_z^2}{b_x H_c}\left[\ln\left(\frac{2H_c}{b}\right) + \frac{1}{2}\right]. \tag{17}$$

The dependences of the critical thicknesses H_c and H_c', which characterize isolated MDs (of 90° and 60° types) and their dipoles respectively, on parameter $4\pi(1+\nu)\varepsilon_0$ are shown in figure 3 for $p = T/2$ and $\alpha H = 1.256$. (This value of αH minimizes the misfit strain energy W^f (Glas 1987).) As follows from figure 3, the critical thicknesses H_c and H_c' given by equations (17) and (16) respectively decrease with increasing modulation amplitude ε_0.

In figure 4, the critical thicknesses H_c and H_c' are plotted against the normalized modulation frequency αH. As seen in figure 4, for small enough αH, the critical

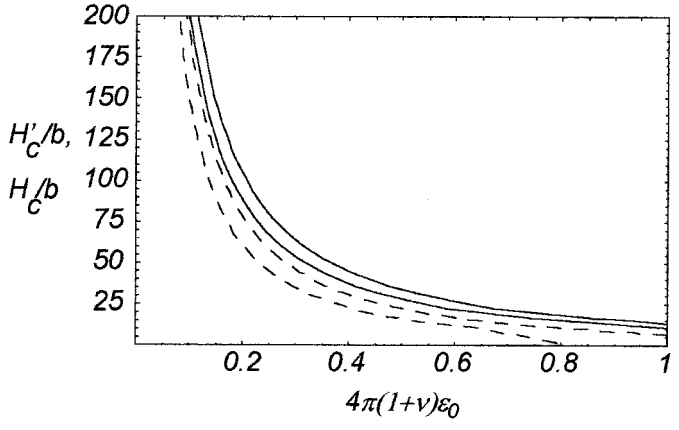

Figure 3. Dependences of the critical thickness H_c/b for the generation of 60° and 90° dislocations (upper and lower solid curves respectively) and the critical thickness H_c'/b for the formation of 60° and 90° dislocation dipoles (upper and lower broken curves respectively) on the modulation magnitude ε_0, for $\alpha H = 1.256$.

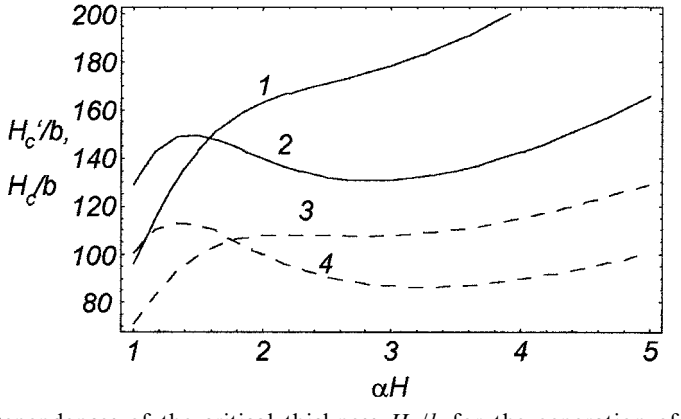

Figure 4. Dependences of the critical thickness H_c/b for the generation of 90° and 60° dislocations (solid curves 1 and 2 respectively) and the critical thickness H_c'/b for the formation of 90° and 60° dislocation dipoles (broken curves 3 and 4 respectively) on the parameter αH, for $4\pi(1+\nu)\varepsilon_0 = 0.15$.

thicknesses H_c and H_c' for the generation of 90° dislocations and dislocation dipoles are smaller than those for the formation of 60° dislocations and dislocation dipoles. With increasing αH, however, the critical thicknesses for the nucleation of dislocations and dislocation dipoles of 90° type become greater than those for the formation of 60° dislocation and dislocation dipoles. The most important conclusion which follows from figures 3 and 4 is that, for any modulation magnitude ε_0 and frequency α, the critical thickness H_c which characterizes isolated MDs in films with periodically modulated chemical composition is larger than the critical thickness H_c' specifying the generation of the MD dipoles in such films. Thus, in compositionally inhomogeneous films, the generation of the MD dipoles becomes energetically favourable at smaller film thicknesses than the formation of isolated MDs. This conclusion is in agreement with experimental data (Wang *et al.* 2000) on observation of dislocation dipoles in nanoscale $Ga_{0.5}In_{0.5}P$ films with compositional inhomogeneities.

§ 5. Concluding remarks

Here we have suggested a first approximation model of the experimentally observed (Wang *et al.* 2000) MD dipoles (MD configurations of a new type) in nanoscale films with compositional inhomogeneities. In the framework of the suggested model, the critical thickness H_c' is estimated, which characterizes the formation of MD dipoles in compositionally inhomogeneous films. With this estimate, it has been theoretically revealed that the generation of MD dipoles is more energetically favourable in compositionally inhomogeneous films than that of isolated MDs. It is contrasted with the case of conventional, compositionally homogeneous films. (Generally speaking, MD dipoles can exist in conventional capped films (Jain *et al.* 1993). In these circumstances, however, two MDs composing a dipole configuration are located at various interphase boundaries, namely at the boundary between the capped film and the substrate and the boundary between the capped film and the upper layer (Jain *et al.* 1993). This case is essentially different from that described in our model.) Also, it has been demonstrated that the formation of MD dipole configurations is energetically favourable in compositionally inhomogeneous films even in the situation where the mean film lattice parameter coincides with the substrate lattice parameter.

Thus, according to our theoretical analysis, MD dipoles are general structural elements of compositionally inhomogeneous films, effectively contributing to accommodation of misfit stresses in such films. This specific feature of compositionally inhomogeneous films, described theoretically in this paper and confirmed by experimental data (Wang *et al.* 2000), should be definitely taken into account in further fundamental and applied research of such films. In particular, degradation of the structure and the functional properties of compositionally inhomogeneous films should be associated with the new mechanism (the formation of MD dipoles) of misfit stress relaxation, which is more effective than the standard mechanism (the formation of isolated MDs) playing the dominant role in conventional, compositionally homogeneous films.

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