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MISFIT DISLOCATION WALLS IN MULTILAYERED FILMS

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Abstract. We suggest a new mechanism for misfit strain accommodation in misfitting multilayer films with alternate layers through the generation of misfit dislocation walls. The detailed analysis of the necessary conditions for their appearance shows that, similar to one-layer films, misfit dislocation walls form above some critical film thicknesses which can be increased by varying the relation between thicknesses of adjoining film layers.

1. INTRODUCTION

Solid films and composite multilayer coatings are widely used in contemporary high technologies due to their unique electronic and optical properties (e. g., [1-3]). The properties of films and coatings strongly depend on the elastic strains induced by the misfit of crystal lattices of different layers and may degrade in the presence of defects. In single crystalline films, the most effective and wide-spread pathway for the accommodation of such misfit strains lies in the generation of misfit dislocations (MDs) at or near interfaces [4-7]. For thin one-layer films, MDs form commonly two-dimensional ensembles in the interface plane. In this case, if the substrate is much thicker than the film, the critical film thickness above which MDs are likely to nucleate depends on the misfit parameter only. For multilayer films on similar substrates, the system energy and the critical thickness depend also on the relations between the crystal lattice misfits and layer thicknesses. The extra degrees of freedom connected with the existence of different film layers lead to the opportunity of the formation of MD configurations which are not found in one-layer films. In particular, in multilayer systems MDs can be arranged into the arrays of dislocation dipoles, with dislocations of opposite sign located at different interface boundaries. This situation has been extensively studied for the case of capped films (which are sandwiched between two substrate layers) (e. g., [7-11]). In recent work [12], another situation has been analyzed for the film consisting of alternate layers where MDs are situated either at the film/ substrate interface or at the interface nearest to the film/substrate one.

In the present paper, we will focus on the generation of the walls of complete edge MDs at the interfaces of the film composed of alternate layers. The rotational mechanisms of misfit strain accommodation realized through the formation of disclinations or dislocation walls in the case of one-layer film have already been observed experimentally [13-18] and analyzed theoretically [17-22]. It has been demonstrated that misfit disclinations [13-22] or MD walls [19-22] can be generated at twin or grain boundaries, in single crystalline films on amorphous substrates and in nano- or polycrystalline films. In multilayer films, dislocation walls can be formed by slip of dislocations from their sources. In the following, we will find the conditions for the formation of a first (individual) MD wall. For simplicity, we will suppose that dislocations composed the wall, are regularly spaced and separated from each other by a pair of alternate film layers.

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2. MISFIT DISLOCATION WALL IN MULTILAYER FILM

Consider a system that consists of a semi-infinite substrate and a multilayer film of thickness H comprised of N pairs of alternate layers α and β of thickness h_1 and h_2, respectively [$H = N(h_1 + h_2)$] (Fig. 1). The film and the substrate are supposed to be elastically isotropic solids and have the same shear modulus G and Poisson's ratio ν. The misfit of the substrate, layers α and layers β crystal lattices is assumed to be dilatational and characterized by the misfit parameters $f_1 = (a_s - a_\alpha)/a_s$ and $f_2 = (a_s - a_\beta)/a_s$ equal in the absence of MDs to the elastic strain within layers α and β, respectively. In the latter relation, a_s, a_α and a_β denote the crystal lattice parameters of the substrate, layers α and layers β. Let the misfit parameters f_1 and f_2 be positive, and the lines and Burgers vectors of the generated MDs be parallel to the film/substrate interface (Fig. 1).

To determine the necessary conditions for the formation of an MD wall, we will use the standard technique comparing the energies of the system with and without the wall. The energy W of the system with an MD wall extending from the film/substrate interface to the free surface (Fig. 1a) per unit length of dislocations can be presented as the sum of three terms:

$$W = W^f + W^w + W^{f-w}, \qquad (1)$$

where W^f is the misfit strain energy, W^w the self-energy of the MD wall and $W^{f\text{-}w}$ the interaction energy of the misfit strain and the MD wall. When the MD wall is absent, the energy of the system is related to the misfit strain only and equals to W^f. The necessary condition for the MD wall generation is that the system energy W with the MD wall be smaller than the system energy W^f prior to the MD wall introduction, $\Delta W = W - W^f < 0$, which yields

$$W^w + W^{f-w} < 0. \qquad (2)$$

The energies W^w and $W^{f\text{-}w}$ appearing in (2) will be calculated in the next section.

3. ENERGY OF MISFIT DISLOCATION WALL

The self-energy W^w of the MD wall shown in Fig. 1a can be written as

$$W^w = \sum_{i=1}^{N}\left[W_i^d + \sum_{j=1}^{i-1} W_{i,j}^{d-d}\right] + NW^c, \qquad (3)$$

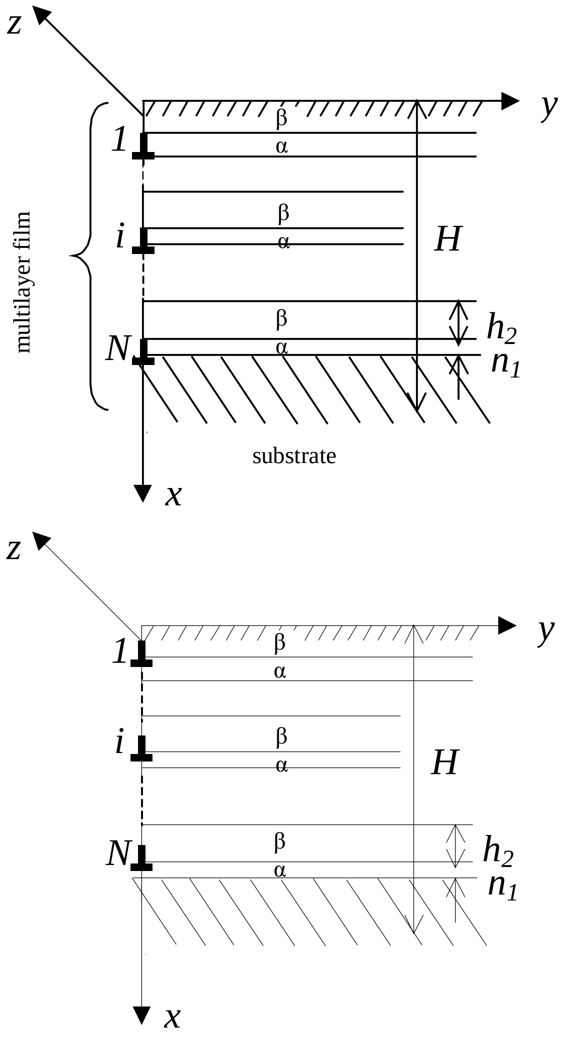

Fig. 1. Misfit dislocation walls in a multilayer film.

where W_i^d is the self-energy of the i-th MD (MDs are enumerated sequentially from 1, for the MD nearest to the free surface, to N for the MD at the film/substrate interface), $W_{i,j}^{d-d}$ is the interaction energy of the i-th and j-th dislocations, and W^c is the dislocation core energy. The self-energies W_i^d of MDs are given by [23]

$$W_i^d = \frac{Db^2}{2}\left(\ln\frac{2d_i}{r_0} - \frac{1}{2}\right), \qquad (4)$$

where b is the value of the dislocation Burgers vectors, $D = G/[2\pi(1-\nu)]$, $d_i = iH/N$ and r_0 is the cut-off radius of dislocation elastic fields on its core; we put $r_0 = b$. The energy $W_{i,j}^{d-d}$ follows from [24] as

$$W_{i,j}^{d-d} = -b\int_{r_0}^{\infty}\sigma_{xy}^{d}(x = d_j - d_i, y, d_i)\,dy, \qquad (5)$$

where $\sigma^{d}_{xy}(x,y,d_i)$ is the stress component of the i-th dislocation lying at a distance d_i from the free surface. The expression for $\sigma^{d}_{xy}(x,y,d_i)$ can be adapted from [25]:

$$\sigma^{d}_{xy}(x,y,d_i) = Db\left\{-\frac{y}{r_-^2}+\frac{2y(x-d_i)^2}{r_-^4}+\frac{y}{r_+^2}+\frac{2y(d_i^2+4d_ix+x^2)}{r_+^4}+\frac{16d_iyx(x+d_i)^2}{r_+^6}\right\}, \tag{6}$$

where $r^2_\pm = (x \pm d)^2 + y^2$. It follows from (5), (6) and the condition $H/N >> b$ that

$$W^{d-d}_{i,j} = Db^2\left(\ln\frac{d_i+d_j}{|d_i-d_j|}-\frac{2d_id_j}{(d_i+d_j)^2}\right). \tag{7}$$

The dislocation core energy is approximated [25] as $W^c \approx Db^2/2$.

Combining (3), (4) and (7), one obtains:

$$W^w = \frac{Db^2}{2}\left\{\sum_{i=1}^{N}\left[\ln\frac{2d_i}{b}+2\sum_{j=1}^{i-1}\left(\ln\frac{d_i+d_j}{d_i-d_j}-\frac{2d_id_j}{(d_i+d_j)^2}\right)\right]+\frac{N}{2}\right]. \tag{8}$$

The energy $W^{f\text{-}w}$ of the interaction of the MD wall with the misfit stress field is given by

$$W^{f-w} = -b\sum_{i=1}^{N}\int_0^{d_i}\sigma^{f}_{yy}(x)dx, \tag{9}$$

where $\sigma^{f}_{yy}(x)$ is the misfit stress tensor component, which is given by [4, 26]

$$\sigma^{f}_{yy} = 4\pi(1+\nu)D\sum_{k=0}^{N-1}\left\{f_1[\Theta(x-x^{(2k+1)})-\Theta(x-x^{(2k+2)})]+f_2[\Theta(x-x^{(2k)})-\Theta(x-x^{(2k+1)})]\right\}, \tag{10}$$

where $\Theta(t)$ is the Heaviside function, $\Theta(t) = 1$ for $t > 0$ and $\Theta(t) = 0$ for $t < 0$; $x^{(2i+1)} = iH/N + h_2$, $x^{(2i)} = iH/N$, $i = 0,...,N$. Substitution of (10) to (9) gives

$$W^{f-w} = -2\pi(1+\nu)(N+1)DbHf_e, \tag{11}$$

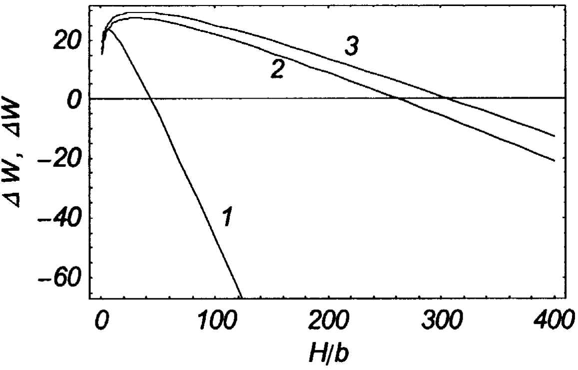

Fig. 2. Dependences of energy variations ΔW (curve 1) and $\Delta\widetilde{W}$ (curves 2 and 3) (in units of $Db^2/2$ due to misfit dislocation walls on film thickness H/b. Curve 2 is drawn for $4\pi(1+\nu)f_1 = 0.1$ and $h_2/h_1 = 1$, while curve 3 corresponds to $4\pi(1+\nu)f_1 = 0$ and $h_2/h_1 = 2$.

where the effective misfit $f_e = (f_1h_1 + f_2h_2)/(h_1 + h_2)$ is introduced. The energy change ΔW due to the MD wall shown in Fig. 1a is now given by the sum of (8) and (11). Taking into account (8) and (11), necessary condition (2) for the generation of the MD wall can be rewritten as

$$\frac{b}{(N+1)H}\left\{\sum_{i=1}^{N}\left[\ln\frac{2d_i}{b}+2\sum_{j=1}^{i-1}\left(\ln\frac{d_i+d_j}{d_i-d_j}-\frac{2d_id_j}{(d_i+d_j)^2}\right)\right]+\frac{N}{2}\right\}<4\pi(1+\nu)f_e. \tag{12}$$

The energy change $\Delta\widetilde{W} = \widetilde{W} - W^f$ due to the generation of the MD wall shown in Fig. 1b, which is defined as the difference between the system energy $\widetilde{W}$ with such an MD wall and the system energy W^f without MD walls, is derived similarly to the expression for the $\Delta\widetilde{W}$. The final result for $\Delta\widetilde{W}$ is

$$\Delta\widetilde{W} = \frac{Db^2}{2}\left\{\sum_{i=1}^{N}\left[\ln\frac{2\tilde{d}_i}{b}+2\sum_{j=1}^{i-1}\left(\ln\frac{\tilde{d}_i+\tilde{d}_j}{\tilde{d}_i-\tilde{d}_j}-\frac{2\tilde{d}_i\tilde{d}_j}{(\tilde{d}_i+\tilde{d}_j)^2}\right)\right]+\frac{N}{2}-4\pi(1+\nu)\frac{H}{b}\left[(N+1)f_e-\frac{2f_1h_1}{h_1+h_2}\right]\right\}, \tag{13}$$

where $\tilde{d}_i = iH/N - h_1$. From (13) and the condition $\Delta\widetilde{W} < 0$ it follows that

$$\frac{b}{(N+1)H}\left\{\sum_{i=1}^{N}\left[\ln\frac{2\tilde{d}_i}{b}+2\sum_{j=1}^{i-1}\left(\ln\frac{\tilde{d}_i+\tilde{d}_j}{\tilde{d}_i-\tilde{d}_j}-\frac{2\tilde{d}_i\tilde{d}_j}{(\tilde{d}_i+\tilde{d}_j)^2}\right)\right]+\frac{N}{2}\right\}< 4\pi(1+\nu)\left(1-\frac{2f_1h_1}{(N+1)(f_1h_1+f_2h_2)}\right)f_e. \tag{14}$$

The plots ΔW and $\Delta\widetilde{W}$ in units of $Db^2/2$ against H/b are shown in Fig. 2 for the case $h_2/h_1 = 2$, $N =$

5 and $4\pi(1+\nu)f_e = 0.15$. Curve 1 corresponds to the dependence of ΔW on H/b, while curves 2 and 3 display the dependences of $\Delta\tilde{W}$ on H/b for the following two cases: $4\pi(1+\nu)f_1 = 0.1$ and $h_2/h_1 = 1$ (curve 2); and $4\pi(1+\nu)f_1 = 0$ and $h_2/h_1 = 2$ (curve 3). One can conclude that the MD walls can nucleate in the film above some critical thicknesses H_c and $\tilde{H}_c$ corresponding to zero values of ΔW and $\Delta\tilde{W}$, respectively. As follows from Fig. 2, if f_1 and f_2 as well as h_1 and h_2 are of the same order, ΔW is lower than $\Delta\tilde{W}$ and $H_c < \tilde{H}_c$ that could easy be predicted. For given f_e, N and H, $\Delta\tilde{W}$ and $\tilde{H}_c$ decrease with increasing f_1 or decreasing the ratio h_2/h_1. At $H < H_c$ the MD walls shown both in Fig. 1a and in Fig. 1b (hereafter referred to as MD walls of first type and MD walls of second type, respectively) do not form. At $H_c < H < \tilde{H}_c$ the generation of an MD wall of first type is possible. If $H > \tilde{H}_c$, MD walls of both first and second type are favoured.

Fig. 3 illustrates the plots of the critical thicknesses H_c (curves 1 and 2) and $\tilde{H}_c$ (curves 1' and 2') against $4\pi(1+\nu)f_e$ for the case $4\pi(1+\nu)f_1 = 0.1$ and $h_2/h_1 = 1$. The plots of H_c and $\tilde{H}_c$ (curves 1 and 1', respectively) shown for $N = 5$ lie below the corresponding plots (curves 2 and 2') drawn for the case $N = 10$. It is seen in Fig. 3 that both H_c and $\tilde{H}_c$ increase with N. This result may seem paradoxical but it becomes clear if one takes in mind that within our model, the number of MDs in the wall increases automatically with N, thus giving rise to the energy (3). In fact, (3) contains terms which vary linearly with N (dislocation self strain energies and core energies) as well as the interaction energy term which is proportional to N^2. On the other hand, the energy (11) of elastic interaction between MD walls and misfit stress varies linearly with N. As a result, the positive energy term (3) grows faster than negative one (11) as N increases, and their balance needs larger H_c (or $\tilde{H}_c$).

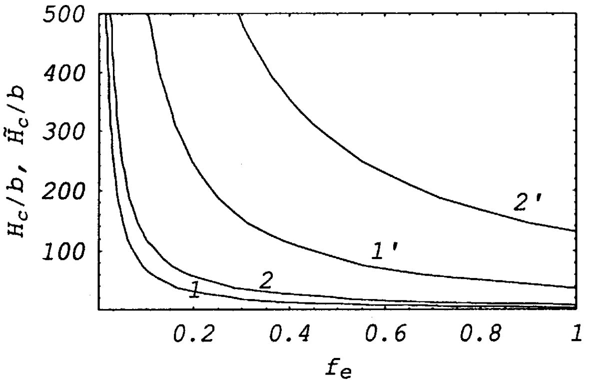

Fig. 3. The critical thicknesses H_c (curves 1 and 2) and $\tilde{H}_c$ (curves 1' and 2') as functions of $4\pi(1+\nu)f_e$ shown for $N = 5$ (curves 1 and 1') and $N = 10$ (curves 2 and 2').

4. CONCLUDING REMARKS

In this paper, we have determined the generation conditions for misfit dislocation walls in the multilayer film with alternate layers. Within our model, the film and the substrate are isotropic solids with equal elastic moduli, G and ν, and two dilatational misfits, f_1 and f_2, of adjoining crystal lattices. Misfit dislocations composed the walls have been spaced regularly with a separation of two film layers.

The principal results of our work are as follows:

(*i*) misfit dislocation walls in a film with alternate layers can form in some range of parameters including film thickness H, misfits f_1 and f_2, the number N of film layers and the ratio h_2/h_1 of adjoining layer thicknesses;

(*ii*) for given f_1, f_2, N and h_2/h_1, misfit dislocation walls can be generated above some critical thicknesses;

(*iii*) the critical film thicknesses increase with the number N of film layers;

(*iv*) for large enough number N of film layers, the critical thicknesses depend mainly on the effective misfit $f_e = (f_1 h_1 + f_2 h_2)/(h_1 + h_2)$ which can be varied (in particular, decreased) by varying the ratio h_2/h_1 of layer thicknesses.

The results of this paper are important for technological applications of multilayer film/substrate composites. In particular, they can be used for estimating the structural and functional stability of real multilayer single crystal film/substrate composites as well as for describing possible accommodation processes in multilayer films with nanocrystalline layers of alternate composition obtained by thermal spray synthesis [27-29].

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