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Faceted Grain Boundaries in Polycrystalline Films

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Abstract—A theoretical model is proposed for describing a new mechanism of misfit-stress relaxation in polycrystalline films, namely, the formation of faceted grain boundaries whose facets are asymmetric tilt boundaries. The ranges of parameters (the film thickness, misfit parameter, angle between facets) in which the nucleation of faceted grain boundaries is energetically favorable are calculated. The nucleation of faceted grain boundaries is shown to be facilitated as the film thickness increases. © *2003 MAIK "Nauka/Interperiodica".*

1. INTRODUCTION

Polycrystalline films are objects of intensive fundamental and applied research, as they are widely applied in modern high-technological production processes. The stability of the physical properties of such films is of primary importance for their technological application and depends significantly on the defects and stress fields in them (see, e.g., reviews [1–5] and papers [6, 7]). For example, a difference between the lattice parameters in substrate and film materials causes internal stresses in the films. These misfit stresses significantly affect the evolution of the structure and the functional properties of the films. When the thickness of a film reaches a certain critical value, misfit stresses, as a rule, are partially accommodated into misfit dislocations at the substrate–film interface [1–15]. However, the presence of grain boundaries in polycrystalline and nanocrystalline films causes alternative efficient mechanisms of relaxation of misfit stresses (and in general, residual stresses of a different nature) via grain-boundary dislocations and disclinations [16–19]. In analyzing these alternative mechanisms, particular attention has been given to the theoretical description of grain-boundary defects in symmetric planar tilt boundaries. In general, however, films also contain asymmetric and faceted grain boundaries [20]. In this work, we propose a theoretical model of a new misfit-stress relaxation mechanism which can operate in polycrystalline films and is related to the formation of faceted grain boundaries whose facets are asymmetric tilt boundaries.

2. FACETED GRAIN BOUNDARIES IN FILMS: MODEL

Let us consider a model of the film–substrate system consisting of a bicrystal film of thickness H and a semi-infinite substrate (Fig. 1). The film and substrate are assumed to be elastically isotropic solids with the same shear modulus G and Poisson ratio ν. The film–substrate interface is characterized by the one-dimensional misfit parameter

$$f = \frac{2(a_f - a_s)}{a_f + a_s}, \tag{1}$$

where a_f and a_s are the lattice parameters of the film and substrate, respectively.

In this work, we analyze two physical states of the film, namely, the state with a straight-line symmetric tilt boundary (Fig. 1a) and the state with a faceted boundary whose facets are asymmetric tilt boundaries (Fig. 1b). For the sake of definiteness, we study low-angle tilt boundaries simulated as ensembles of edge lattice dislocations. However, the results of our consideration can be generalized to the case of a high-angle grain boundary simulated as a grain boundary having dislocations with Burgers vectors of the complete coincidence lattice of the boundary [20]. The plane of the

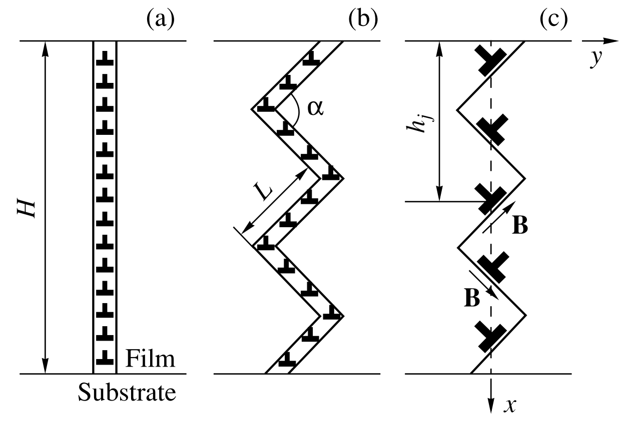

Fig. 1. Dislocation structure of grain boundaries in a bicrystal film: (a) symmetric plane tilt boundary, (b) faceted grain boundary, and (c) faceted grain boundary modeled as a wall of superdislocations having Burgers vectors with alternating directions (schematic).

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symmetric tilt boundary is assumed to be normal to the free surface (Fig. 1a). Such a boundary contains M periodically ordered edge dislocations with a Burgers vector $\mathbf{b}$ parallel to the interface and normal to the plane of the tilt boundary. The misorientation θ of the low-angle symmetric tilt boundary is connected with dislocation parameters by the Frank formula $b = 2(H/M)\sin(\theta/2)$ [20].

A faceted grain boundary consists of facets (segments) having the dislocation structure of an asymmetric tilt boundary (Fig. 1b). For simplicity, we consider facets of only two types, having the same structure and length L; the angle between adjacent facets is α, and their number is N. Like a symmetric boundary, a faceted boundary contains M edge dislocations with a Burgers vector $\mathbf{b}$ that is parallel to the film–substrate interface. Therefore, the orientations of the crystal lattices far from a faceted boundary are identical to those in the case of a symmetric tilt boundary. Since the Burgers vectors $\mathbf{b}$ of dislocations making up a faceted grain boundary are not normal to the facet planes, each facet is an asymmetric tilt boundary.

A symmetric tilt boundary and a faceted boundary in the film differ in the spatial arrangement of the dislocation ensemble. This causes the different character of interaction between the grain boundaries under study and the misfit-stress field in the film. The periodic wall of edge dislocations making up a symmetric tilt boundary is essentially characterized by short-range stress fields. The stress fields of dislocations making up a periodic wall (Fig. 1a) completely compensate (shield) each other at distances exceeding the wall period H/M. Therefore, a symmetric tilt boundary with a periodic dislocation structure weakly interacts with the field of misfit stresses.

Dislocations in a faceted boundary are arranged so that the mutual shielding of their stress fields is substantially weakened. Consequently, a faceted boundary is a source of long-range stress fields and strongly interacts with misfit-stress fields in the film. In particular, dislocations in a faceted boundary can provide efficient relaxation of misfit stresses, which decreases the total elastic energy of the system as compared to the case of a symmetric tilt boundary. This decrease is the driving force for the formation of faceted grain boundaries, which have been detected experimentally in films (see, e.g., review [21] and references therein). In this work, we perform a theoretical study of the conditions of formation of faceted boundaries in the context of the model of the dislocation structure of such boundaries shown in Fig. 1c.

If facets are not long, the dislocation structure of each of them responsible for the asymmetry can be simulated, to a first approximation, as one edge superdislocation placed at the center of a facet. The Burgers vector $\mathbf{B}$ of the superdislocation is parallel to the facet plane, and its magnitude is equal to the sum of the projections of the Burgers vectors of lattice dislocations (the facet elements) along its direction. Thus, a faceted grain boundary in a film subjected to a misfit-stress field is simulated as a vertical wall of N edge superdislocations with alternating Burgers vectors $\mathbf{B}$ (differing in direction but having the same magnitude) in the misfit-stress field (Fig. 1c).

Within this model, we consider the physical states (Figs. 1a, 1b) as independent states, which are actualized upon film growth. It is assumed that either a symmetric or a faceted grain boundary (depending on the ratio of the elastic energies of the film in these physical states) forms in the film. We do not analyze transformations between these film states or possible energy barriers between them.

A certain similarity between the formation of faceted grain boundaries and the faceting of the free surfaces of crystals should be noted. However, the cause of the spontaneous faceting of a crystal flat surface is an orientation dependence of the surface free energy (see, e.g., [22]), whereas the driving force of the formation of faceted grain boundaries is their involvement in the relaxation of misfit stresses.

3. ENERGY CHARACTERISITCS OF GRAIN BOUNDARIES IN THE FILM

The energy of the film with a faceted grain boundary (Fig. 1b) is higher than the energy of the film with a symmetric boundary (Fig. 1a) by the elastic energy of the superdislocations (Fig. 1c) and by the surface energy of the boundary related to an increase in the boundary length caused by facet formation. In the course of the facet formation, however, misfit stresses relax, which should lead to a decrease in the total energy of the film. The competition of these factors specifies whether the formation of a faceted grain boundary is favorable or not as compared to the symmetric boundary. Thus, the characteristic difference ΔW between the energies of the faceted and symmetric grain boundaries in the film consists of three parts: the elastic energy of superdislocations W^{el} (which includes the energy of superdislocations and the energy of their interactions), the surface energy of the boundary W^{s}, and the interaction energy of the superdislocations with misfit stresses W^{f}:

$$\Delta W = W^{\mathrm{el}} + W^{\mathrm{s}} + W^{\mathrm{f}}. \quad (2)$$

If $\Delta W < 0$, faceting is energetically favorable. Note that the total volume of grains in the film is the same in both physical states (Figs. 1a, 1b).

Below, we calculate the terms in Eq. (2). The energy W^{el} can be represented in the form

$$W^{\mathrm{el}} = \sum_{i=1}^{N} W_i^{\mathrm{d}} + \frac{1}{2}\sum_{\substack{i,j=1 \\ i \neq j}}^{N} W_{ij}^{\mathrm{d-d}}. \quad (3)$$

Here, $W_i^{\rm d}$ is the self-energy of the ith dislocation and $W_{ij}^{\rm d\text{–}d}$ is the interaction energy between the ith and jth dislocations ($i, j = 1, \dots, N$, where the first dislocation is the nearest to the free surface and the dislocation index increases as the interface is approached). According to the general procedure for calculating the interaction energy of defects [23], the energy of interaction of two dislocations can be written as the work done in the nucleation of one dislocation in the stress field of the other dislocation (in the coordinate system shown in Fig. 1c):

$$W_{ij}^{\rm d\text{–}d} = -\int_0^{h_j} (B_{jx}\sigma_{xy}^{(i)}(x, y=0) + B_{jy}\sigma_{yy}^{(i)}(x, y=0))dx. \quad (4)$$

Here, B_{jx} and B_{jy} are the components of the Burgers vector of the jth dislocation, $\sigma_{xy}^{(i)}$ and $\sigma_{yy}^{(i)}$ are the components of the stress tensor of the ith dislocation, and h_j is the distance from the free surface to the jth dislocation. According to Fig. 1c, h_j can be represented in the form

$$h_j = \frac{2j-1}{2}L\sin\frac{\alpha}{2}. \quad (5)$$

The stress tensor components are sums of the individual contributions from each component of the Burgers vector of a dislocation, i.e., $\sigma_{xy}^{(i)} = \sigma_{xy}^{(ix)} + \sigma_{xy}^{(iy)}$ and $\sigma_{yy}^{(i)} = \sigma_{yy}^{(ix)} + \sigma_{yy}^{(iy)}$. The components of the stress tensor of the ith dislocation near the free surface have the form [23]

$$\sigma_{xy}^{(ix)}(x, y) = \frac{GB_{ix}}{4\pi(1-\nu)}\left(-2\frac{x_1}{r_1^2} + 4\frac{x_1^3}{r_1^4} + 2\frac{x_2}{r_2^2} - 4\frac{x_2^3}{r_2^4} - 2h_i\left[\frac{2}{r_2^2} - 16\frac{x_2^2}{r_2^4} + 16\frac{x_2^4}{r_2^6} + 2h_i\left(6\frac{x_2}{r_2^4} - 8\frac{x_2^3}{r_2^6}\right)\right]\right), \quad (6)$$

$$\sigma_{yy}^{iy}(x, y) = \frac{GB_{iy}}{4\pi(1-\nu)}\left(6\frac{x_1}{r_1^2} - 4\frac{x_1^3}{r_1^4} - 6\frac{x_2}{r_2^2} + 4\frac{x_2^3}{r_2^4} + 2h_i\left[-\frac{2}{r_2^2} + 16\frac{x_2^2}{r_2^4} - 16\frac{x_2^4}{r_2^6} - 2h_i\left(6\frac{x_2}{r_2^4} - 8\frac{x_2^3}{r_2^6}\right)\right]\right), \quad (7)$$

where $x_1 = x - h_i$, $x_2 = x + h_i$, and $r_n^2 = x_n^2 + y^2$, with $n = 1, 2$. The components $\sigma_{xy}^{(iy)}$ and $\sigma_{yy}^{(ix)}$ vanish at $y = 0$ and, hence, are not presented here. Substituting Eqs. (6) and (7) into Eq. (4), we obtain the pair interaction energy

$$W_{ij}^{\rm d\text{–}d} = D(B_{ix}B_{jx} + B_{iy}B_{jy}) \times\left(\ln\frac{h_i + h_j}{|h_i - h_j|} - \frac{2h_ih_j}{(h_i + h_j)^2}\right), \quad (8)$$

where $D = G/2\pi(1-\nu)$. In our model (Fig. 1c), the components B_{iy} are the same for all dislocations irrespective of their index: $B_{iy} = B\cos(\alpha/2)$. However, the sing of the components B_{ix} alternates with their index; i.e., $B_{ix} = (-1)^iB\sin(\alpha/2)$. Thus, Eq. (8) can be rewritten in the form

$$W_{ij}^{\rm d\text{–}d} = DB^2\left(\cos^2\frac{\alpha}{2} + (-1)^{i+j}\sin^2\frac{\alpha}{2}\right) \times\left(\ln\frac{h_i - h_j}{|h_i - h_j|} - \frac{2h_ih_j}{(h_i + h_j)^2}\right), \quad (9)$$

which is more convenient for further analysis.

Using a calculation procedure similar to that used above to calculate $W_{ij}^{\rm d\text{–}d}$, we find the self-energy $W_i^{\rm d}$ of the ith dislocation at a distance h_i from the free surface to be

$$W_i^{\rm d} = -\frac{1}{2}\int_0^{h_i - r_0}(B_{ix}\sigma_{xy}^{(i)}(x, y=0) + B_{iy}\sigma_{yy}^{(i)}(x, y=0))dx = \frac{DB^2}{2}\left(\ln\frac{2h_i - r_0}{r_0} - \frac{2h_i(h_i - r_0)}{(2h_i - r_0)^2}\right), \quad (10)$$

where r_0 is the dislocation core radius.

Substituting Eqs. (9) and (10) into Eq. (3), we obtain the elastic energy

$$W^{\rm el} = \frac{DB^2}{2}\sum_{i=1}^{N}\left[2\sum_{\substack{j=1\\ i\neq j}}^{N}\left(\cos^2\frac{\alpha}{2} + (-1)^{i+j}\sin^2\frac{\alpha}{2}\right) \times\left(\ln\frac{h_i + h_j}{|h_i - h_j|} - \frac{2h_ih_j}{(h_i + h_j)^2}\right) + \left(\ln\frac{2h_i - r_0}{r_0} - \frac{2h_i(h_i - r_0)}{(2h_i - r_0)^2}\right)\right]. \quad (11)$$

The difference between the surface energies of the faceted and symmetric boundaries is

$$W^{\rm s} = N\gamma L - \gamma H = \gamma(NL - H). \quad (12)$$

Here, N is the number of facets, L is the facet length, H is the film thickness, and γ is the surface energy density of the grain boundary.

In general, the interaction energy between the ith dislocation and the misfit stress field is given (by analogy with Eq. (4)) by the formula

$$W_i^{\rm f} = -\int_0^{h_j} (B_{ix}\sigma_{xy}^{(\rm f)}(x, y = 0) + B_{iy}\sigma_{yy}^{(\rm f)}(x, y = 0))dx, \quad (13)$$

where $\sigma_{xy}^{(\rm f)}$ and $\sigma_{yy}^{(\rm f)}$ are the components of the misfit-stress tensor. Since the off-diagonal components of the misfit-stress tensor are zero and the diagonal ones have the form $\sigma^{(\rm f)} = 4\pi D(1+\nu)f$ [6], we have from Eq. (13)

$$W_i^{\rm f} = -B_{iy}\int_0^{h_j}\sigma^{(\rm f)}dx = -4\pi DB\cos\frac{\alpha}{2}(1+\nu)fh_i. \quad (14)$$

Summing Eq. (14) over i and using Eq. (5), we obtain the energy $W^{\rm f}$:

$$\begin{aligned} W^{\rm f} &= \sum_{i=1}^{N} W_i^{\rm f} = -4\pi DB\cos\frac{\alpha}{2}(1+\nu)f\sum_{i=1}^{N}h_i \\ &= -4\pi DB\cos\frac{\alpha}{2}(1+\nu)f\sum_{i=1}^{N}\frac{(2i-1)}{2}L\sin\frac{\alpha}{2} \\ &= -\pi DB(1+\nu)fL\sin\alpha\sum_{i=1}^{N}(2i-1) \\ &= -\pi DB(1+\nu)fLN^2\sin\alpha. \end{aligned} \quad (15)$$

Thus, we found all components of the difference between the energies of the faceted and symmetric boundaries, ΔW. Substituting Eqs. (11), (12), and (15) into Eq. (2), we obtain the final expression

$$\begin{aligned} \Delta W = \frac{DB^2}{2}\sum_{i=1}^{N}\Bigg[2\sum_{\substack{j=1\\ i\neq j}}^{N}\left(\cos^2\frac{\alpha}{2} + (-1)^{i+j}\sin^2\frac{\alpha}{2}\right) \\ \times\left(\ln\frac{h_i+h_j}{|h_i-h_j|} - \frac{2h_ih_j}{(h_i+h_j)^2}\right) \\ + \left(\ln\frac{2h_i-r_0}{r_0} - \frac{2h_i(h_i-r_0)}{(2h_i-r_0)^2}\right)\Bigg] \\ + \gamma(NL-H) - \pi DB(1+\nu)fLN^2\sin\alpha. \end{aligned} \quad (16)$$

4. RESULTS OF MODEL CALCULATIONS

Using Eq. (16), derived for the characteristic difference between the energies of the faceted and plane symmetric grain boundaries, we find the dependence of ΔW on the parameters of the system. First, we determine the dependence of ΔW on the misfit parameter f at various values of the Burgers vector of a superdislocation. The following values of the parameters are used: elastic modulus $G = 100$ GPa, $\nu = 0.3$, facet length $L = 10$ nm, angle between adjacent facets $\alpha = 90°$, number of facets $N = 100$ [therefore, the film thickness is $H = NL\sin(\alpha/2) \approx 700$ nm], and surface energy density characteristic of aluminum $\gamma = 0.6$ J/m^2 [24]. The Burgers vector of a superdislocation is $B = nb$, where b is the Burgers vector of a lattice dislocation. The superdislocation core radius is taken to be $r_0 = B$.

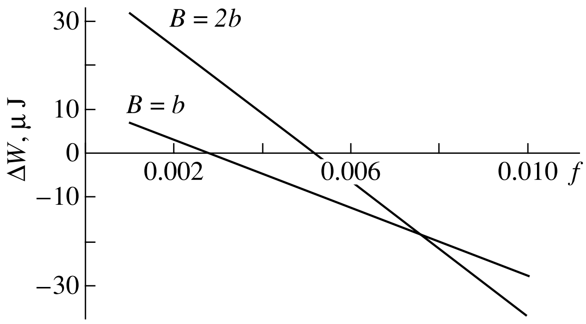

Fig. 2. Dependence of the difference ΔW between the energies of faceted and plane grain boundaries on the misfit parameter f in a film $H = 700$ nm thick at various values of the Burgers vector of superdislocations.

The calculated $\Delta W(f)$ dependence from Eq. (16) at $b = 0.4$ nm and $H = 700$ nm is given in Fig. 2. As the misfit parameter f increases, the energy difference ΔW is seen to decrease linearly and reach negative values, which means that the contribution of the relaxation term is predominant. Therefore, the formation of a faceted boundary becomes energetically favorable as compared to a symmetric tilt boundary. The data in Fig. 2 allow the following conclusion about the effect of the Burgers vectors of superdislocations on ΔW: at small values of the misfit parameter, an increase in the Burgers vector leads to an increase in ΔW, since the elastic energy of superdislocations increases in proportion to B^2 and the interaction energy between superdislocations and the misfit-stress field depends linearly on B. However, at large values of the misfit parameter, when misfit-stress relaxation becomes predominant, an increase in the Burgers vector of a superdislocation decreases ΔW.

The $\Delta W(B)$ dependence calculated from Eq. (16) at various values of f and the system parameters given above is shown in Fig. 3. The value of B was varied from b to $5b$. The plots in Fig. 3 exhibit a very strong dependence of ΔW on the Burgers vector of superdislocations. The maximum value of B at which the formation of a faceted boundary is still favorable as compared to a symmetric tilt boundary is equal to $3b$ at realistic values of the misfit parameter. Figure 4 shows the dependence of ΔW on the film thickness for various values of the misfit parameter at $B = b$. These dependences exhibit three possible types of behavior: (1) a faceted boundary is energetically unfavorable over the whole

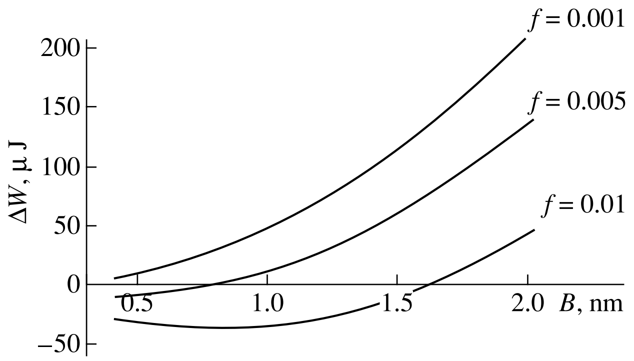

Fig. 3. Dependence of the difference ΔW between the energies of faceted and plane grain boundaries on the Burgers vector of superdislocations in a film H = 700 nm thick at various values of the misfit parameter f.

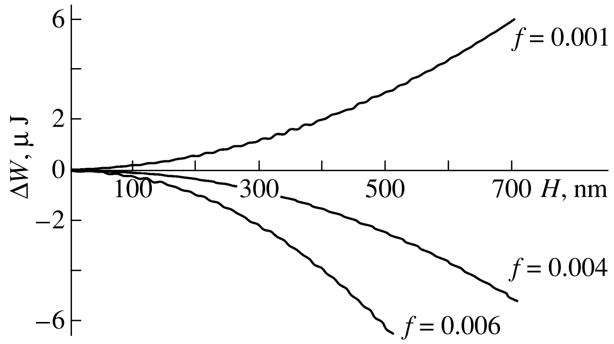

Fig. 4. Dependence of ΔW on the film thickness at various values of the misfit parameter.

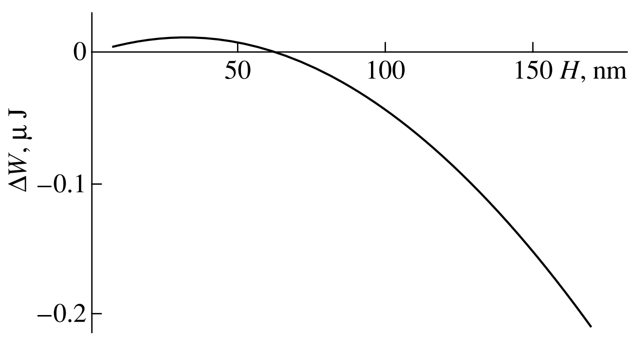

Fig. 5. Dependence of ΔW on the film thickness H at f = 0.004 and $B = b$.

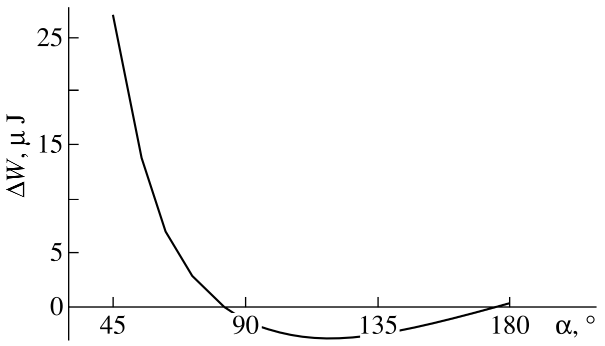

Fig. 6. Dependence of ΔW on the angle α between facets at f = 0.003 and $B = b$.

thickness range (Fig. 1b), (2) a faceted boundary is energetically favorable over the whole thickness range, and (3) a faceted boundary is energetically unfavorable in a thin film and becomes favorable at film thicknesses greater than a certain critical value. The curve at f = 0.004 in Fig. 4 illustrates the third type of behavior. The same curve is shown in Fig. 5 on a larger scale.

The plot in Fig. 6 shows the dependence of ΔW of the angle α between facets (at $f = 0.003$, $B = b$, $H \cong 700$ nm). Since both the elastic and relaxation terms in ΔW depend on the angle between facets (the latter term vanishes at $\alpha = 180°$), the $\Delta W(\alpha)$ dependence exhibits a minimum corresponding to the most favorable angle between facets.

5. CONCLUSIONS

Thus, in this work, we have theoretically studied a new mechanism of misfit-stress relaxation in polycrystalline films, namely, the formation of faceted grain boundaries whose facets are asymmetric tilt boundaries. We have constructed a model to describe a faceted grain boundary in a film placed on a semi-infinite substrate in the presence of misfit stresses. Using this model, we calculated the difference between the energies characterizing the states of the film with a faceted boundary (Fig. 1b) and a symmetric tilt boundary (Fig. 1a). The parameters of the system that exhibit a significant effect on the formation of faceted grain boundaries in films are the misfit parameter, facet asymmetry (which is characterized by the Burgers vector of a superdislocation), film thickness, and the angle between facets. Within the model proposed, the ranges of the system parameters were found in which faceted grain boundaries are energetically favorable. The model agrees with the experimental data (see review [21] and references therein) on faceted grain boundaries observed in superconducting films. The results obtained within the model can be used to prepare polycrystalline films with a given (faceted or nonfaceted) structure of grain boundaries, which significantly affects the physical (in particular, superconducting [25, 26]) properties of films.

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