J. Phys.: Condens. Matter **11** (1999) 6521–6527. Printed in the UK PII: S0953-8984(99)01065-6

Misfit dislocation walls in solid films

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Received 19 January 1999, in final form 18 May 1999

Abstract. A theoretical model is proposed which describes a new physical micromechanism for relaxation of misfit stresses in crystalline films, namely the formation of misfit dislocation walls. Energetic characteristics of films with misfit dislocation walls are estimated and compared with those of films with 'standard' misfit dislocation rows. Misfit dislocation walls are recognized as misfit defect configurations that can be formed, in particular, in films resulting from convergence of island films.

Misfit stresses occur in crystalline films due to the geometric mismatch at interphase boundaries between crystalline lattices of films and substrates. In most cases a partial relaxation of misfit stresses is realized via generation of misfit dislocations (MDs) that form dislocation rows in interphase boundary planes; see e.g. [1–5]. Generally speaking, the formation of MD rows at interphase boundaries is either desirable or disappointing, from an applications viewpoint, depending on the roles of the films and interphase boundaries in applications of heteroepitaxial systems. So, if the properties of a film are exploited, the formation of MD rows commonly is desirable as it results in a (partial) compensation for misfit stresses in the film. If the properties of an interphase boundary are exploited, the formation of MD rows commonly is undesirable, since the MD cores formed violate the pre-existing ideal (coherent) structure of the interphase boundary. The main aim of this paper is to suggest and theoretically examine an alternative physical micromechanism for relaxation of misfit stresses which results in a 'weaker' violation of the ideal (coherent) interphase boundary structure than the 'standard' formation of MD rows. This new micromechanism is the formation of MD walls in crystalline films.

Let us consider a model heteroepitaxial system consisting of an elastically isotropic semi-infinite crystalline substrate and an elastically isotropic thin crystalline film with thickness h. For simplicity, hereinafter we confine our examination to the situation with one-dimensional misfit characterized by the misfit parameter $f = (a_2 - a_1)/a_1 < 0$, where a_1 and a_2 are the crystal lattice parameters of the substrate and the film, respectively. The shear stress G and the Poisson ratio ν are assumed to be identical for the substrate and the film.

Let us consider the model heteroepitaxial system in the situation with a coherent interphase boundary. Owing to the geometric mismatch between the crystalline lattices of the film and the substrate, the film is elastically uniformly distorted. It is characterized by the elastic strain $\varepsilon = -f$. Since $f < 0$, the corresponding misfit stresses are tensile in the film.

The standard physical micromechanism for relaxation of misfit stresses is the formation of a row of MDs in the interphase boundary plane (figure 1(a)) that induce compressive stress fields

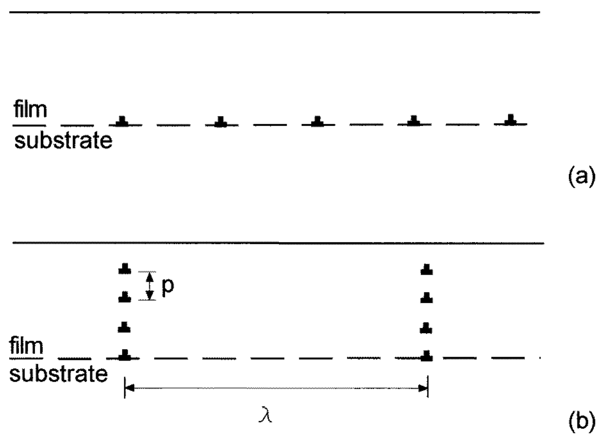

Figure 1. Physical micromechanisms for relaxation of misfit stresses: (a) formation of a misfit dislocation row, and (b) formation of a misfit dislocation walls. λ and p are the spacings between the walls and between the dislocations in a wall, respectively.

partly compensating for the tensile misfit stresses (or, in other words, partly accommodating the misfit f) [1–6]. We think that an effective alternative to the standard micromechanism is the formation of walls of MDs in the film (figure 1(b)) that induce compressive stress fields. Whether it is the standard or the alternative micromechanism for relaxation of misfit stresses that is realized depends on kinetic factors (related to the technology of the deposition of the film on the substrate) and the degree of misfit stress relaxation caused by such micromechanisms.

Let us briefly discuss a situation where formation of MD walls is kinetically favourable. Regardless of the values of the 'equilibrium' parameters (critical thickness of a film, elastic energy density, etc) which characterize the MD walls, their formation can occur at non-equilibrium conditions in films, resulting from convergence of island films. In fact, misfit stresses in an island film partly relax via the sloping of the edge surfaces of the island film (figure 2(a)). In these circumstances, when two island films converge, their contact-edge surfaces are crystallographically misoriented. As a corollary, the convergence process results in the formation of a boundary with a low-angle crystallographic misorientation in the contact area of the films (figure 2). That is, the convergence of two island films leads to the transformation of their contact-edge surfaces (being crystallographically misoriented) into an interface, a low-angle grain boundary (figure 2). At the same time, any low-angle boundary in a crystal is represented as a wall of dislocations [7]. In the situation discussed, a low-angle boundary in the film resulting from the convergence of two island films (figure 2) is naturally interpreted as a wall of MDs.

Now let us turn to analysis of the 'equilibrium' characteristics of MD walls, specifying their contribution to the misfit stress relaxation. In order to evaluate the degree of misfit stress relaxation in the situation with MD walls (figure 1(b)), let us estimate the elastic energy density W of a film with MD walls. In doing so, for simplicity, we assume the following: MD walls are periodically arranged with period λ along the interphase boundary; MDs are edge dislocations of the 90° type—that is, edge dislocations with a glide plane perpendicular to the normal to the interphase boundary plane; the Burgers vectors of MDs are identical and equal to $b = a_2$; and the distances between neighbouring MDs in dislocation walls are identical and equal to p. Also, it should be noted that MD walls are of finite extent, as a result of which (in the spirit of the theory of disclinations; see e.g. [8]) a stress-field source of the disclination type exists

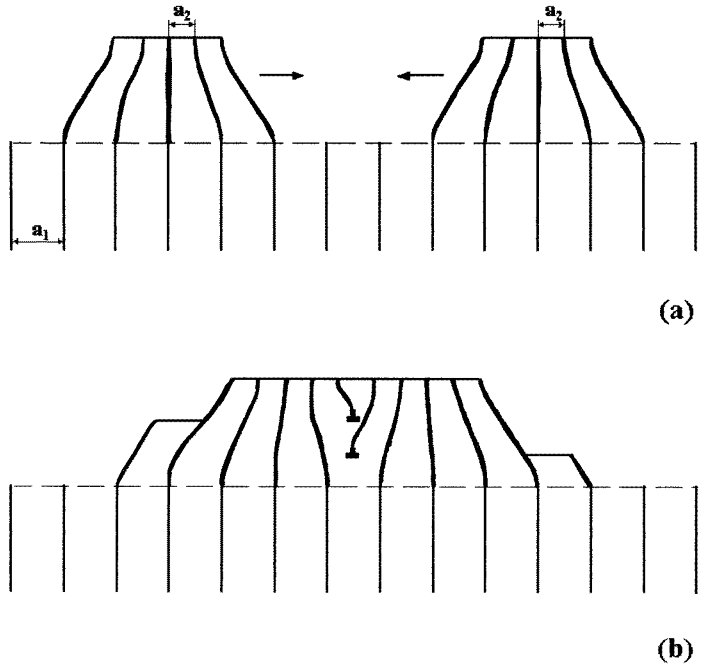

Figure 2. Convergence of island films (shown schematically). (a) Island films migrate towards each other. (b) Island films converge, whereupon a MD wall (a low-angle boundary) is formed.

at the 'internal' termination point of every MD wall. In other words, disclinations exist at junctions of the MD walls and the interphase boundary (figure 1(b)). In these circumstances, in the first approximation (which corresponds to the Matthews approximation [4, 9] for MD rows), the elastic energy density W has the three basic constituents:

$$W = W^f + W^d + W^\omega \tag{1}$$

where W^f is the proper elastic energy density of the residual misfit (uncompensated for by MDs), and W^ω and W^d are respectively the energy densities of MD walls and the disclinations associated with MD walls.

Generally speaking, misfit disclinations provide only a partial relaxation of misfit stresses, as a result of which there is some residual elastic strain ε which corresponds to the residual misfit stresses in the film. The corresponding elastic energy density W^f is determined in the standard way [3, 4] as follows:

$$W^f = 2G\varepsilon^2 h(1+\nu)/(1-\nu). \tag{2}$$

In calculations of W^d, we use the results of calculations [8] that deal with the proper elastic energy of a wedge disclination located near a free surface in a semi-infinite solid. In doing so, for simplicity, we restrict our consideration to the situation with the film thickness h lower than the distance λ between neighbouring MD walls and, as a corollary, between neighbouring disclinations located at termination points of MD walls at the interphase boundary. Such a situation ($h < \lambda$) can often arise in real heteroepitaxial systems. In fact, MDs, in general, provide only a partial accommodation of the misfit f, as a result of which the residual strain is $\varepsilon = |f| - B/\lambda$, where B denotes the Burgers vector sum of MDs in one wall, and B/λ the part of the pre-existing misfit f which is accommodated by the MDs. The parameters of the MD wall in our model (figure 1(b)) force the relationship $B = hb/p$, where h/p is the number of MDs in one wall. As a result, we have the following relationship between h and λ:

$$\lambda = hb/p(|f| - \varepsilon). \qquad (3)$$

Since $|f| - \varepsilon \leqslant |f|$ and $|f|$ ranges from 10^{-3} or 10^{-2} in real heteroepitaxial systems, the situation with $h < \lambda$ (or, as results from formula (3), the situation with the distance between neighbouring MDs in a MD wall $p \leqslant 10^2b$–10^3b) can often arise in such systems. With this taken into account, in terms of our model, we find with the help of calculations [8] the proper elastic energy density of the misfit disclinations, whose periodic distribution along the interphase boundary is characterized by the linear density λ^{-1}, as follows:

$$W^d = G\omega^2h^2/4\pi(1-\nu)\lambda \qquad (4)$$

where the thickness h of the film plays the role of the distance between misfit disclinations and the free surface of the film, and ω is the disclination power (in our model, $\omega \approx b/p \ll 1$). It should be noted that the elastic interaction between the disclinations is negligibly small in the situation discussed (figure 1(b)), since the screening length h (the distance between a disclination and the free surface) for disclination stress fields is lower than the spacing λ between the disclinations ($h < \lambda$).

Let us estimate the energy density W^ω which specifies the MD walls (without taking into account the contribution related to the disclinations; see above). W^ω can be calculated with the help of a formula (known in the theory of dislocations; see e.g. [7]) for the energy density of an infinite dislocation wall as follows:

$$W^\omega = \frac{Gb^2h}{4\pi(1-\nu)p\lambda}\left(\ln\frac{R}{r_0} + Z\right). \qquad (5)$$

Here R denotes the screening length for stress fields of MDs composing a wall ($R \approx p$), r_0 is the radius of a MD core ($r_0 \approx a_2$), and Z is a factor taking into account the contribution of the MD core to the elastic energy density ($Z \approx 1$).

From (1), (2), (4), and (5) for the characteristic values $R \approx p$, $Z \approx 1$, $\omega \approx b/p$, and $r_0 \approx a_2$, we obtain the following formula for the elastic energy density of the film with MD walls:

$$W \approx 2G\frac{1+\nu}{1-\nu}\varepsilon^2h + \frac{Gb^2h}{4\pi(1-\nu)p\lambda}\left(\ln\frac{p}{a_2} + 1\right) + \frac{Gh^2b^2}{4\pi(1-\nu)\lambda p^2}. \qquad (6)$$

From (3) and (6) one can the find dependence $W(\varepsilon)$ whose minimum corresponds to the so-called equilibrium value $\tilde{\varepsilon}$ of the residual strain in the film. This value characterizes the elastically deformed film with MD walls at equilibrium conditions. From (3) and (6) it follows that

$$\tilde{\varepsilon} = -\frac{b}{16\pi(1+\nu)h}\left(\ln\frac{p}{a_2} + \frac{h}{p} + 1\right). \qquad (7)$$

Another important parameter of the film is its critical thickness h_c which, as for the situation with MD rows (see e.g. [3, 4]), is defined as follows. For the film with thickness h higher (lower, respectively) than h_c, the existence of misfit disclinations is energetically preferable (not preferable, respectively) as compared to the coherent state of the interphase boundary. h_c is derived from equation (7) with $\tilde{\varepsilon}$ substituted for with $-f$, in which case we find

$$h_c = \frac{bp[\ln(p/a_2) + 1]}{16\pi f(1+\nu)p - b}. \qquad (8)$$

Let us consider two situations: the situation with a film containing walls of MDs (figure 1(b)) and the situation with a film containing a row of MDs (figure 1(a)); they are characterized by the same averaged MD density. For characteristic values of $h/p > 1$, the elastic energy W of the film with MD walls, given by formula (6) (with (3) taken into account) is larger than the energy density W^* of the film with a row of MDs, given by the Matthews formula (see e.g. [4, 9]):

$$W^* = 2G\frac{1+\nu}{1-\nu}\varepsilon^2 h + \frac{Gb(|f|-\varepsilon)}{4\pi(1-\nu)}\left(\ln\frac{h}{a_2}+1\right). \tag{9}$$

For a quantitative characterization of the difference between the elastic energy densities of MD rows (figure 1(a)) and MD walls (figure 1(b)), let us estimate the characteristic ratio $r = (W - W^*)/W^*$ defined by the following formula, which follows from formulae (5), (6), and (9):

$$r = \frac{b(|f|-\varepsilon)[(h/p) - \ln(h/p)]}{8\pi(1+\nu)\varepsilon^2 h + b(|f|-\varepsilon)[\ln(h/a_2)+1]}. \tag{10}$$

At the initial stage of formation of MD structures (figures 1(a) and 1(b)), the values of $|f| - \varepsilon$ are small, in which case r is small. So, for characteristic values of $|f| - \varepsilon \approx 10^{-3}|f|$, $\varepsilon \approx |f| \approx 10^{-3}$–$10^{-2}$, $h/p \approx 3$–10, $h \approx (10^2$–$10^3)a_2$, $b \approx a_2$, $\nu \approx 1/3$, from formula (10) we find that the characteristic ratio r ranges from 7×10^{-6} to 2.5×10^{-3}. At the late stages of evolution of MD structures (figures 1(a) and (b)), the values of $|f| - \varepsilon$ are of the same order as $|f|$, in which case r is comparatively large. So, for characteristic values of $|f| - \varepsilon \approx |f|$ and $\varepsilon \approx 10^{-3}|f|$, and the above values of the other parameters ($|f|$, h/p, h, b, and ν), from formula (10) we obtain that r ranges from 0.25 to 1.5.

Our estimates of r indicate that MD rows (figure 1(a)) in films, from an energetic viewpoint, are misfit defect configurations that are nearer equilibrium (more stable) than MD walls (figure 1(b)). This conclusion is supported by numerous experimental observations of MD rows at interphase boundaries in heteroepitaxial systems; see e.g. [1–5]. At the same time, the difference between the energy densities of MD rows (figure 1(a)) and MD walls (figure 1(b)) at the initial stage of their formation in a film is small ($r \ll 1$), with the result that a MD wall can be formed due to some kinetic factors.

If MD walls are formed, they exist as metastable misfit defect configurations in films. In fact, the movement of a dislocation (in our case, a MD) from a dislocation wall (in our case, a MD wall) into an adjacent crystalline region requires the overcoming of an energetic barrier [7]. In other words, in order to remove a MD from a MD wall and to place this MD at the interphase boundary far from the wall or, in the general situation, in order to transform a MD wall (figure 1(b)) into a MD row (figure 1(a)), energetic barriers have to be overcome.

In these circumstances, MD walls in films (figure 1(b)) are metastable configurations, since $W > W^*$. The formation of such metastable MD walls can occur under non-equilibrium conditions—for example, in films resulting from convergence of island films (figure 2). This serves as a natural explanation of experimental data [10] indicative of special interfaces (different from high-angle grain boundaries) being formed in films resulting from the convergence of island films.

Also, in general, MD walls can be formed, for kinetic reasons, in films with threading dislocations which (by definition [4, 5]) are extended from ‘parent’ dislocations entering from the substrate into the interphase boundary. For instance, a MD wall can be formed due to a kinetically favourable rearrangement of several dislocations which are extensions of neighbouring threading dislocations (with the same Burgers vectors) that enter the interphase boundary in some local region (figure 3). In other words, for kinetic reasons related to a high local density of the parent dislocations, a ‘beam’ of threading dislocations can be extended into a MD wall (rather than an ‘equilibrium’ planar row of MDs at the interphase boundary) in the film (figure 3).

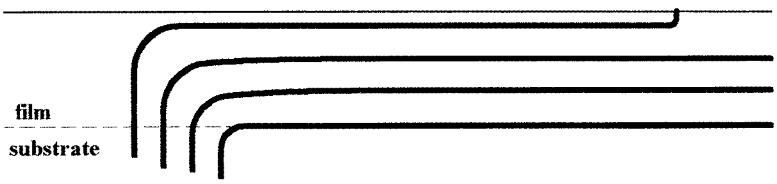

Figure 3. The arrangement of threading dislocations in a MD wall.

In general, MD walls can be formed also under equilibrium conditions—for instance, in multilayer films consisting of alternate layers with various parameters; say, with various values of the layer thickness (figure 4). In this situation (figure 4), the misfit stress distribution and the energetic characteristics of MD walls are different from those in the situation with single-layer films (figure 1), and, therefore, MD walls can be stable—energetically favourable—in multilayer films at some values of their parameters. Detailed (labour-intensive) estimations relating to the peculiarities of MD walls resulting from the rearrangement of threading dislocations (figure 3) as well as MD walls in multilayer films (figure 4) represent separate problems whose consideration is beyond the scope of this paper.

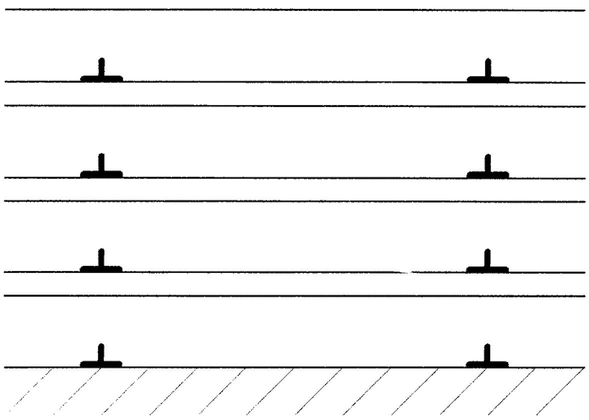

Figure 4. A MD wall in a multilayer film.

Thus the suggested physical micromechanism for the relaxation of misfit stresses—generation of MD walls (figure 1(b))—is an effective alternative to the standard micromechanism—generation of MD rows (figure 1(a))—in films resulting from the convergence of island films. MD walls provide a 'weaker' violation of the ideal (coherent) interphase boundary structure, in which case the formation of MD walls is preferable, from an applications viewpoint, to that of MD rows in heteroepitaxial systems with interphase boundaries used as functional elements in applications.

Acknowledgments

Many thanks are due to M Yu Gutkin and S A Kukushkin for helpful discussions. This work was supported, in part, by the Russian Foundation of Basic Research (Grant 98-02-16075), the Office of US Naval Research (Grant N00014-99-1-0569), and the Volkswagen Foundation (Research Project No 05019225).

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