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Competing relaxation mechanisms in strained semiconducting and superconducting films

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Abstract
A theoretical model is suggested which describes the crossover from the disclination (rotational) mechanism of misfit stress relaxation to the dislocation (translational) mechanism in strained solid films, with the emphasis on the case of semiconducting and high-transition-temperature superconducting films. In the framework of the model, the ranges of parameters (film thickness, characteristics of disclination arrangement, misfit parameter) of disclinated films for which the generation of misfit dislocations is energetically favourable are calculated. The specific features of films with misfit disclinations and dislocations are discussed in relation to the technologically interesting possibility of exploiting either disclinated or dislocated films for applications.

1. Introduction

Semiconducting and high-transition-temperature (T_C) superconducting films are the subject of intensive studies in condensed matter physics, motivated by their diverse applications and the interest of fundamental physical phenomena occurring in these films. Defect structures in semiconducting and high-T_C superconducting films are among the most important issues for their physical properties. For instance, grain boundaries in high-T_C superconducting films dramatically suppress the critical current density [1–3]. Dislocations and stacking faults play an important role in the degradation of optoelectronic functional properties of semiconducting continuous films and self-assembled nanoislands [4, 5].

In many cases, defects in solid films are generated, causing relaxation of misfit stresses that occur due to geometric mismatch between crystal lattice parameters of films and substrates. The most widespread physical mechanisms for relaxation of misfit stresses in continuous and island films are associated with the formation of misfit dislocations and their complicatedly arranged configurations; see, e.g., [6–20]. However, non-conventional misfit defects—misfit disclinations (rotational defects)—are also capable of causing effective relaxation of misfit stresses; see the experimental data [21–25] and theoretical models [26–30]. For instance, misfit disclinations at junctions of grain and twin boundaries in Ge films on Si substrates have been experimentally detected [21–23, 25]. Recently, reference [24] has reported experimental observation of misfit disclination dipoles and quadrupoles in epitaxial rhombohedral ferroelectric films.

In general, disclinations in real solids are located at grain boundaries where they terminate grain boundaries of finite extent or separate grain boundary segments with different misorientation parameters; see, e.g., [21–32]. In this context, formation of grain boundaries in growing solid films indirectly indicates that the disclination mechanism of misfit stress relaxation can effectively come into play when misfit dislocations are not generated. In particular, this is the case for high-T_C superconducting films where the relaxation of misfit stresses through the formation of conventional misfit dislocations is possible only for small misfit parameter values, such as in the case of YBaCuO film growth on $LaAlO_3$ or $SrTiO_3$; see the review [33]. Even in these cases, one frequently observes low-angle grain boundaries. When the misfit parameter is much larger, as in the case of YBaCuO film growth on MgO or ZrO_2, the high-T_C superconducting films are no longer single crystalline; grain boundaries are formed in such films [33]. These experimental data indicate that the formation of grain boundaries accompanies growth of high-T_C superconducting films on substrates, for large misfit parameter values, in which case disclinations at grain boundaries are capable of causing more effective relaxation of misfit stresses than conventional misfit dislocations.

Current theoretical models of misfit stress relaxation through formation of misfit dislocations at grain boundaries in solid films [25–28, 30] are based on the oversimplifying assumption that, if such disclinations are formed at the initial stage of the film growth, the disclination mechanism for stress relaxation always dominates, suppressing the formation of misfit dislocations. In general, however, both the disclination and dislocation mechanisms for stress relaxation compete during all the stages of the film growth. As a result, the crossover from one relaxation mechanism to another can occur at some critical values of the parameters of the growing films. When the crossover occurs, it results in a dramatic change of the defect structure of a growing film, that strongly affects its physical properties. This raises substantial interest in both experimental identification and theoretical analysis of the phenomenon discussed. The main aim of this paper is to suggest a theoretical model describing the crossover from the disclination relaxation mechanism to the dislocation relaxation mechanism in strained solid films, with emphasis on the case of semiconducting and high-T_C superconducting films.

2. Misfit disclinations and dislocations in strained films; model

Let us consider a model film/substrate system consisting of a crystalline film of thickness H and a semi-infinite crystalline substrate. The film and substrate are assumed to be isotropic solids having the same values of the shear modulus G and the same values of Poisson ratio ν. The film/substrate boundary is characterized by the misfit parameter $f = 2(a_f - a_s)/(a_f + a_s)$, where a_s and a_f are the crystal lattice parameters of the substrate and the film, respectively.

Let us consider a periodic row of misfit disclinations located at positions I in the film (figure 1). They induce stress fields that compensate, in part, for misfit stresses [26–30]. For certain ranges of parameters of strained films, the formation of misfit disclinations is more energetically favourable than formation of conventional misfit dislocations [28]. However, generally speaking, in the situation where misfit disclinations are energetically preferred at the initial stage of the film growth, conventional misfit dislocations may become preferred at further stages of the film growth. If this is so, misfit dislocations are effectively generated due to the motion of disclinations which, following the general theory of disclinations in solids [34], serve as dislocation sources.

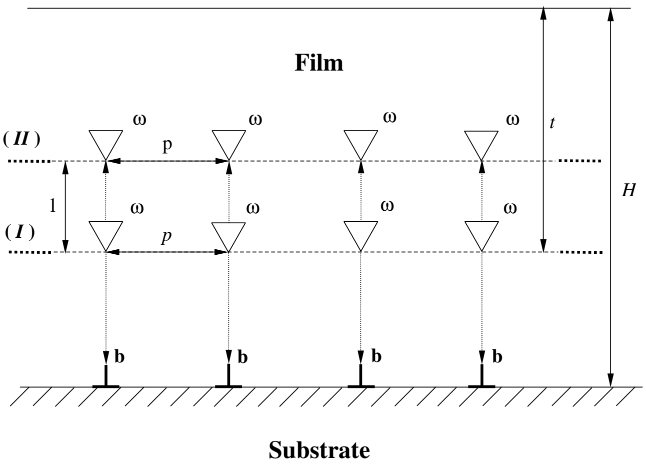

Figure 1. Transformation of the misfit defect ensemble in a solid film of thickness H. Misfit disclinations initially located at positions I move to new positions II, emitting misfit dislocations that move towards the film/substrate boundary.

In the context discussed, the first elementary act of the crossover from the disclination relaxation mechanism to the dislocation mechanism is as follows. Misfit disclinations initially located at position I move to a new position II (figure 1), emitting misfit dislocations that move towards the film/substrate boundary. The disclination transfer from position I to II, accompanied by emission of misfit dislocations that form a periodic dislocation row at the film/substrate boundary (figure 1), is characterized by the difference Δw between the energy densities (per unit area) of the film with the defect configuration before and after transfer of disclinations. The transfer in question is energetically favourable (unfavourable, respectively), if $\Delta w < 0$ (>0, respectively). The condition $\Delta w = 0$ allows one to reveal the ranges of critical parameters of the defected film/substrate system—that is, parameters at which the transition from the disclination relaxation mechanism to the dislocation mechanism occurs.

3. Energetic characteristics of misfit defect ensembles in strained films

Let us calculate the energetic characteristics of the film/substrate system with a row of disclinations (located at position I; see figure 1(a)) as well as with rows of disclinations (located at position II; see figure 1) and dislocations (located at the film/substrate boundary; see figure 1). To do so, first, it is convenient to calculate the difference Δw_1 between energies (per unit length) that characterize the state I (figure 1) and the coherent (defect-free) state of the film/substrate system. Following the standard scheme for calculation of energies characterizing disclinated films (see, for instance, [29, 30]), the energy difference has three terms:

$$\Delta W_1 = E_1^{\Delta} + E_1^{\Delta-f} + E_1^{\Delta-\Delta}. \tag{1}$$

Here E_1^{Δ} denotes the sum proper energy of disclinations located at positions I, $E_1^{\Delta-f}$ the energy that characterizes interaction between disclinations and misfit stresses, and $E_1^{\Delta-\Delta}$ the energy that characterizes interaction between disclinations.

The sum proper energy of disclinations located at positions I is calculated using the standard formula [34]:

$$E_1^{\Delta} = \tfrac{1}{2} N D \omega^2 t^2, \tag{2}$$

where $D = G/[2\pi(1-\nu)]$, N denotes the number of disclinations, ω the disclination strength, t the distance between the row of disclinations located at position I (figure 1(a)) and the film free surface. This formula is obtained in the framework of the linear elasticity theory in which the disclination stress fields weakly diverge with decrease in the distance r from the disclination line (as $\ln(r/a_f)$ when $r \to 0$). As a result, formula [34] used here in the calculation of the disclination energy is rather accurate (as is confirmed by computer modelling of grain boundary disclinations [35]) and involves the disclination core contribution. This is contrasted with a description of dislocations by methods of linear elasticity theory in which the dislocation stress fields diverge as $1/r$ when $r \to 0$ [36], and, as a corollary, the dislocation core contribution to the elastic energy is well described by the non-linear elasticity theory only [36].

The energy $E_1^{\Delta-f}$ that characterizes the interaction between disclinations (located at positions I) and misfit stresses is given by the following formula [29, 30]:

$$E_1^{\Delta-f} = 2\pi ND(1+\nu)f\omega t^2. \tag{3}$$

The sum energy $E_1^{\Delta-\Delta}$ that characterizes the interaction between all the disclinations located at positions I is calculated as the sum (over disclinations) of the energies of the pair disclination–disclination interactions, using a standard formula given in review article [34]. As a result, we get

$$E_1^{\Delta-\Delta} = 2ND\omega^2\left(S(t) - \frac{t^2}{2}\right). \tag{4}$$

Function $S(t)$ appearing in brackets in (4) is given as $S(t) = (2\pi/p)\int_0^t \tilde{t}^2 \coth[2\pi\tilde{t}/p]\,\mathrm{d}\tilde{t}$.

Now let us calculate the difference ΔW_2 between the energies (per unit length) that characterize the film/substrate system in state II (figure 1(b)) and the coherent (defect-free) state. The energy difference ΔW_2 consists of seven terms:

$$\Delta W_2 = E_2^{\Delta} + E_2^{d} + E_2^{\Delta-f} + E_2^{d-f} + E_2^{\Delta-\Delta} + E_2^{d-d} + E_2^{\Delta-d}. \tag{5}$$

Here E_2^{Δ} denotes the sum proper energy of disclinations located at positions II, E_2^d the sum proper energy of dislocations (including the contribution of dislocation cores), $E_2^{\Delta-f}$ the energy that characterizes the interaction between disclinations and misfit stresses, E_2^{d-f} the energy that characterizes the interaction between dislocations and misfit stresses, $E_2^{\Delta-\Delta}$ the energy that characterizes the interaction between disclinations, E_2^{d-d} the energy that characterizes the interaction between dislocations, and $E_2^{\Delta-d}$ the energy that characterizes the interaction between disclinations and dislocations.

State II is characterized by the following parameters of the defect system: the dislocation Burgers vector magnitude b, the interspacing p (period) between neighbouring dislocations, and the distance $l \approx b/\omega$ moved by disclinations from positions I to II. Then, the sum proper energy of disclinations located at positions II is calculated using standard formula [34] as follows:

$$E_2^{\Delta} = \tfrac{1}{2}ND\omega^2(t-l)^2. \tag{6}$$

The sum proper energy E_2^d of dislocations is represented as that consisting of their elastic energy [37] and the dislocation core contributions [36], in which case we have

$$E_2^d = \frac{NDb^2}{2}\left(\ln\frac{2H}{b} + \frac{1}{2}\right). \tag{7}$$

The sum energy $E_2^{\Delta-f}$ that characterizes the interaction between disclinations (located at positions II) and misfit stresses is given by the following formula [29, 30]:

$$E_2^{\Delta-f} = 2\pi ND(1+\nu)f(t-l)^2. \tag{8}$$

The sum energy that characterizes interaction between dislocations and misfit stresses is given by the standard formula [36]:

$$E_2^{d-f} = 4\pi N D(1+\nu) f b H. \tag{9}$$

With the standard formula given in review article [34], the energy $E_2^{\Delta-\Delta}$ that characterizes the interaction between disclinations located at positions II is as follows:

$$E_2^{\Delta-\Delta} = 2ND\omega^2\left(S(t-l) - \frac{(t-l)^2}{2}\right). \tag{10}$$

The sum energy E_2^{d-d} of the interaction between dislocations is given by the following formula [38, 39]:

$$E_2^{d-d} = NDb^2\left(\ln\frac{\sinh(t_2-t_1)}{t_2-t_1} + (t_2-t_1)\left[\coth(t_2-t_1) - \frac{1}{2}(t_2-t_1)\,\mathrm{cosech}^2(t_2-t_1)\right] - \frac{1}{2}\right), \tag{11}$$

where $t_1 = \pi(t-H)/p$, $t_2 = \pi(t+H)/p$.

Finally, the energy $E_2^{\Delta-d}$ that characterizes the interaction between disclinations and dislocations is calculated using formula [17] as follows:

$$E_2^{\Delta-d} = -\frac{NDb\omega}{2}\left(2(t-H)\ln\frac{\sinh t_1}{\sinh t_2} - \frac{4tH}{p}\coth t_2\right). \tag{12}$$

Formulae (1)–(12) are written for energies. In order to characterize the transition from one state of the film/substrate composite to another state, it is convenient to operate with the energy densities. The difference ΔW_i between the energies of the ith state of a disclinated film ($i = 1, 2$; see figure 1) and its coherent (defect-free) state is in the following relationship to the difference between the corresponding energy densities Δw_i: $\Delta w_i = \Delta W_i/(Np)$. With this relationship, from formulae (1)–(12) we find that the characteristic difference Δw between the energy densities of the states 1 and 2 of the disclinated film (figure 1) is as follows:

$$\begin{aligned}\Delta w = \Delta w_2 - \Delta w_1 = \frac{\Delta W_2}{Np} - \frac{\Delta W_1}{Np} = \frac{Db^2}{2p}\Bigg(&\ln\frac{2H}{b} + 2\ln\frac{\sinh(t_2-t_1)}{t_2-t_1}\\ &+ (t_2-t_1)[2\coth(t_2-t_1) - (t_2-t_1)\,\mathrm{cosech}^2(t_2-t_1)] - \tfrac{1}{2}\\ &- \frac{2}{l}\left((t-H)\ln\frac{\sinh t_1}{\sinh t_2} - \frac{2tH}{p}\coth t_2 + \frac{2f'H + (2t-l)(\omega - f')}{2\omega}\right)\\ &+ \frac{4}{l^2}(S(t-l) - S(t))\Bigg),\end{aligned} \tag{13}$$

where $f' = 4\pi(1+\nu)f$.

4. Results of model

The film/substrate system under consideration (figure 1) has several structural and technologically controlled parameters: the distance t between the disclination row and the film free surface, the misfit parameter f, the film thickness H, the disclination strength ω, and the period p of the disclination and dislocation rows. With formula (13), for characteristic values of $f = 10^{-2}$ and $\omega = 0.1$, we have calculated the dependences of Δw on the three parameters in the following ranges: $H = 40b$–$100b$, $t = 20b$–$40b$, $p = 10b$–$100b$. (The disclination strength is taken as $\omega = 0.1$ ($\approx 5.7°$), because it is close to the mean value of

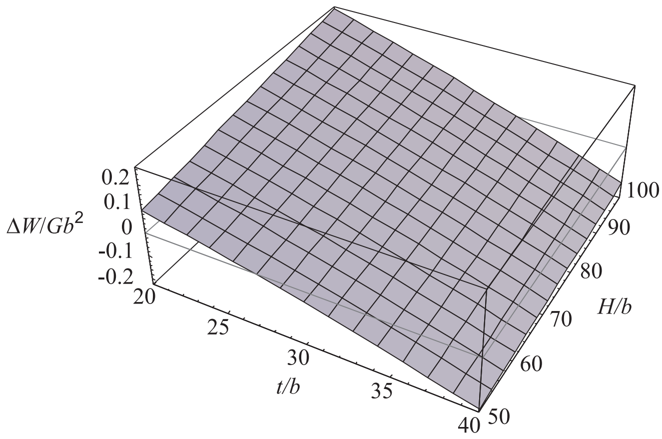

Figure 2. The dependence of energy density difference Δw on film thickness H and the distance t between the disclination row and film free surface, for $\omega = 0.1$, $p = 30b$, and $f = 10^{-2}$.

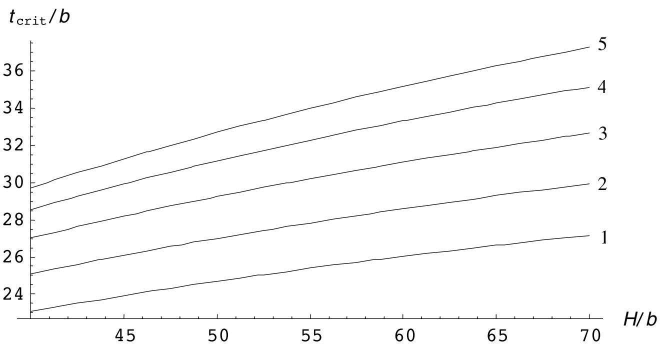

Figure 3. The dependence of critical distance t_{crit} on film thickness H at different values of the disclination row period: $p = 10b$, $30b$, $50b$, $70b$, and $90b$ (see curves 1, 2, 3, 4, and 5, respectively), for $\omega = 0.1$ and $f = 10^{-2}$.

the strength values 3.678° and 7.356° that characterize experimentally observed [22, 23, 25] disclinations in Ge films deposited onto Si substrates.)

Thus, dependence of Δw on t and H is shown in figure 2. From figure 2 it follows that the character of the $\Delta w(t)$ dependence does not change with varying film thickness H. That is, Δw essentially decreases with rising t at any H. At the same time, Δw just weakly decreases with increase of H.

Dependences of the critical distance t_{crit} between the disclinations and the film free surface on H at different values of p are shown in figure 3, where t_{crit} corresponds to the case of $\Delta w = 0$. The disclination motion accompanied by emission of dislocations (figure 1) is energetically favourable ($\Delta w < 0$) and unfavourable ($\Delta w > 0$) if $t < t_{crit}$ and $t > t_{crit}$, respectively. As follows from figure 3, t_{crit} increases with rising H (at fixed p) and p (at fixed H).

With formula (13), we have calculated the state diagram of the film/substrate system considered. It is presented as figure 4 in the coordinates (t, H, f). Surface $t = t_{crit}\ (f, H)$ separates the state region α where the disclination transfer (figure 1) is energetically favourable from the state region β where the disclination transfer (figure 1) is energetically unfavourable. As follows from figure 4, t_{crit} grows with rising misfit parameter f at fixed H.

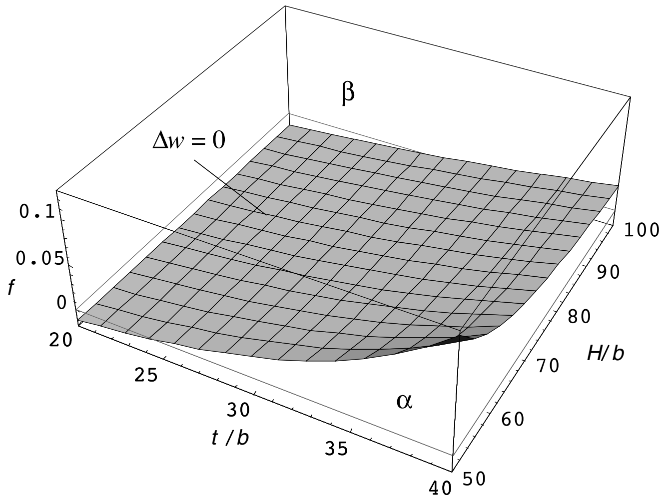

Figure 4. The state diagram of the film/substrate system with misfit disclinations. Surface $t_{crit}(f, H)$ separates region α where the transfer of disclinations is energetically favourable ($\Delta w < 0$) from region β where the transfer of disclinations is energetically unfavourable ($\Delta w > 0$).

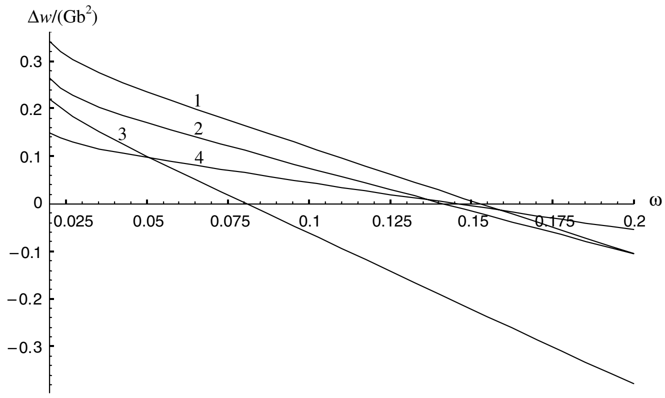

Figure 5. Dependences of the energy density difference Δw on the disclination strength ω for the following values of the parameters: $f = 10^{-2}$; $H = 80b$, $p = 30b$, $t = 20b$ (curve 1); $H = 60b$, $p = 30b$, $t = 20b$ (curve 2); $H = 60b$, $p = 30b$, $t = 30b$ (curve 3); and $H = 60b$, $p = 40b$, $t = 20b$ (curve 4).

Finally, with formula (13), we have calculated the dependences of Δw on the disclination strength ω at various values of H, p, and t. These dependences shown in figure 5 indicate that Δw is very sensitive to the disclination strength ω.

5. Concluding remarks

Here we have suggested a theoretical model which describes the crossover from the disclination mechanism to the dislocation mechanism of misfit stress relaxation in strained solid films. In the framework of the model suggested, we have calculated energetic characteristics of misfit disclinations that move towards the film free surface, emitting misfit dislocations (figure 1). This transformation of the misfit defect ensemble represents an elementary act of crossover from the disclination relaxation mechanism to the dislocation one in a growing film. We have revealed the ranges of parameters (misfit parameter, characteristics of misfit disclination arrangement, film thickness) of a film/substrate composite for which the crossover is energetically favourable. It is found that the most important parameter influencing the crossover is the distance t between the film free surface and misfit disclinations in the initial state of the system (figure 1). More precisely, the crossover from the disclination relaxation mechanism to the dislocation one is energetically favourable if $t < t_{crit}$ and unfavourable if $t > t_{crit}$. The critical distance t_{crit} increases with rising film thickness H at fixed values of the other parameters, and with rising misfit parameter f at fixed values of the other parameters of the film/substrate composite.

These results are important for technological applications of films, because they potentially allow one to influence the defect structure and, therefore, the properties of films using technologically controlled parameters (misfit parameter f, film thickness H). In fact, in general, the formation of either misfit disclinations or misfit dislocations is desirable from an applications viewpoint, depending on the roles of the films and interphase boundaries in applications of heteroepitaxial structures. So, if the properties of a film are exploited, the formation of misfit dislocation rows at the interphase boundary is commonly desirable, as it results in a (partial) compensation for misfit stresses in the film and does not disrupt the film interior. If the properties of an interphase (film/substrate) boundary are exploited, the formation of misfit dislocations at this boundary is commonly undesirable, since the dislocation cores commonly the pre-existing ideal (coherent) structure of the interphase boundary. In this case, formation of misfit disclinations is desirable, because they are commonly formed in the film interior [21–25] and, therefore, do not disrupt the ideal structure of interphase boundaries. Also, the weak-link behaviour of grain boundaries in polycrystalline thin-film cuprates forms a basis for their use in low-current microelectronics. In these applications, the disclination relaxation mechanism associated with the formation of grain boundaries is desirable, because it 'produces' grain boundaries.

With experimental data directly indicating the existence of misfit disclinations in semiconducting films [21–25] and experimental data indirectly indicating the potential existence of misfit disclinations in high-T_C superconducting films (see the review article [33] and references therein), the results of this paper are applicable to the analysis of real defect structures in these films. Generally speaking, the notion of the disclination relaxation mechanism is essential in a theoretical description of the structural features and behaviour of polycrystalline and nanocrystalline films of other systems [40, 41], in which case the results of this paper may be used in fundamental and applied research into such films, too.

Acknowledgments

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