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Surface Self-Organization in Multilayer Film Coatings

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Abstract. It is a recognized fact that during film deposition and subsequent thermal processing the film surface evolves into an undulating profile. Surface roughness affects many important aspects in the engineering application of thin film materials such as wetting, heat transfer, mechanical, electromagnetic and optical properties. To accurately control the morphological surface modifications at the micro- and nanoscale and improve manufacturing techniques, we design a mathematical model of the surface self-organization process in multilayer film materials. In this paper, we consider a solid film coating with an arbitrary number of layers under plane strain conditions. The film surface has a small initial perturbation described by a periodic function. It is assumed that the evolution of the surface relief is governed by surface and volume diffusion. Based on Gibbs thermodynamics and linear theory of elasticity, we present a procedure for constructing a governing equation that gives the amplitude change of the surface perturbation with time. A parametric study of the evolution equation leads to the definition of a critical undulation wavelength that stabilizes the surface. As a numerical result, the influence of geometrical and physical parameters on the morphological stability of an isotropic two-layered film coating is analyzed.

PROBLEM FORMULATION

Multilayered thin film structures are extensively used in engineering systems to accomplish a wide range of specific functions. However, these materials can be subjected to thermomechanical, electromechanical, electromagnetic, chemical or radioactive impacts causing a stress concentration near the roughened surface that may reduce the durability of a device [1, 2]. Numerous experimental and theoretical results demonstrate that surface effects become important in the mechanical behavior of nanosized structural elements [3, 4]. A high device quality can only be maintained if defects in film are kept to minimum. Throughout the years, numerous deposition techniques have been developed to grow defect-free thin film coatings, but the defect nucleation continues to be a problem. To improve the fabrication of devices based on thin film technology, it is necessary to understand the processes by which various defects are formed. It is well known that multilayer film structures are stressed due to lattice mismatch of different layers [5–7]. In order to minimize the total free energy of such material, the surface shape can be self-organized by mass transfer [8, 9]. Thus, the pattern formation can be considered as the mechanism of morphological instability under the constraints set by the material balance. It should be noted that many researchers analyzed morphological evolution of solid surface [10]. However, we are cognizant of only a few papers where the multilayer thin film materials were considered [11, 12]. Unfortunately, they neglected the effect of volume diffusion. In this paper, we extend the theoretical analysis of morphological instability of multilayered structures performed in our previous studies [13] and demonstrate that volume fluxes could promote surface roughening with tensile stress as well as smoothing of initial perturbation with compressive stress.

Consider an isotropic multilayer film coating of a total thickness $h_{\mathrm{f}} = \sum_{r=1}^{N} h_r$ which consists of N dissimilar layers and is deposited on a substrate with Poisson's ratio ν_{N+1} and shear modulus μ_{N+1} under plane strain conditions. The layer of thickness h_r has Poisson's ratio ν_r and shear modulus μ_r.

The substrate is modeled as an elastic half-plane of complex variable $z = x_1 + ix_2$

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$$\Omega_{N+1} = \{z: x_2 < 0, x_1 \in R^1\}. \quad (1)$$

The coating is presented as coherently bonded strips Ω_r

$$\Omega_r = \{z: H_{r+1} < x_2 < H_r, x_1 \in R^1\},$$

$$H_N = h_N, H_{N+1} = 0, H_r = H_{r+1} + h_r, r = \overline{2, N} \quad (2)$$

with rectilinear boundaries

$$\Gamma_r = \{z: z \equiv z_r = x_1 + iH_r\}, r = \overline{2, N+1}. \quad (3)$$

We assume that the film surface has a sinusoidal small perturbation that changes with time τ

$$\Gamma_1 = \{z: z \equiv z_1 = x_1 + i[H_1 + A(\tau)\cos(kx_1)]\}, \quad (4)$$

$$A(\tau)/\lambda = \varepsilon(\tau) \ll 1 \, \forall\tau, k = 2\pi/\lambda, A(0) = a. \quad (5)$$

The conditions at the free surface, interfaces and infinity are respectively

$$\sigma(z_1) = 0, z_1 \in \Gamma_1, \quad (6)$$

$$\Delta u(z_r) = u^+ - u^- = 0, \ \Delta\sigma(z_r) = \sigma^+ - \sigma^- = 0, \quad (7)$$

$$\sigma_{22}^\infty = \sigma_{12}^\infty = 0, \ \sigma_{11}^\infty = \sigma_{N+1}, \omega^\infty = 0. \quad (8)$$

In Eqs. (6)–(8), $u = u_1 + iu_2, \sigma = \sigma_{nn} + i\sigma_{nt}, u_1, u_2$ are the displacements along corresponding axes of the Cartesian coordinates x_1, x_2, σ_{nn}, σ_{nt} are the components of stress vector σ at the area with normal n in the local Cartesian coordinate system n, t, $u^\pm = \lim_{z \to z_r \pm i0} u(z)$, $\sigma^\pm = \lim_{z \to z_r \pm i0} \sigma(z), z_r \in \Gamma_r, r = \overline{2, N+1}$, $\sigma_{\alpha\beta}^\infty = \lim_{x_2 \to -\infty} \sigma_{\alpha\beta}$, $\omega^\infty = \lim_{x_2 \to -\infty} \omega, \sigma_{\alpha\beta}$ (α, β = 1, 2) are the components of the stress tensor in the axes x_1, x_2 and ω is the rotation angle of a material particle.

As it was mentioned above, the analysis of morphological instability is based on a combined effect of surface and volume diffusion that are assumed to take place in the region close to the free surface. The motion of the surface is given by the following equation [8, 9]

$$\frac{\partial h(x_1, t)}{\partial t} = K_s \frac{\partial^2}{\partial x_1^2}\left[U(x_1, t) - \gamma \frac{\partial^2 h(x_1, t)}{\partial x_1^2}\right] + K_v k \left[\gamma \frac{\partial^2 h(x_1, t)}{\partial x_1^2} + \Delta P(x_1, t)\right], \quad (9)$$

where $K_s = D_s C_s \Omega^2 / k_B T_a$, $K_v = D_v C_v \Omega / k_B T_a$, Ω is the atomic volume, D_s is the surface self-diffusivity, C_s is the number of diffusing atoms per unit area, k_B is the Boltzmann constant, T_a is the absolute temperature, D_v is the vacancy self-diffusivity in bulk of top layer, C_v is the concentration of vacancies in the bulk of top layer in equilibrium with a flat film surface under a remote stress, γ is the surface energy, U is the elastic strain energy at the perturbed film surface, and ΔP is the variation of the hydrostatic pressure at rough and flat free surface.

Here, the elastic deformation caused by surface perturbation is treated as a quasi-static state. Thus, in order to integrate surface evolution equation (9), we solve the corresponding boundary-value problem of plane elasticity for the multiply connected domain $\Omega = \bigcup_{r=1}^{N+1} \Omega_r$ under boundary conditions (6), (7) and conditions at infinity (8). Using the complex variable representations, superposition method and boundary perturbation technique, the original boundary value problem is reduced to a successive solution of the set of Fredholm integral equations [5].

NUMERICAL RESULTS

After the elasticity problem was solved, we could determine the elastic strain energy and the variation of the hydrostatic pressure. Then, evolution equation (9) is integrated and we obtain the amplitude of perturbation (4) as a function of time

TABLE 1. The effect of compressive and tensile stresses on the critical value of undulation wavelength

μ_1/μ_2		0.3	0.3	3	3
μ_2/μ_3		0.3	3	0.3	3
h_1, μm	h_2, μm	$(\lambda_{cr}^{-}-\lambda_{cr}^{+})/\lambda_{cr}^{+}$			
0.6	0.6	0.36	0.30	0.18	0.17
1.2	0.6	0.24	0.24	0.21	0.21
0.6	1.2	0.33	0.32	0.18	0.18

$$\ln\left(\frac{A(\tau)}{a}\right) = P(\lambda, h_1, ..., h_N, \mu_1, ..., \mu_{N+1}, \nu_1, ..., \nu_{N+1}, \gamma, D, \sigma_{N+1})\tau. \quad (10)$$

The critical value of perturbation wavelength λ_{cr} that stabilize the surface is determined from the following condition

$$P(\lambda, h_1, ..., h_N, \mu_1, ..., \mu_{N+1}, \nu_1, ..., \nu_{N+1}, \gamma, D, \sigma_{N+1}) = 0, \; D = \frac{D_v C_v}{D_s C_s}. \quad (11)$$

For surface undulations with wavelengths less than critical value, the amplitude of initial perturbation decrease with time [7–11]. According to the practical interest, the dependence of λ_{cr} on geometrical and physical parameters of the problem should be analyzed.

For a numerical result, we consider a two-layered film coating with Ni-based top layer [13]. The stress-strain state of the film is defined in the first-order approximation of the perturbation method. Table 1 shows the relative difference of critical wavelengths λ_{cr}^{-} and λ_{cr}^{+} that was calculated for compressive and tensile residual stresses, consequently. The shear modulus of the top layer is $\mu_1 = 100$ GPa, Poisson's ratios are $\nu_1 = \nu_2 = \nu_3 = 0.3$, the surface energy is $\gamma = 1$ J/m^2, the volume to surface diffusion ratio is $D = 10^{-25}$ m^2, the atomic volume is $\Omega = 4.29 \times 10^{-29}$ m^3, and value of residual stress in the top layer is equal to $\sigma_1 = \pm 700$ MPa.

The influence of residual stresses σ_1 and diffusional coefficient D on the critical value of perturbation wavelength λ_{cr} is shown in Fig. 1a and 1b, correspondingly, for different stiffness ratios $r_j = \mu_j / \mu_{j+1}, \; j = \overline{1,2}$ in the case of $h_1 = h_2 = 0.08$ μm. To plot the curves of Fig. 1a, we take the value of the diffusional coefficient equal to $D = 10^{-25}$ m^2. For graphs of Fig. 1b, residual stresses in the top layer are equal to $\sigma_1 = \pm 700$ MPa.

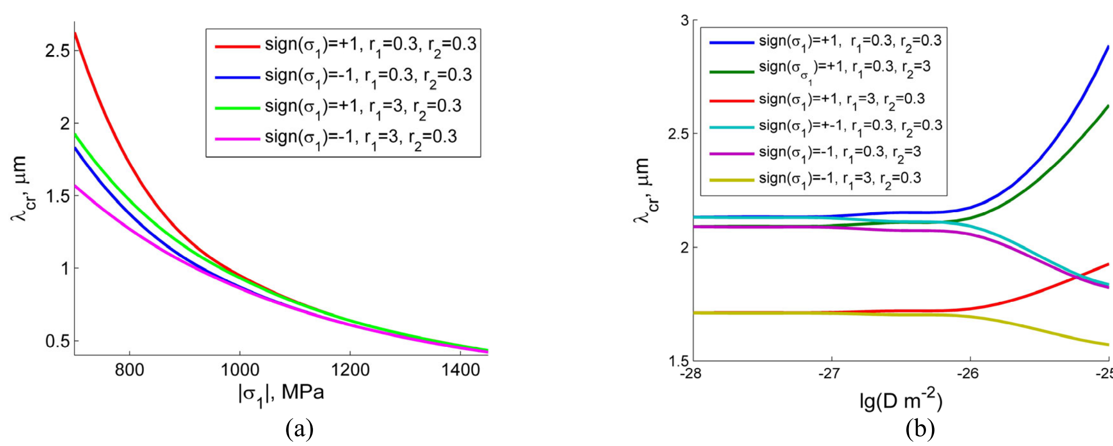

FIGURE 1. Influence of residual stress σ_1 (a) and diffusion coefficient D (b) on critical undulation wavelength λ_{cr} in the case of different values of stiffness ratios $r_1 = \mu_1/\mu_2$ and $r_2 = \mu_2/\mu_3$

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