Morphological stability of thin film materials during annealing

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Abstract

Multilayer thin film materials are extensively used in engineering systems to accomplish a wide range of specific functions. The layered structure could be used for improving mechanical, optical, electrical, magnetic and thermal properties of microelectronic devices. However, multilayer thin film structures are inherently stressed owing to lattice mismatch between different layers. Similar to other stressed solids, such materials can self-organize a surface shape with mass redistribution to minimize a total energy. But the morphological stability is very important in fabrication of defect-free microelectronic devices. In this paper, we present a model of surface pattern formation in multilayer thin film structure with an arbitrary number of layers by considering combined effect of volume and surface diffusion. Based on Gibbs thermodynamics and linear theory of elasticity, we design a procedure for constructing a governing equation that gives the amplitude change of surface perturbation. A parametric study of this equation leads to the definition of a critical undulation wavelength which stabilizes the surface. As an application of presented solution, we analyze the surface stability of two-layered film under different conditions.

1 Introduction

Nowadays it's a well-established phenomena that during film deposition and subsequent thermal processing the film surface evolves into an undulating profile. Surface roughness affects on many important aspects in the engineering application of thin film materials such as wetting, heat transfer, mechanical, electromagnetic and optical properties. Numerous experimental results demonstrate that surface effects become important in mechanical behavior of nanosized structural elements. Analyzing a regular surface patterns in mono- and multilayer film coatings, it was found that even a slight undulation in surface morphology can lead to nucleation of microcracks and film delamination. It should be noted, that there are some positive aspects of surface roughening. For instance, control annealing of thin film causes to break up it to nanosized islands, which exhibit unusual electrical and optical properties. So, to accurately control the morphological surface modifications at the micro- and nanoscale and improve manufacturing techniques, we need to model this process to gain a better theoretical understanding.

2 Problem formulation

Consider an isotropic multilayer film coating of a total thickness $h_f = \sum\limits_{r=1}^{N} h_r$, 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 (see Fig. 1). The layer of thickness h_j has Poisson's ratio ν_r and shear modulus μ_r.

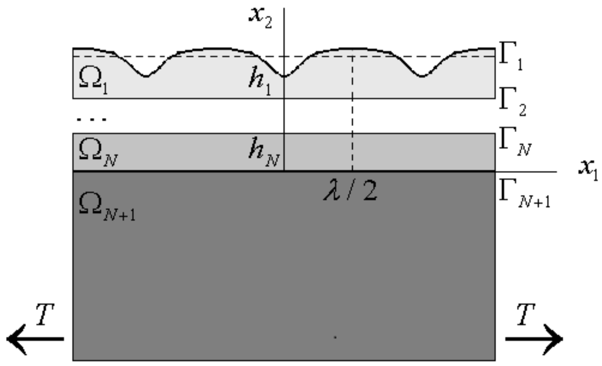

Figure 1: Multilayer film coating with curved surface.

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

$$\Omega_{N+1} = \left\{ z: \ x_2 < 0, \, x_1 \in \mathrm{R}^1 \right\}. \tag{1}$$

The coating is modeled as coherently bonded strips Ω_r

$$\begin{gathered} \Omega_r = \left\{ z: \ H_{r+1} < x_2 < H_r, \ x_1 \in \mathrm{R}^1 \right\}, \\ H_N = h_N, \ H_{N+1} = 0, \ H_r = H_{r+1} + h_r, \ r = \overline{2, N} \end{gathered} \tag{2}$$

with rectilinear boundaries

$$\Gamma_r = \left\{ z: \ z \equiv z_r = x_1 + iH_r \right\}, \ r = \overline{2, N+1}. \tag{3}$$

Taking into account the results of experimental studies, we assume that the film surface has an arbitrary small perturbation which changes with time τ through the mass transport

$$\begin{gathered} \Gamma_1 = \left\{ z: \ z \equiv z_1 = x_1 + i \left[H_1 + g(x_1, \tau) \right] \right\}, \\ g(x_1, \tau) = \sum_{n=1}^{+\infty} A_n(\tau) \cos kx_1, \ A_n(0) = a_n, \\ \max_n \left| A_n(\tau) \right| / \lambda = \varepsilon(\tau) \ll 1 \ \forall \tau, \ k = 2\pi n / \lambda. \end{gathered} \tag{4}$$

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

$$\sigma(z_1) = 0, \ z_1 \in \Gamma_1, \tag{5}$$

$$\Delta u(z_r) = u^+ - u^- = 0, \ \Delta\sigma(z_r) = \sigma^+ - \sigma^- = 0, \tag{6}$$

$$\sigma_{22}^\infty = \sigma_{12}^\infty = 0, \ \sigma_{11}^\infty = T, \ \omega^\infty = 0. \tag{7}$$

In Eqs. (5)–(7), $u = u_1 + iu_2$, $\sigma = \sigma_{nn} + i\sigma_{nt}$; u_1, u_2 are displacements along corresponding axes of Cartesian coordinates x_1, x_2; σ_{nn}, σ_{nt} are components of the stress vector σ at the area with unit normal $\mathbf{n}$ in the local Cartesian coordinate system n, t (vector $\mathbf{n}$ is perpendicular to the boundary Γ_1 in Eq. (5) and the interface Γ_r in Eq. (6); $u^\pm = \lim\limits_{z \to z_r \pm i0} u(z)$, $\sigma^\pm = \lim\limits_{z \to z_r \pm i0} \sigma(z)$, $z_r \in \Gamma_r$, $r = \overline{2, N+1}$; $\sigma_{\alpha\beta}^\infty = \lim\limits_{x_2 \to -\infty} \sigma_{\alpha\beta}$, $\omega^\infty = \lim\limits_{x_2 \to -\infty} \omega$; $\sigma_{\alpha\beta}$ $(\alpha, \beta = 1, 2)$ are the components of the stress tensor in the axes x_1, x_2; ω is the rotation angle of a material particle.

As it was mentioned above, the analysis of morphological instability is based on combined effect of surface and volume diffusion that are assumed to take place in the region close to the free surface Γ_1. Following Panat et al.[1], the normal velocity of the surface can be computed as

$$\begin{aligned} \frac{\partial g(x_1, \tau)}{\partial \tau} &= K_s \frac{\partial^2}{\partial x_1^2} \left[U(x_1, \tau) - \gamma \frac{\partial^2 h(x_1, \tau)}{\partial x_1^2} \right] + \\ &+ K_v k \left[\gamma \frac{\partial^2 h(x_1, \tau)}{\partial x_1^2} + \Delta P(x_1, \tau) \right], \end{aligned} \tag{8}$$

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 sel-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 perturbated film surface, 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 the surface evolution equation (8), we solve the corresponding boundary-value problem of plane elasticity for multiply connected domain $\Omega = \bigcup\limits_{r=1}^{N+1} \Omega_r$ under boundary conditions (5)–(6) and conditions at infinity (7).

3 Perturbation Solution

In accordance with the superposition technique [2, 3], the solution of formulated problem of linear elasticity (1)–(7) is represented as

$$G(z) = \begin{cases} G_k^k(z, \eta_k) + G_k^{k+1}(z, \eta_k), & z \in \Omega_k, \\ \\ G_{N+1}^{N+1}(z, \eta_{N+1}), & z \in \Omega_{N+1}, \end{cases} \tag{9}$$

where $k = \overline{1, N}$.

In Eq. (9), the following notations are introduced

$$G(z, \eta_j) = \begin{cases} \sigma(z), \ \eta_j = 1, \\ & z \in \Omega_j, \\ -2\mu_j \upsilon(z), \ \eta_j = -\kappa_j, \end{cases} \tag{10}$$

$$G_j^r(z,\eta_j)=\begin{cases}\sigma^r(z),\ \eta_j=1,\\ -2\mu_j\upsilon^r(z),\ \eta_j=-\kappa_j,\end{cases}\quad z\in\Omega_j. \tag{11}$$

Here, $\kappa_j=3-4\nu_j$; $\upsilon=\dfrac{du}{dz}$; $\upsilon^r=\dfrac{du^r}{dz}$; σ^r and u^r are the stress and displacement vectors in the problem with number r, similar to σ and u; $r,j=\overline{1,N+1}$. The derivative is taken along the area with normal $\mathbf{n}$, i.e. in the direction of the axis t.
In the first problem, it is supposed that unknown self-balanced periodic load p is applied to the periodic curvilinear boundary Γ_1 of the homogeneous half-plane with the same period λ. The longitudinal load at infinity is equal to T_1^1.
In the problem r ($r=\overline{2,N+1}$), the coupled deformation of two dissimilar half-planes Θ_{r-1} and Θ_r with elastic properties of the corresponding phases Ω_{r-1} and Ω_r is caused by the unknown jumps of tractions $\Delta\sigma^r$ and displacements Δu^r at the rectilinear interface Γ_r under longitudinal remote load T_j^r in Θ_j ($j=r-1,\,r$).
Quantities T_1^1, T_{r-1}^r, T_r^r ($r=\overline{2,N+1}$) are found from recurrence relations which follow from conditions (6) and equations $\Delta\sigma^r=\Delta u^r=0$ corresponding to the case of the coating with the flat surface.
Boundary conditions (5) and (6) at Γ_i lead to the system of boundary equations for unknown functions p, $\Delta\sigma^r$ and Δu^r.
According to papers [2, 3], the stresses σ^r and displacements u^r are related to Goursat-Kolosov complex potentials Φ_j^r and Υ_j^r by the equality

$$\begin{aligned}G_j^r(z,\eta_j)&=\eta_j\Phi_j^r(w_k)+\overline{\Phi_j^r(w_k)}-\big(\Upsilon_j^r(\overline{w_k})+\\&+\overline{\Phi_j^r(w_k)}-(w_k-\overline{w_k})\,\overline{\Phi_j^{r\prime}(w_k)}\big)\,e^{-2i\alpha},\ z\in\Omega_j,\end{aligned} \tag{12}$$

where α is the angle between axis t of the local coordinates n,t and axis x_1, the prime denotes differentiation with respect to the argument; $r,j=\overline{1,N+1}$; $w_1=z+i(g(x_1)-H_1), w_k=z+iH_k$, $k=\overline{r-1,r}$, $k\neq j$.
Following boundary perturbation technique, we expand functions Φ_j^r, Υ_j^r and p in power series of small parameter ε

$$p(z_1)=\sum_{n=0}^{\infty}\frac{\varepsilon^n}{n!}p_n(z_1),\ \Phi_j^r(w_k)=\sum_{n=0}^{\infty}\frac{\varepsilon^n}{n!}\Phi_{jn}^r(w_k),\ \Upsilon_j^r(\overline{w_k})=\sum_{n=0}^{\infty}\frac{\varepsilon^n}{n!}\Upsilon_{jn}^r(\overline{w_k}). \tag{13}$$

And boundary values of functions Φ_{1n}^1, Υ_{1n}^1 and p_n at Γ_1 into Taylor series in the vicinity of the line $\operatorname{Im} w_1=0$, i.e. $z=iH_1$, considering x_1 as parameter

$$\begin{aligned}&\Phi_{1n}^1(w_1)=\sum_{m=0}^{\infty}\frac{[i\varepsilon f(x_1)]^m}{m!}\Phi_{1n}^{1(m)}(x_1),\Upsilon_{1n}^1(\overline{w_1})=\sum_{m=0}^{\infty}\frac{[-i\varepsilon f(x_1)]^m}{m!}\Upsilon_{1n}^{1(m)}(x_1),\\&p_n(z_1)=\sum_{m=0}^{\infty}\frac{[i\varepsilon f(x_1)]^m}{m!}p_n^{(m)}(x_1).\end{aligned} \tag{14}$$

In view of relation $\varepsilon f'(x_1) = \mathrm{tg}\,(\alpha_1)$ and condition $|\varepsilon f'(x_1)| < 1$, one can write

$$e^{-2i\alpha_1} = 1 + 2\sum_{m=0}^{\infty} \left(-i\varepsilon f'(x_1)\right)^{m+1}. \tag{15}$$

Based on the solution of Riemann-Hilbert problem for holomorphic functions $\Phi^r_{1n}(w_1)$, $\Upsilon^r_{1n}(\overline{w_1})$ $(r = \overline{1, N+1})$, representations (12)-(15) allows us to transform the system of boundary equations for unknown functions p, $\Delta\sigma^r$ and Δu^r into Fredholm integral equations of the second kind in expansion coefficients σ^r_n and v^r_n $(r = \overline{2, N})$ and their conjugates

$$\begin{gathered}
\Delta\sigma^r_n(x_1) + \int\limits_{-\infty}^{+\infty} K_{r1}(x_1,\xi)\Delta\sigma^r_n(\xi)d\xi + \int\limits_{-\infty}^{+\infty} K_{r2}(x_1\xi)\overline{\Delta\sigma^r_n(\xi)}d\xi + \\
+ \int\limits_{-\infty}^{+\infty} K_{r3}(x_1,\xi)\Delta v^r_n(\xi)d\xi + \int\limits_{-\infty}^{+\infty} K_{r4}(x_1,t)\overline{\Delta v^r_n(t)}dt = H^r_{1n}(x_1), \\
\Delta v^r_n(x_1) + \int\limits_{-\infty}^{+\infty} K_{r5}(x_1,\xi)\Delta\sigma^r_n(\xi)d\xi + \int\limits_{-\infty}^{+\infty} K_{r6}(x_1,\xi)\overline{\Delta\sigma^r_n(\xi)}d\xi + \\
+ \int\limits_{-\infty}^{+\infty} K_{r7}(x_1,\xi)\Delta v^r_n(\xi)d\xi + \int\limits_{-\infty}^{+\infty} K_{r8}(x_1,\xi)\overline{\Delta v^r_n(\xi)}d\xi = H^r_{2n}(x_1).
\end{gathered} \tag{16}$$

Here the kernels $K_{rj}(x_1,\xi)$, $j = \overline{1, 8}$ are the same for every order of approximation and belong to the class of continous functions.The right hand sides $H^r_{1n}(x_1)$, $H^r_{2n}(x_1)$ are known continuous functions which depend on solutions of all previous approximations.

Periodicity of a surface perturbation (4) makes it possible to solve the problem in a form of Fourier series as in the case of the single layer coating [2, 3, 4]

$$\Delta\sigma^r_n(x_1) = \sum_{k=-\infty}^{+\infty} A^r_{kn}E_k(x_1),\ \ \Delta v^r_n(x_1) = \sum_{k=-\infty}^{+\infty} B^r_{kn}E_k(x_1) \tag{17}$$

where $A^r_{kn}\ B^r_{kn} \in C$, $E_k(x_1) = \exp(b_k x_1)$, $b_k = \dfrac{2\pi i k}{\lambda}$.

Functions $H^r_{1n}(x_1)$ and $H^r_{2n}(x_1)$ are periodic as well and can be represented by Fourier series with known coefficients

$$\begin{gathered}
H^r_{1n}(x_1) = \sum_{k=-\infty}^{+\infty} C^r_{kn}E_k(x_1),\ \ C^r_{kn} = \frac{1}{\lambda}\int\limits_{-\lambda/2}^{\lambda/2} H^r_{1n}(t)E_{-k}(x_1)dt, \\
H^r_{2n}(x_1) = \sum_{k=-\infty}^{+\infty} D^r_{kn}E_k(x_1),\ \ D^r_{kn} = \frac{1}{\lambda}\int\limits_{-\lambda/2}^{\lambda/2} H^r_{2n}(t)E_{-k}(x_1)dt
\end{gathered} \tag{18}$$

Using expansions (17) and (18), the system of $2N-2$ integral equations (16) is reduced to the linear system of algebraic equations in the unknown coefficients A^r_{kn}, B^r_{kn}.

4 Stability conditions

Using the method described above, a stress and strain distribution modified by surface perturbation (4) is obtained in the first-order approximation

$$\begin{aligned} \sigma_{ij}(x_1,\tau) &\approx \sigma_{ij(0)}(x_1,\tau)+\varepsilon(\tau)\sigma_{ij(1)}(x_1,\tau), \\ \varepsilon_{ij}(x_1,\tau) &\approx \varepsilon_{ij(0)}(x_1,\tau)+\varepsilon(\tau)\varepsilon_{ij(1)}(x_1,\tau). \end{aligned} \tag{19}$$

Substituting obtained equations for the elastic strain energy U at the wavy surface and the hydrostatic pressure variation ΔP into Eq. (8), equating coefficients of $\cos(kx_1)$ and then integrating over the time we derive the governing equations which give the exponential growth of each Fourier wavemodes A_n with time [5]

$$\ln\left(\frac{A_n(t)}{a_n}\right)=P_n(\lambda,h_1,\ldots,h_N,\mu_1,\ldots,\mu_{N+1},\nu_1,\ldots,\nu_{N+1},\gamma,D,T)\tau, \tag{20}$$

while $\lambda>\lambda_{cr}$, where critical wavelength λ_{cr} is determined from equations

$$P_n(\lambda,h_1,\ldots,h_N,\mu_1,\ldots,\mu_{N+1},\nu_1,\ldots,\nu_{N+1},\gamma,D,T)=0,\ D=\frac{D_vC_v}{D_sC_s}. \tag{21}$$

As an example, we consider two-layered film structure where the surface undulation is specified by the periodic function [4]

$$f(x_1)=\frac{\lambda}{d}\left[Imctg(\frac{\pi x_1}{\lambda}-iy)-1\right],\ \ d=Imctg(iy)+1, \tag{22}$$

here the real quantity $y\in(0,+\infty)$ plays the role of the parameter determining the surface shape. Fig. 2 presents the film surface relief for y = 0.5 and 5.

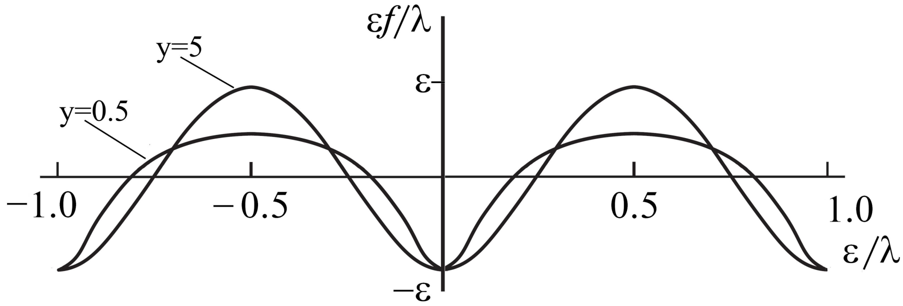

Figure 2: The surface shape with different values of parameter y.

Table 1 shows the critical values of surface perturbation wavelength where shear modulus is $\mu_1=100GPa$, Poisson ratios are $\nu_1=\nu_2=\nu_3=0.3$, surface energy is $\gamma=1J/m^2$, volume to surface diffusion ratio is $D=10^{-25}m^2$ and atomic volume is $\Omega=4.29\times10^{-29}m^3$. Young modulus ratios E_1/E_2, E_2/E_3; thicknesses of layers h_1, h_2 and parameter y are varying in Eq. (22).
As one can see from the table, the surface shape has most significant effect on critical wavelength. The relative difference of critical values in the case of $y=0.5$ and $y=5$

Table 11: The critical perturbation wavelength for various system parameters.

E_1/E_2			0.3	0.3	3	3
E_2/E_3			0.3	3	0.3	3
$h_1, \mu m$	$h_2, \mu m$	y	$\lambda_{cr}, \mu m$			
0.6	0.6	0.5	1.287	1.287	1.248	1.248
		5	2.887	2.625	1.926	1.911
1.2	0.6	0.5	1.264	1.264	1.263	1.263
		5	2.190	2.186	2.100	2.098
0.6	1.2	0.5	1.287	1.287	1.247	1.247
		5	2.728	2.694	1.917	1.916

Table 12: The effect of different longitudinal load T signs.

E_1/E_2			0.3	0.3	3	3
E_2/E_3			0.3	3	0.3	3
$h_1, \mu m$	$h_2, \mu m$	y	$(\lambda_{cr}^{+} - \lambda_{cr}^{-})/\lambda_{cr}^{+}$			
0.6	0.6	0.5	0.153	0.152	0.138	0.138
		5	0.365	0.306	0.185	0.180
1.2	0.6	0.5	0.144	0.144	0.143	0.143
		5	0.242	0.240	0.215	0.214
0.6	1.2	0.5	0.153	0.153	0.138	0.138
		5	0.330	0.322	0.182	0.181

ranges from 53% to 125% for different parameters. In the case of a sinusoidal surface ($y = 5$), effect of Young modulus ratios E_1/E_2 and E_2/E_3 and thicknesses of layers h_1 and h_2 are also considerably (33%, 10%, 25%, respectively). However, in the case of $y = 0.5$ variation of these parameters has insignificant effect on the result.
The contribution of volume diffusion depends on the sign of the stress T [1]. The relative differences of critical wavelengths λ_{cr}^{-} and λ_{cr}^{+} for compressive and tensile stresses, consequently, are presented in the Table 2. According to the results, the load sign has greater influence in the case of the soft film coating.

5 Conclusion

In the present study, we designed the theoretical model of multilayer thin film coating in order to analyze the stability of free surface against diffusional perturbations. Using the complex variable representations, superposition method and boundary perturbation technique, the original boundary value problem is reduced to the successive solution of the set of Fredholm integral equations, which is given in the terms of Fourier series. As a result, governing equation is derived and gives the amplitude of morphological evolution as a function of time.

Acknowledgements

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