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Stability analysis of solid morphology incorporating surface elasticity and surface tension

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ABSTRACT

This paper is primarily focused on exploring the morphological instability conditions inherent in nanostructured solid surfaces. Employing the constitutive equations of Gurtin–Murdoch model, we examine how surface elasticity and surface tension exert their influence on surface relief formation. Within this framework, we posit that the surface instability of the solid surface is instigated by surface diffusion processes propelled by the nuanced interplay of surface and bulk energy across the undulated surface. To distinguish the strain field along the undulated surface, we navigate the solution space of the plane elasticity problem, accounting for plane strain conditions. Our investigation tracks the linearized evolution of the surface, capturing the change in the amplitude of surface perturbations with time. Thus, the presented linear stability analysis sheds light on the precise conditions that initiate the early-stage increase in surface relief amplitude. This nuanced exploration provides not only a theoretical foundation, but also practical insights into the intricate mechanisms governing the morphological stability of nanostructured solid surfaces.

KEYWORDS

morphological stability • surface elasticity • surface diffusion • plane strain

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Introduction

The study of nanostructured materials is important for the development of modern electronic and optoelectronic devices. The roughness of free surfaces and interfacial boundaries has a significant effect on optical properties [1,2]. It is also used to enhance the coupling between electronic circuit components when creating flexible electronics [3]. Additionally, the formation of the surface relief can be applied to produce quantum dots [4]. However, the created relief may be unstable and evolve during the manufacture and operation of devices. Morphological instability can lead to decreased reliability or functionality of the devices based on patterned structures, especially in applications where high levels of mechanical stress are present. Stress concentrations at valley regions of an undulated surface can aid the nucleation of defects [5,6]. Therefore, in addition to other processes [7–9], morphological instability is one of the key factors in the stress-assisted degradation of materials. This highlights the significance of understanding the self-organization of solid surfaces.

Mullins [10] conducted pioneering research on the morphological instability of free solid surfaces influenced by surface diffusion, particularly observing the formation of surface grooves at the grain boundaries of a heated polycrystal. Later studies revealed the morphological instability of the stressed solid bodies due to diffusion perturbations with wavelengths exceeding a critical value [11]. The determination of this critical wavelength is based on the ratio between the surface energy and the energy of elastic deformation calculated on the surface.

A series of studies were conducted to analyze the stability of free and interfacial surfaces in solids with different topological defects [12–14]. However, in most works dedicated to studying surface/interface morphological instability, the influence of surface and interface elasticity was neglected, as it was considered to be small compared to bulk elastic behavior. Nevertheless, in nanostructured materials, the ratio of surface to volume increases, which leads to an increase in the influence of surface deformation [15]. So, to accurately predict the conditions under which morphological instability occurs, we need to take surface elasticity into account.

In this paper, we examine the phenomenon of surface nanosized relief instability, considering its surface elastic properties and surface tension. Our study is based on the complete model of surface elasticity developed by Gurtin and Murdoch [16] and the Asaro–Tiller–Grinfeld model of morphological instability [11,17,18].

Problem formulation

We consider film coating under plane strain loading (Fig. 1). It is assumed that the film thickness significantly exceeds the surface relief period. Therefore, we neglect the deformation of the substrate and the interfacial boundary and come to the 2D elastic problem for a homogeneous half-plane B with a curvilinear boundary S, which profile is described by an arbitrary periodic function f:

$$S = \{z: z \equiv \zeta = x_1 + i\varepsilon(\tau) f(x_1)\}, \quad B = \{z: x_2 < \varepsilon(\tau) f(x_1)\}, \quad z = x_1 + ix_2, \quad i^2 = -1,$$
$$f(x_1) = f(x_1 + a), \quad \max|f(x_1)| = a, \quad \varepsilon(\tau) = \frac{A(\tau)}{a} \forall\, \tau, \quad A(0) = A_0. \qquad (1)$$

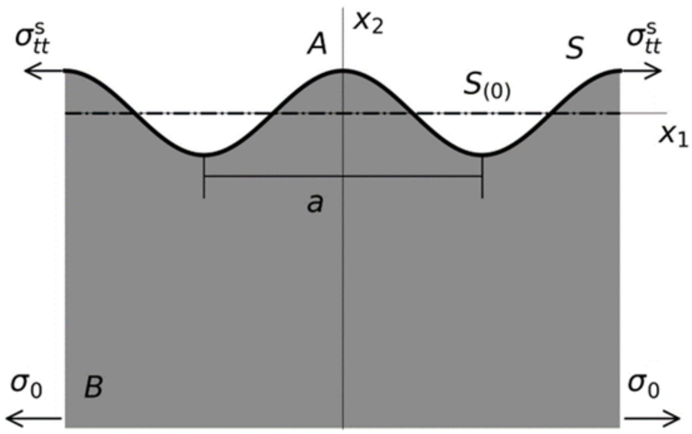

Fig. 1. The model of a solid with a slightly undulated surface

According to the principles of Gurtin–Murdoch surface elasticity theory, the surface domain is considered as an exceptionally thin layer that firmly adheres to the bulk without any slipping. The continuity of displacements across the surface region is provided by the following conditions:

$$u_s(\zeta) = u(\zeta), \quad \zeta \in S, \tag{2}$$

where $u^s = u_1^s + iu_2^s$, $u = u_1 + iu_2$; u_1^s, u_2^s and u_1, u_2 are the displacements of the surface and bulk phases along Cartesian axes x_1 and x_2.

The mechanical equilibrium conditions on a curved solid surface are captured by the generalized Young–Laplace equations [19]:

$$\sigma(\zeta) = \gamma_0\kappa + \left[M\kappa\mathrm{Re}\frac{\partial u}{\partial \zeta} + \gamma_0\mathrm{Im}\left(\frac{\partial^2 u}{\partial \zeta^2}e^{i\alpha_0}\right)\right] + i\left[M\mathrm{Re}\left(\frac{\partial^2 u}{\partial \zeta^2}e^{i\alpha_0}\right) - \gamma_0\kappa\mathrm{Im}\frac{\partial u}{\partial \zeta}\right], \tag{3}$$

where $\sigma(\zeta) = \sigma_{nn}(\zeta) + i\sigma_{nt}(\zeta)$ is the complex stress vector, σ_{nn} and σ_{nt} are the components of bulk stress tensor, defined in the Cartesian coordinates (n, t) ($\boldsymbol{n}$ is a normal to S), γ_0 is residual surface stress (surface tension), $M = \lambda_s + 2\mu_s$ is surface stiffness, λ_s and μ_s are the Lame parameters for the surface domain, κ is the local curvature of S, α_0 is the angle between the tangent to S and x_1-axis at the point ζ.

The boundary conditions at infinity are defined as

$$\lim_{x_2\to-\infty} \omega = \lim_{x_2\to-\infty} \sigma_{22} = \lim_{x_2\to-\infty} \sigma_{12} = 0, \lim_{x_2\to-\infty} \sigma_{11} = \sigma_0, \tag{4}$$

where ω is the rotation angle, σ_{ij} $(i, j = \{1,2\})$ are components of stress tensor in Cartesian coordinates (x_1, x_2) and the longitudinal stress σ_0 may means either misfit stress or mechanical loading.

As it was mentioned in introduction, the surface profile of a stressed solid may change under the influence of surface diffusion due to nonuniform distribution of chemical potential. So, the surface atoms are moving from a region with high chemical potential to a region with a lower one, i.e. atomic flow along the surface is proportional to the gradient of chemical potential. According to [18], the chemical potential is defined as the sum of bulk strain elastic energy and surface energy. The mass conservation law leads to the following differential equation, which describes the change of surface profile $g(x_1, \tau) = \varepsilon(\tau)f(x_1)$ over the time [18,20]:

$$\frac{\partial g(x_1,\tau)}{\partial \tau} = K_s h(x_1, \tau)\frac{\partial^2}{\partial s^2}[U(\zeta, \tau) - \kappa(\zeta, \tau)U_s(\zeta, \tau)],\ K_s = D_s C_s \Omega^2/(k_b T), \tag{5}$$

where U is the strain elastic energy density and U_s is the surface energy, h is the metric coefficient, D_s is the self-diffusivity coefficient; C_s is the number of diffusing atoms per unit area; k_b is the Boltzmann constant, T is the absolute temperature; and s is arc length along S.

To prevent or minimize morphological instability in stressed films, it is crucial to consider various factors such as the elastic properties of bulk and surface phases, the stress levels, and the shape of the initial surface profile. In this paper, our attention was directed towards the examination of the conditions that facilitate the occurrence of morphological instability.

Linear stability analysis

To integrate the evolution equation (5) and establish the stability conditions for a stressed solid surface, we must first calculate the elastic strain energy U and the surface energy U_s. It is important to note that the effect of surface elasticity was assumed to be insignificant in previous studies dedicated to investigating the morphological instability of the surface microrelief. However, to accurately predict the instability conditions for a stressed surface with nanosized relief, it is essential to consider the effect of surface elasticity.

The elastic strain energy U and the surface energy U_s can be expressed as follows [21]:

$$U = \left(\frac{1}{2}\lambda + \mu\right)\left((\varepsilon_{tt})^2 + (\varepsilon_{nn})^2\right) + \lambda\varepsilon_{nn}\varepsilon_{tt} + 2\mu(\varepsilon_{nt})^2, \quad (6)$$

$$U_s = \gamma_0 + \left(\frac{1}{2}\lambda_s + \mu_s\right)(\varepsilon_{tt}^s)^2, \quad (7)$$

where ε_{ij} are the components of bulk and surface stress tensors, respectively, defined in the Cartesian coordinates (n, t) ($\boldsymbol{n}$ is a normal to S), and λ and μ are the Lame constants of the solid B.

Therefore, in order to integrate (5), we have to obtain the stress-strain state near the undulated surface. To achieve this, we solve the corresponding plane strain problem for a homogeneous elastic half-plane with a curved boundary (1) – (4), using the original approach suggested in [19,22].

Here we consider a weak undulation of the surface relief ($\varepsilon \ll 1$), and therefore we calculate components of the strain tensors of the bulk and surface phases as well as metric coefficient h and the surface curvature κ using the linear approximation of the boundary perturbation method [19]:

$$\varepsilon_{ij} = \varepsilon_{ij(0)} + \varepsilon\varepsilon_{ij(1)}, \; \varepsilon_{tt}^s = \varepsilon_{tt(0)}^s + \varepsilon\varepsilon_{tt(1)}^s, \quad (8)$$

$$\kappa(x_1, \tau) = \varepsilon(\tau)f''(x_1), \quad h(x_1, \tau) = 1, \quad (9)$$

where a prime denotes the derivative with respect to the argument.

Substituting Eqs. (6) – (9) into (5), and integrating over the interval $[0;\ x_0]$ ($x_0 \in [0, a/2]$ and $f(x_0) = 0$), we receive an ordinary differential equation that reveals the variation of surface relief amplitude with time:

$$\frac{dA(\tau)}{d\tau}\int_0^{x_0} f(x_1)dx_1 = \frac{A(\tau)K_s}{2}\int_0^{x_0}\frac{d^2}{dx_1^2}\Big[\lambda\left(\varepsilon_{tt(0)}\varepsilon_{nn(1)}(x_1) + \varepsilon_{tt_0}\varepsilon_{nn(1)}(x_1)\right) +$$
$$+ \left(\frac{1}{2}\lambda + \mu\right)\left(2\varepsilon_{tt(0)}\varepsilon_{tt(1)}(x_1) + 2\varepsilon_{nn(0)}\varepsilon_{nn(1)}(x_1)\right) + 4\mu\varepsilon_{nt(0)}\varepsilon_{nt(1)}(x_1) - \quad (10)$$
$$- f''(x_1)\left(\gamma_0 + \left(\frac{1}{2}\lambda_s + \mu_s\right)\left(\varepsilon_{tt(0)}^s\varepsilon_{tt(0)}^s\right)\right)\Big]dx_1.$$

We seek the functions $\varepsilon_{ij(1)}$, $\varepsilon_{tt(1)}^s$ in the form of a Fourier series with unknown coefficients P and Q:

$$\varepsilon_{ij(1)} = \sum_{k=1}^{\infty}\left[P_{(\varepsilon_{ij})k}\sin(b_k x_1) + Q_{(\varepsilon_{ij})k}\cos(b_k x_1)\right], \quad (11)$$

$$\varepsilon_{tt(1)}^s = \sum_{k=1}^{\infty}\left[P_{(\varepsilon_{tt}^s)k}\sin(b_k x_1) + Q_{(\varepsilon_{tt}^s)k}\cos(b_k x_1)\right], \quad (12)$$

where $b_k = 2\pi k/a$.

In the present paper, we do not give explicit expressions for these functions due to their tremendous size, but they can be found using the original algorithm, presented in [19].

The Fourier series approximation is also utilized to represent the known function $f(x_1)$, which describes the surface profile:

$$f(x_1) = \sum_{k=1}^{\infty} R_k\cos(b_k x_1), \quad R_k = \frac{2}{a}\int_{-a/2}^{a/2} f(x_1)\cos(b_k x_1)\,dx_1. \quad (13)$$

The solution of the elasticity problem gives the unknowns components of surface and bulk strain tensors required to determine the amplitude as a function of time. Due to the enormous size of the explicit solution of evolution equation, we write it in the following form:

$$\ln\left(\frac{A(\tau)}{A_0}\right) = \frac{K_s}{J_f}\sum_{k=1}^{\infty} V_k\, k^{-1}\sin(b_k x_0)\,\tau, \; J_f = \sum_{k=1}^{\infty} R_k k^{-1}\sin(b_k x_0). \quad (14)$$

where functions $V_k = V_k(a, \lambda, \mu, \lambda_s, \mu_s, \gamma_0, \sigma_0, R_k)$ are known functions depending on the physical and geometrical parameters of the problem but are not presented here because of their cumbersomeness.

Numerical results

As per the findings of experimental research, it has been determined that the diverse relief configurations, encompassing a cusp-like relief and a smoothly undulated relief, may be occurred in the process of surface rearrangement because of morphological instability. The following function is used to investigate the impact of surface profiles on morphological instability:

$$f(x_1) = -\frac{a}{d}\left[\operatorname{Im}\operatorname{ctg}\left(\frac{\pi x_1}{a} - iy\right) - 1\right], \quad d = \operatorname{Im}\operatorname{ctg}(iy) + 1, \tag{15}$$

where the parameter $y \in (0, +\infty)$ defines the shape of surface profile. We take $y = 2$ for the smoothly undulated surface, and y = 0.7 for cusp-like relief.

For example, we use following bulk Lame parameters for isotropic solid: λ =58.17 GPa, μ =26.13 GPa. Also, we set surface stiffness $M =$ 6.099 N/m and surface tension $\gamma_0 = 1$ N/m, which correspond to aluminum surface Lame parameters calculated in [23] using molecular dynamics. As the thin film systems are often subjected to large stress, typically in giga-Pascal range [5], we consider the values of σ_0 in the range from 1 to 2 GPa.

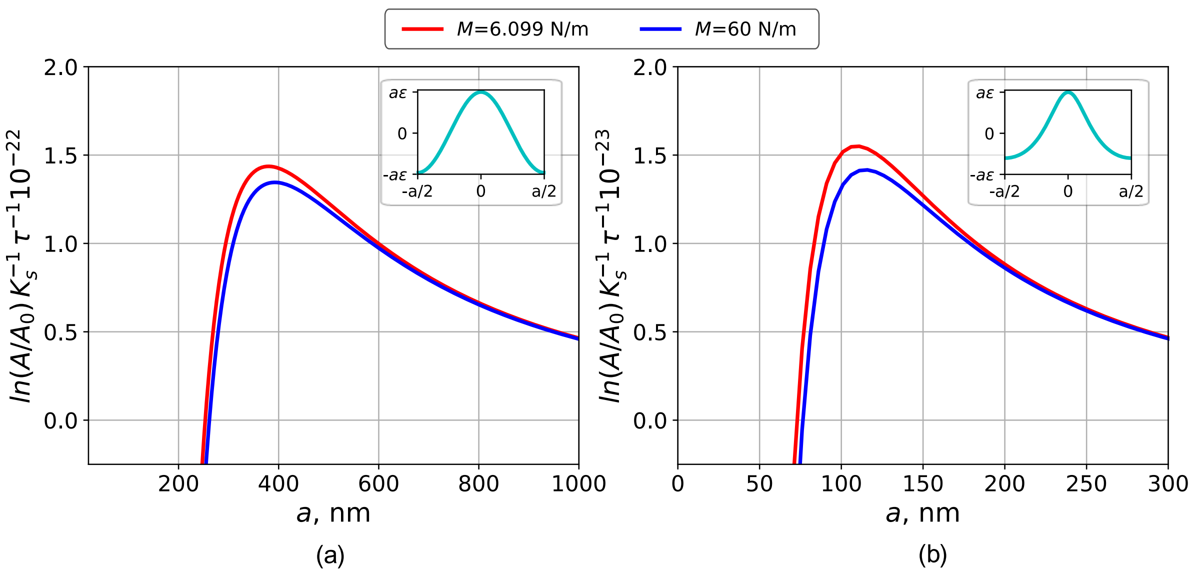

Fig. 2. The dependence of normalized amplitude changes $A(\tau)/A_0$ on the perturbation wavelength a for surface profile parameters y = 2 (a) and y = 0.7 (b)

Figure 2 shows the dependence of the normalized amplitude changes $A(\tau)/A_0$ of the surface relief on the perturbation wavelength a for $\sigma_0 = 1$ GPa, different surface stiffness values $M = \{6.099, 60\}$ N/m (red and blue lines, respectively) and surface profiles with y = 2 (a) and y = 0.7 (b). The abscissas of the intersections of the x-axis and each line lead to the determination of the critical undulation wavelengths a_{cr} corresponding to the thermodynamic equilibrium. If the wavelength of initial undulation $a \in (0, a_{cr})$, the surface relief amplitude will decrease with time, and vice versa, surface undulation will grow in the case $a \in (a_{cr}, \infty)$. The critical perturbation wavelength a_{cr} for the considered parameters and for the case, when surface elasticity is neglected ($M = 0$), are presented in the Table 1.

Table 1. The critical wavelength a_{cr} of considered system for various parameters

Surface stiffness M, N/m	6.099	60	0
Shape parameter a_{cr}, nm			
y = 2	253.3	261.6	252.3
y = 0.7	72.8	76.5	72.2

Figures 3–5 demonstrate the dependence of critical undulation wavelength a_{cr} on surface stiffness M, surface tension γ_0 and misfit stress σ_0. A succinct examination of the results is presented in the conclusions. It is noteworthy that these findings are in good agreement with the outcomes of previous studies where the simplified Gurtin-Murdoch model was considered omitting the normal component of the surface gradient of the displacement field [24,25]. From the results of current study, it follows that accounting of this term doesn't affect the critical value of perturbation wavelength found within the linear analysis of morphological stability. The main goal of linear stability analysis is to predict the conditions under which changes in morphology might occur. Nonetheless, it's worth noting that linear stability analysis is limited to minor changes in surface profile amplitude. To examine how the surface changes over longer periods, it's essential to take into account the nonlinear terms in the surface evolution equation. By keeping terms up to a certain order in the perturbation expansion, we can grasp the behavior at higher amplitudes when cusp-like grooves appear. However, higher amplitudes of undulation profile may significantly amplify the misfit stresses and lead to the nucleation of dislocations [5,6]. The generation of dislocations lowers the local strain energy in the surface layer decreasing the driving force behind relief formation [26].

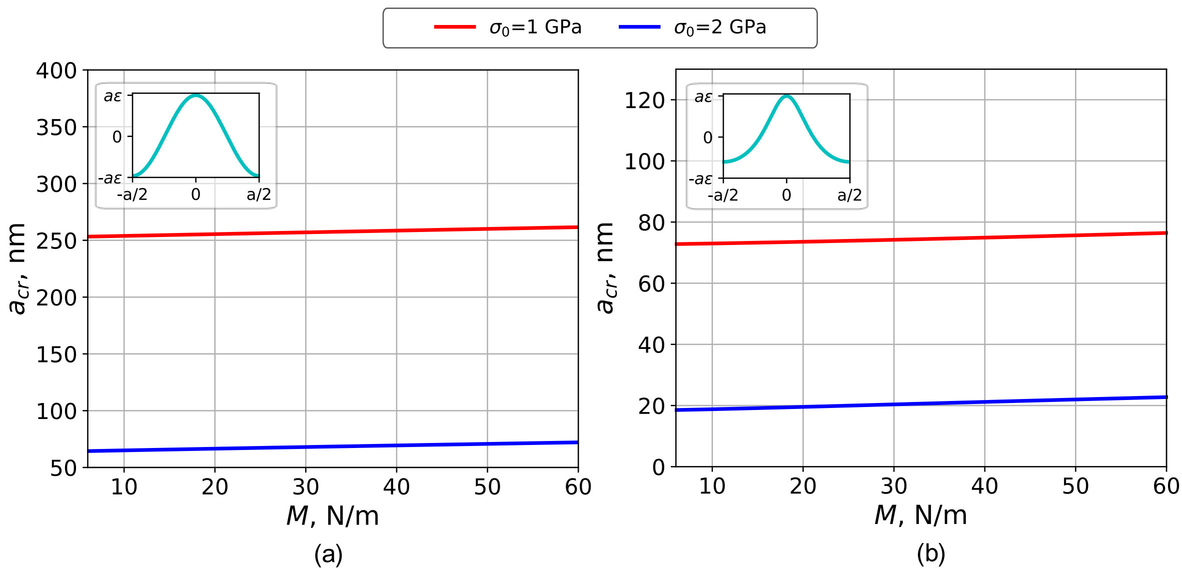

Fig. 3. The effect of surface stiffness M on critical undulation wavelength a_{cr} for surface profile parameters y = 2 (a) and y = 0.7 (b)

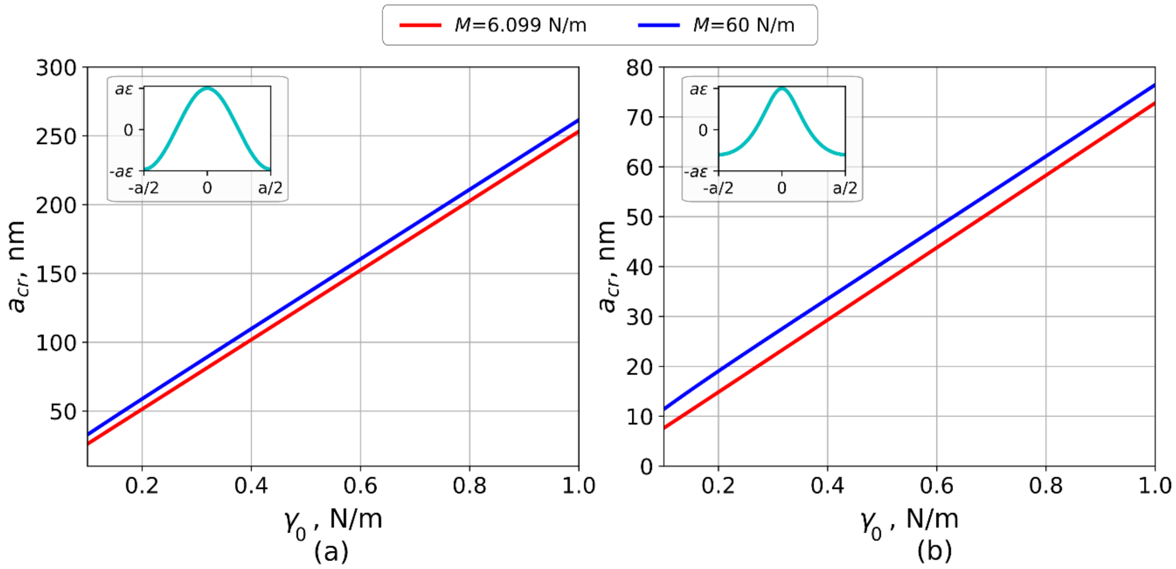

Fig. 4. The effect of surface tension γ_0 on critical undulation wavelength a_{cr} for surface profile parameters y = 2 (a) and y = 0.7 (b)

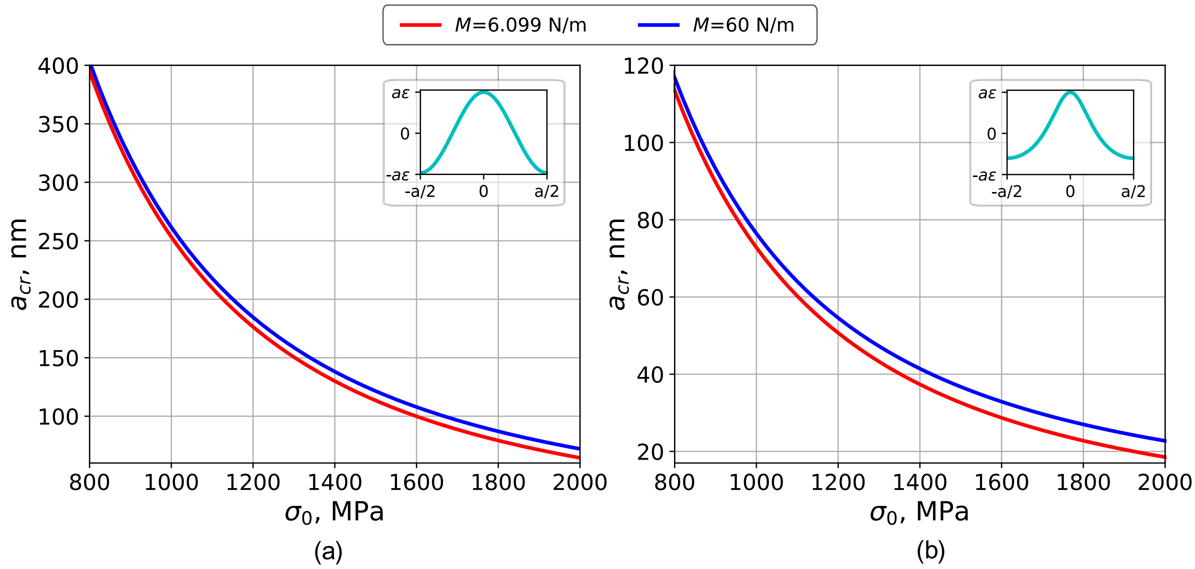

Fig. 5. The effect of longitudinal stress σ_0 on critical undulation wavelength a_{cr} for surface profile parameters y = 2 (a) and y = 0.7 (b)

Conclusions

In this paper, we investigate the joint effect of surface elasticity, tension and shape on the morphological instability in nanopatterned solid films based on Gurtin – Murdoch theory of surface elasticity and the Asaro – Tiller – Grinfeld model of morphological instability. In line with [18], the morphological instability of the film surface results from surface diffusion driven by variations in surface and bulk energy along the undulated solid surface.

Assuming that the film thickness is significantly greater than the initial surface relief wavelength, we defined the elastic strain energy and the surface energy from the solution of the elastic problem for a homogeneous elastic half-plane with curved boundary under plane strained conditions. The solution of the linearized evolution equation allowed us to determine the amplitude of arbitrary periodic surface relief as a function of time. The critical conditions that correspond to surface morphological instability are determined through the analysis of this solution.

We investigated the dependence of the critical undulation wavelength on the misfit stress, surface tension, surface stiffness and the shape of the initial surface relief, and based on the obtained data, we have come to the following conclusions:

– an increase in the curvature radius of the initial surface profile, surface stiffness, surface tension, and a decrease in longitudinal stress result in an augmentation of the critical perturbation wavelength.

– a decrease in the influence of surface stiffness is observed with an increase in surface tension as well as in curvature radius of the initial surface profile, and a decrease in longitudinal stress.

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