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Internal friction in thin films and membranes*

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Abstract

This paper presents experimental results of dynamic measurements of thin films and membranes. Through the dynamic properties of film/substrate or multilayer composites, reflected by their eigen frequencies, the adhesion between film and substrate and between multilayers was also studied. The same approach was used to study *in situ* the stress evolution in growing films. Each addition of a monoatomic layer caused upward stress relaxation, and internal friction measurements indicate concurrent relaxations. The *in situ* dynamic measurement provides unique information on the evolution of growing ultrathin films. Further studies on their surface morphology and microstructure are in progress.

1. Introduction

The field of internal friction in thin films and membranes was introduced and pursued by Berry and Pritchet [1] and reviewed by him in detail [2]. A common feature of thin film dynamic studies was that the film and the substrate were treated as a parallel composite which is perfectly elastically bonded. Under this assumption the internal friction and the resonance frequency of the composite can be expressed as [2]

$$\frac{\omega_c^{-1}-\omega_s^{-1}}{\omega_f^{-1}}=\left(3\frac{E_f}{E_s}-\frac{\rho_f}{\rho_s}\right)\frac{d_f}{d_s}$$

$$\frac{Q_c^{-1}-Q_s^{-1}}{Q_f^{-1}}=3\frac{E_f d_f}{E_s d_s} \quad (1)$$

where $d_{s,f}$ is the thickness of substrate and film, $E_{s,f}$ is the modulus of substrate and film, $\rho_{s,f}$ is the density of substrate and film, $Q_{s,f}^{-1}$ is the internal friction of film and substrate, and $\omega_{c,s}$ is the resonance frequency of the composite and substrate.

According to these equations the choice of a low internal friction substrate material is important to reduce the substrate contribution to the composite internal friction Q_c^{-1}. Fortunately, the most frequently used substrate material is single-crystal Si in which the intrinsic internal friction, thermal anelastic effects excluded, is as low as 10^{-6} [3]. Other materials such as quartz and fused silica can also serve as both low internal friction and good substrate materials [4, 5]. An appropriate film to substrate thickness ratio is also important in order to minimize the dilution of the thicksubstrate. The ratio is usually chosen to be 1/100 to 1/50.

Thin films exhibit very different mechanical properties from bulk material. For example, the yield stresses in thin films can be 200 times larger than those of annealed bulk materials [6]. It appears stimulating to revisit the classical internal friction problems, *e.g.* point defect anelasticity, grain boundary relaxation and phase transformation induced damping, in this new form of material [7–9]. These studies are motivated by potential applications in the micro-electronic industry. A correspondence exists between grain boundary relaxation and electromigration in metallized devices [10]; the film–substrate interface constraint was found to play a crucial role in the thin film martensitic transformation and thus the output efficiency of microactuators [9]. Research on the anelastic relaxation mechanism in thin films also leads to new insight into long-standing problems. An example is the discrepancy between the activation energy of grain boundary diffusion and relaxation [11].

One fundamental problem which has been largely ignored in thin film dynamic studies is the load transfer through the interface. Equation (1), which acts as the basis of assessing thin film internal friction and dynamic properties, was based on the assumption that the stress due to substrate bending was elastically coupled to the film, *i.e.* the film adheres perfectly to the substrate. The practical situation is quite different since adhesion

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is a poorly characterized parameter and difficult to control [12]. Only in a few cases such as molecular epitaxy growth on matched substrates does the film–substrate interface achieve perfect elastic load transfer. In the other extreme, if the interface appears to be viscous, load would not be able to couple mechanically to the film. Thus, in general it must be expected that the contribution of the thin film internal friction to the measured film–substrate composite internal friction depends on the degree of adhesion. In this contribution we demonstrate that such non-elastic load transfer, or imperfect adhesion, does exist. Hence, an understanding of the interface behavior should precede the understanding of thin film internal friction.

Another application of dynamic measurements in the field of thin film materials is to determine the stress in thin films by configuring the film–substrate composite or film into membranes, the resonance frequency of which is determined by the state of stress. Thermomechanical studies of thin membranes have been an active field of research [13–15]. The commonly low internal friction, or high mechanical Q, of membranes offers uniquely high resolution for the detection of internal stresses in thin films. The disadvantage of the vibrating membrane technique is that the measured stress represents the average over the membrane. Whenever surface roughness becomes important or interface adhesion is not perfect, the analysis of the stress components can be complicated. Especially in the case of multilayer membranes, the interface load transfer may control the mechanical response. A review of experiments, on Ni/Cu multilayer membranes is provided in this paper.

Finally, we introduce results of preliminary *in situ* studies of the stress and internal friction during thin film growth. A vibrating membrane device was incorporated into a very high vacuum thin film growth chamber. Using micromachined single-crystal Si membrane substrates, the stress and internal friction resolution can reach mono-atomic add-layer [16]. The new device allows one to follow the film growth kinetics instantaneously. Unique information on the film growth kinetics and dynamics can be obtained.

2. Elasticity and anelasticity of thin films

The dynamical mechanical properties of a thin film–substrate layered composite cantilever, equivalent to a mechanical parallel composite, *i.e.* the eigenfrequencies ω_c and damping Q_c^{-1}, are given by eqn. (2) [17]:

$$\frac{\omega_c^{-1}-\omega_s^{-1}}{\omega_f^{-1}} = \left(3\gamma^2\frac{E_f}{E_s} - \frac{\rho_f}{\rho_s}\right)\frac{d_f}{d_s}$$
$$\frac{Q_c^{-1}-Q_s^{-1}}{Q_f^{-1}} = 3\gamma^2\frac{E_f d_f}{E_s d_s} \tag{2}$$

where $d_{s,f}$ is the thickness of substrate and film respectively, $E_{s,f}$ is the modulus of substrate and film respectively, and $\rho_{s,f}$ is the density of substrate and film, respectively.

Equations (2) reduce to eqn. (1) if $\gamma = 1$ which is the case for perfectly adhering films. However, $\gamma < 1$ for imperfect adhesion [18]. This can be appreciated by considering a film which does not adhere at all to the substrate. In this case it only contributes to the inertia, thereby lowering the eigenfrequency of the substrate cantilever. The constant γ in eqns. (2) would be zero. If the film adheres perfectly it contributes maximally to the restoring force, hence the constant γ would equal unity. Values $0 < \gamma < 1$ signal partial adherence.

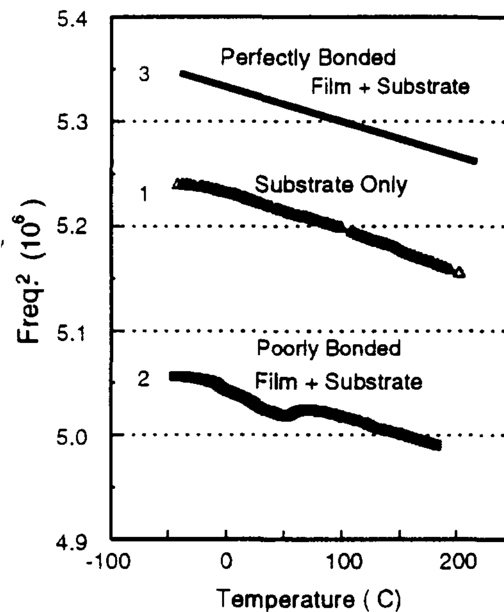


Fig. 1. Comparison of the square of the eigenfrequencies, proportional to the bending stiffness, of an SiO_2/Si reed with and without a 2 μm $Ni_{50}Ti_{50}$ film with the expectation for a perfectly bonded film.

TABLE 1. Coefficients of adhesion γ of amorphous $Ni_{50}Ti_{50}$/SiO_2/Si reeds

Film thickness (μm)	As deposited at 25 °C	As heated to 300 °C	Annealed for 1 h at 300 °C
0.5	0.90	0.90	0.97
1	0.79	0.70	0.74
2	0.77	0.75	0.77

It follows that the adhesion parameter γ can be determined from the difference in the expected and calculated eigenfrequencies of the film–substrate composite. An example can be found in Fig. 1 pertaining to the adhesion of $Ni_{50}Ti_{50}$ on an SiO_2/Si reed. It can be seen from this figure that the eigenfrequency of the $Ni_{50}Ti_{50}/SiO_2$/Si reed is less than that of the substrate, indicating partial adherence; $\gamma \approx 0.7$ in this case. We have studied the variation of the adhesion of these films [18] and found that, as expected, the adhesion deteriorates with increasing film thickness. We also found that it can be improved for very thin films by moderate heat treatments. The results are assembled in Table 1.

3. Elasticity and anelasticity of membranes

The mechanical vibrations of circular membranes have been analyzed [13, 19] and used to characterize stresses of interest in materials science [13, 15, 20]. The eigenfrequencies are given by

$$\omega^2_{m;m,n} = \frac{\alpha^2{}_{m,n}T}{a^2\rho}\left(1+\frac{\alpha^2{}_{m,n}D}{a^2T}+\ldots\right)$$

$$\frac{\alpha^2{}_{m,n}D}{a^2T} \ll 1 \tag{3}$$

where ρ is the areal mass density of the membrane, T is the membrane tension, $D=Eh^3/[12(1-\nu^2)]$, h is the membrane thickness, E is the modulus of the membrane, ν is Poisson's ratio, a is the membrane radius, and $\alpha_{m,n}$ is the eigenvalue of the Bessel function of the mth kind and nth degree.

For the typical values of $\nu=0.3$, $h=5$ μm and $a=5$ mm, the parameter $D/(a^2T) \approx 10^{-7}E/\sigma$. Since $E \approx 100$ GPa and typical internal stresses of metallic and semiconducting substrates range from 50 MPa to 200 MPa, the membrane vibrations are given by the first term of eqn. (3) only, *i.e.*

$$\omega^2_{m;\,m,n} = \frac{\alpha^2{}_{m,n}T}{a^2\rho} \tag{4}$$

The important feature of eqn. (4) is that it does not depend on all on the membrane's modulus, meaning that the contribution of the modulus defect to the damping of membrane vibrations equals zero. Small damping contributions arise from the correction term given by eqn. (3). The situation is different in composite membranes as in this case interfaces may contribute to the damping. It is also possible that the stress itself displays relaxation. Examples of both situations are presented below.

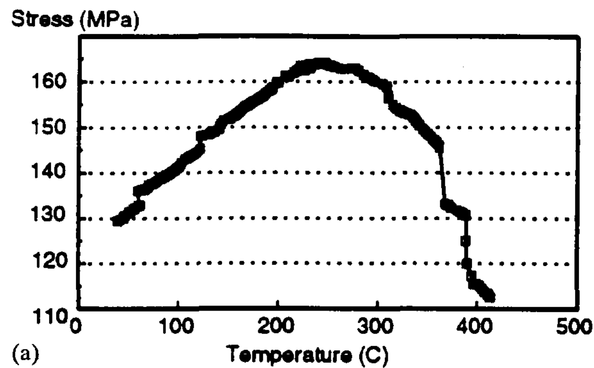
4. Thermal expansion of multilayer membranes

Consider the thermoelasticity of a multilayer membrane on a substrate. The difference in the thermal expansivity of the membrane and substrate will generate a stress in the membrane. It is thus possible to measure the thermal expansivity of the multilayer through the variation of its eigenfrequency with temperature. Figure 2 shows the result of the stress evolution of a Cu/Ni multilayer on Cu. The stress increase signals that the coefficient of thermal expansion of the multilayer is smaller than that of Cu, as must be expected from the expression

$$\alpha_{ave} = \frac{\alpha_{Ni}E_{Ni}d_{Ni}+\alpha_{Cu}E_{Cu}d_{Cu}}{d_{CuNi}E_{ave}} \tag{5}$$

where $\alpha_{ave,Ni,Cu}$ is the thermal expansivity of the multilayer, Ni and Cu, $E_{ave,Ni,Cu}$ is the modulus of the same, and $d_{CuNi,Ni,Cu}$ is the thickness of the multilayer, Ni and Cu.

Quantitatively, eqn. (4) does not agree with the slope $\sigma^{-1}\partial\sigma/\partial T$; it is ten times larger. This difference signals interlayer relaxation as indicated in Fig. 3 which takes into account that the membrane is "anchored" at the bottom of the substrate only and that the interfaces can relax. The average stress in the membrane is then given by



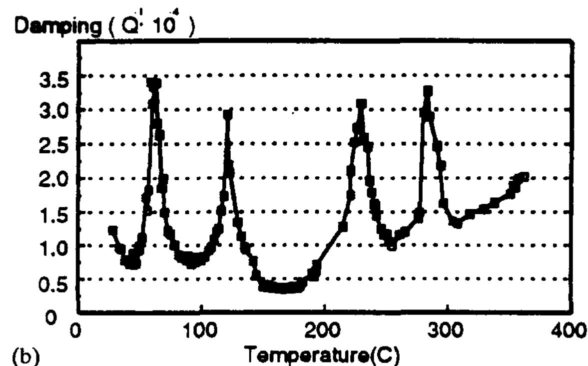


Fig. 2. Temperature dependence of the stress and damping in a electrodeposited 5 μm thick Cu/Ni multilayer membrane with individual layer thicknesses of 2 nm. The substrate is Cu.

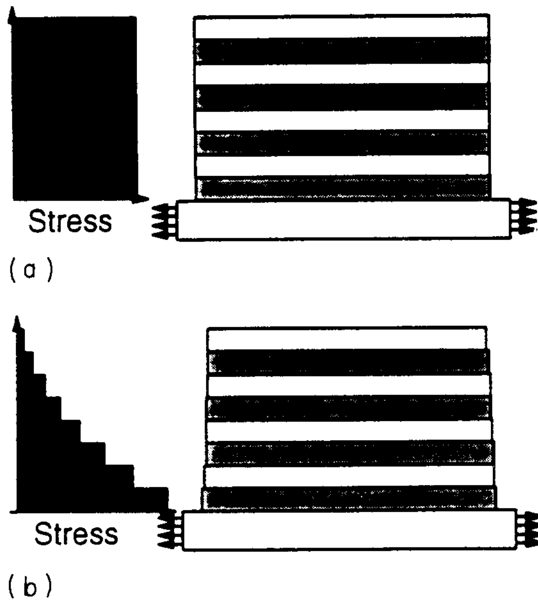


Fig. 3. Stress distribution in a multilayer; top, unrelaxed stress, no interfacial sliding, state (a); bottom, relaxed stress, interfacial sliding occurs, state (b).

$$\sigma_{\text{ave}} = \frac{\sigma + \gamma\sigma + \gamma^2\sigma + \ldots}{n} = \frac{\sigma}{n(1-\gamma)} \tag{6}$$

where σ_{ave} is the average stress in the membrane, γ is the adhesion coefficient, and n is the number of layers.

Comparison of the experimentally determined expansivity with eqn. (6) yields a value of $\gamma \approx 0.99$, *i.e.* the effect is noticeable even for very well adhering multilayers. Equation (6) also implies that the effective thermal expansivity should decrease as the layer density increases. The results of an experiment demonstrating this fact are shown in Fig. 4. It should be noted that the thermal expansivity of Ni calculated on the basis of Fig. 4 agrees with the accepted value. In conclusion, it can be stated that interlayer relaxation can be investigated through studies of membrane vibrations.

The fact that stress relaxation across interfaces in multilayer membranes occurs suggests a source of damping in these stress driven vibrations. Damping will arise whenever a phase difference between the stress driving the vibration and the stress relaxation from state (a) to state (b) exists. The magnitude of this damping must be proportional to the difference in the stresses given by eqns. (5) and (6). It is thus possible to understand the damping in Fig. 2. It occurs whenever stresses are thermally released at the interface, as also indicated by the change in average stress at this temperature.

5. Stress evolution during film growth

It is known that epitaxial films grow coherently to a critical thickness at which the volume elastic energy

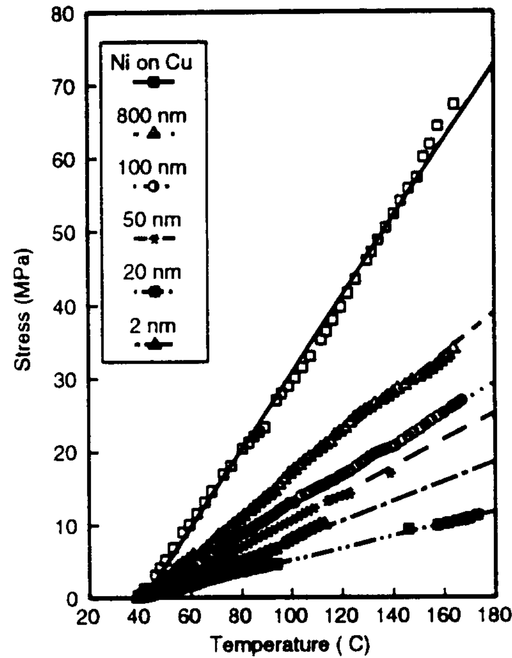


Fig. 4. Evolution of stress in electrodeposited Cu/Ni multilayers of different repeat distances λ.

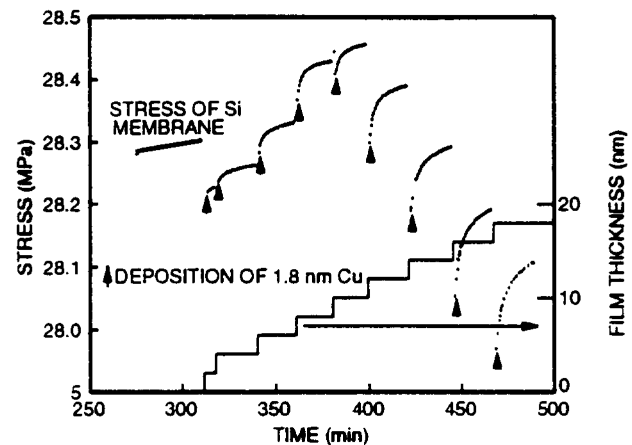


Fig. 5. *In situ* evolution of stress in the course of Cu film growth on an Si membrane.

equals the energy required to create interface dislocations. As these dislocations are created, the stress in the film decreases and it should be possible to detect this change if this event happens in films on membranes. Similar arguments apply if other film–stress reactions occur which involve a balance of the elastic and other fields. The kinetics of this topic was not addressed previously [21–24]. The second example thus deals with the stress generation kinetics and relaxation in ultrathin Cu films deposited onto an Si membrane substrate.

The results of *in situ* measurements during film deposition are shown in Fig. 5. The deposition was performed in a sputtering system with base vacuum 1×10^{-7} Torr. The membrane substrate was carefully

held at the thick edge of the wafer to avoid clamping stresses. The membrane vibrations were excited electrostatically and the electrode and membrane comprise a capacitive distance sensor with a displacement sensitivity of the order of tens of nanometers. The configuration was used to measure the eigenfrequencies of the membrane [16] and through eqn. (4) the stress.

The initial straight line in Fig. 5 indicates the background stress of the bare Si membrane. The deposition was performed in increments of 1.8 nm thick layers and each add-layer was allowed to relax as indicated by the stress and damping measurements. On relaxation of the previous layer another 1.8 nm layer was deposited. Figure 5 summarizes the results. In it, the start and, on the time scale of this figure, end of each deposition are indicated by an arrow and the progress of the deposition is shown by the staircase of the step increase of the total thickness. It can be seen in Fig. 5 that the average stress at the start of each incremental deposition reaches a maximum and that the stress relaxation is always positive.

Figure 6 shows an analysis of the stress relaxation, $\sigma(t)$-σ_i, where σ_i represents the initial stress at the termination of the deposition of each add layer. The lines connecting the experimental data points represent simple exponentials. The characteristic relaxation times are plotted as a function of the accumulated film thickness in Fig. 7. It is interesting to notice that these relaxation times τ depend linearly on the total film thickness d as $\tau=\tau_0+\alpha d$.

Simultaneously with the stress the damping of the film–substrate composite was measured. A typical example is shown in Fig. 8. The stress in this figure is already converted to the film stress [13]. The damping of the add-layer is substantially diluted owing to the Si membrane substrate which is 500 times thicker than the add-layers. The conversion of the total damping into the intrinsic damping of the deposited film depends on the mode of stress transfer in the film–substrate composite [16]. Since this is presently unclear, only the original data are shown. The data shown in Fig. 8 suggest that the characteristic times for both stress relaxation and damping are the same as also follows from a detailed data analysis. It is thus concluded that the decrease in damping and the gradual increase in the stress reflect the same relaxation process.

The film growth data may be summarized by stating that the post-deposition relaxation is characterized by a stress increase and simultaneous decrease in the damping. The relaxation originates in the deposited film, it is exponential and characterized by a linear

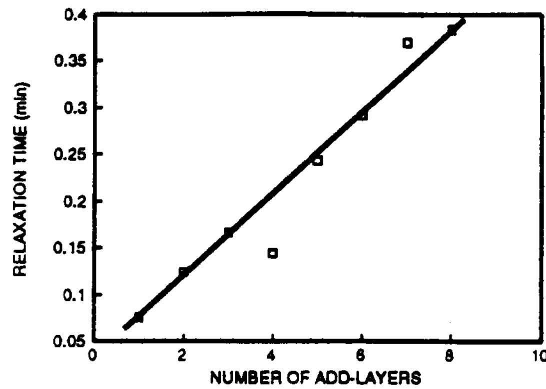


Fig. 7. Stress relaxation time as a function of the number of add-layers deposited.

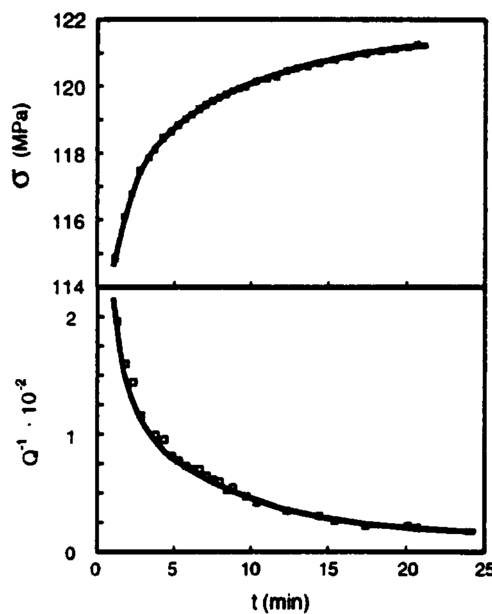


Fig. 8. Simultaneous post-deposition evolution of the damping, Q^{-1} and stress σ.

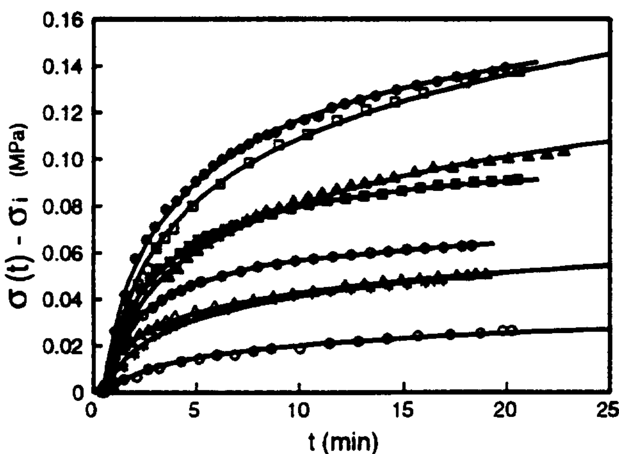


Fig. 6. Analysis of the stress relaxation, $\sigma(t)$-σ_i, occurring at the termination of each add-layer deposition. The lines represent simple exponentials: ○, bottom exponential, first add-layer; △, second add-layer; ○, third add-layer; *, fourth add-layer; ■, fifth add-layer; ▲, sixth add-layer; ●, seventh add-layer; □, eighth add-layer.

thickness dependence of the relaxation time on film thickness. The base value τ_0 in this equation must reflect a surface process whereas the thickness dependent term appears to be a surface initiated bulk process. The underlying microscopic mechanism is probably the reconstruction of a defective add-layer present immediately after deposition, as indicated by the high value of the damping observed at this instance. The increase in the stress in the course of the relaxation can then be driven by the decrease in the free energy on reconstruction.

6. Summary

Adhesion of interface between film–substrate or multilayers can be reflected by dynamic response properties of the composite cantilever or membranes. Understanding of the intrinsic internal friction of thin films requires knowledge of the interface load transfer which can be partially non-elastic if the adhesion is not perfect. The same argument holds in the study of dynamic properties of the multilayer membranes. It is proved that dynamic measurement of the thin layered materials has the potential to assess non-destructively the interface adhesion. Dynamic measurement is also a powerful tool in the study of ultrathin film growth kinetics and dynamics. Work on the dissipation mechanism and stress upward relaxation may provide new information on thin film evolutions.

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