Kinetics of buckling of a compressed film on a viscous substrate

N. Sridhar
Rockwell Science Center, 1049 Camino Dos Rios, Thousand Oaks, California 91360

D. J. Srolovitz[a] and Z. Suo
Princeton Materials Institute and Department of Mechanical and Aerospace Engineering, Princeton University, Princeton, New Jersey 08544

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Compressively-stressed elastic films on finite-thickness viscous substrates can undergo a buckling instability that relieves stresses but destroys the planarity of the film. A linear-stability analysis is performed to determine the onset and maximally unstable mode of this buckling instability as a function of misfit strain, viscous layer thickness, and viscosity. We find that the onset of the buckling instability of the film on a glass layer is the same as that for a compressively stressed free-standing film. However, the maximally unstable wavelength increases as the glass layer thickness increases. Comparisons with experimental data are provided. © *2001 American Institute of Physics.* [DOI: 10.1063/1.1368180]

Compressed films on deformable substrates are ubiquitous in modern technology. For example, heteroepitaxial constraints can lead to compressive stresses in semiconductor films on mismatched substrates, thermal expansion mismatch produces compression in thermal barrier coatings on turbine blades upon cooling from operating temperatures, ion nitriding of the surfaces of metals produces compressive surface layers, etc. One approach to relieving compressive stresses in thin films is to bond them to low viscosity glasses that can flow, leading to an expansion of the film. This is one of the so-called ''compliant substrate'' strategies employed in the growth of low threading dislocation density, relaxed heteroepitaxial semiconductor films.[1] In the recent experiments of Hobart *et al.*,[2] the relaxation of compressively strained heteroepitaxial $Si_{0.7}Ge_{0.3}$ on a low viscosity borophosphorosilicate glass led to the slow buckling of the semiconductor film during annealing, as observed by atomic force microscopy and cross-sectional scanning and transmission electron microscopy. In this letter, we examine conditions under which a compressive film on a viscous substrate will buckle, the wavelength of the buckling, and the time required for buckling to occur. This provides a predictive tool for designing stress relaxation strategies.

Any free-standing film or plate, subjected to compressive stress, will spontaneously buckle at wavelengths that depends on the magnitude of the stress, the elastic constants of the film and the film thickness (see e.g., Ref. 3). Buckling occurs in order to allow for the expansion of the film by bending out of the nominal film plane. The buckling wavelength is a compromise between the relaxation of the in-plane strain and the elastic stresses associated with bending. The constraint provided by an elastic substrate, to which most films are bonded, will prevent buckling. On the other hand, a viscous substrate allows buckling, but on a time scale set by the viscous flow.

Consider a misfitting, elastic film of thickness h_f, Young's modulus E, and Poisson ratio ν bonded to a viscous substrate with viscosity η, and thickness h_g (see Fig. 1). The film substrate system is in turn bonded to a rigid substrate. The film is under a compressive stress, σ_0, which is related to the misfit strain ε_0 by $\sigma_0 = E\varepsilon_0/(1-\nu)$. Upon buckling, the film is under both compression and bending. The classical equilibrium equation describing the vertical displacement of the compressed film, w, is[3]

$$\frac{Eh_f^3}{12(1-\nu^2)}\frac{\partial^4 w}{\partial x^4} + \sigma_0 h_f \frac{\partial^2 w}{\partial x^2} + q = 0, \tag{1}$$

where q is the normal stress exerted on the film by the viscous substrate. This equation is accurate for $w \ll h_f$. If we describe the shape of the film on the substrate as a general, low amplitude ($h_f k \ll 1$) sinusoidal profile $w(x,t) = A(t)\sin(kx)$, we can determine the normal stress that must be provided by the substrate needed to maintain this shape, q. By substituting this form of $w(x,t)$ into Eq. (1), we find that

$$q = \left[\sigma_0 h_f k^2 - \frac{Eh_f^3}{12(1-\nu^2)}k^4\right] A(t)\sin(kx). \tag{2}$$

Next, we turn our attention to the viscous flow of the underlying glass layer. At the interface between the glass layer and the film, the normal stress is prescribed by q [Eq. (2)] and there is no shear stress. At the interface between the glass layer and the rigid substrate, both the tangential and the normal velocities vanish. A similar problem for an infinite

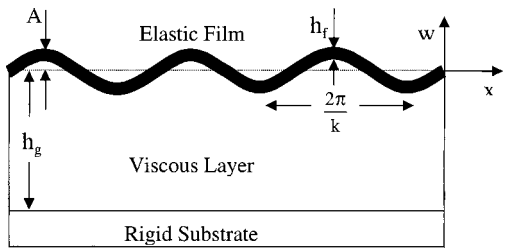

FIG. 1. Schematic illustration of a buckled film on a viscous layer that rests on a rigid substrate.

[a]Electronic mail: srol@princeton.edu

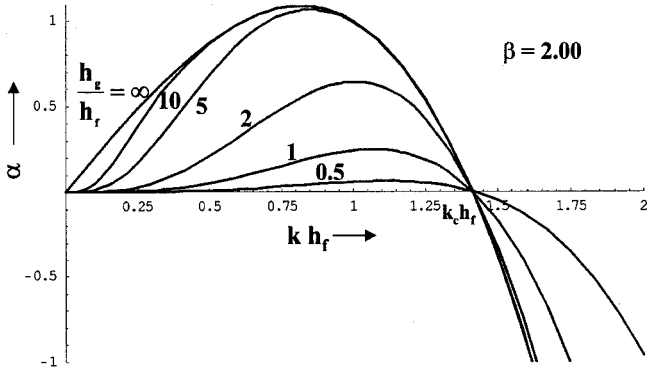

FIG. 2. The normalized growth rate dependence on the dimensionless wave number $h_f k$ for $\beta=2$ and several values of the ratio of the thickness of the glass layer to that of the film, h_g/h_f. The growth rate α initially increases with increasing $h_f k$ and then decays.

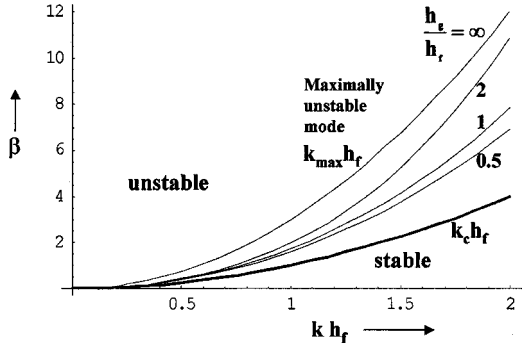

FIG. 3. Stability diagram indicating the conditions (normalized misfit strain β and normalized wave number $h_f k$) under which the film is stable (below the thick line) and unstable against buckling (above the thick line). Also shown are the fastest growing modes as a function of β for several normalized thicknesses, h_g/h_f.

glass layer was solved by Mullins.[4] We have extended his method to solve for the case of a glass layer of finite thickness. For the linear boundary value problem, the velocity of the surface, $\partial w/\partial t$, must be proportional to a representative strain rate, q/η, and to some length scale. There are two length scales in this problem, $1/k$ and h_g. Consequently, the surface velocity can be written as

$$\frac{\partial w}{\partial t}=g\frac{q}{2\eta k}, \tag{3}$$

where g is a dimensionless function of $h_g k$ (if we chose the length scale to be h_g rather than $1/k$, this would simply modify the form of g). The solution of the boundary value problem[5] gives this function

$$g=\frac{\sinh(2h_g k)-2h_g k}{1+\cosh(2h_g k)+2(h_g k)^2}. \tag{4}$$

Note that $g\to 1$ when $h_g k\to\infty$, which reproduces the result of Mullins.[4]

Inserting Eqs. (2) and (4) into Eq. (3) yields a differential equation for the perturbation amplitude: $\partial A(t)/\partial t=\alpha A(t)$, where

$$\alpha=\frac{E}{24\eta(1-\nu^2)}\left[\frac{\sinh(2h_g k)-2h_g k}{1+\cosh(2h_g k)+2(h_g k)^2}\right]\times[\beta(h_f k)-(h_f k)^3], \tag{5}$$

where $\beta=12\varepsilon_0(1+\nu)$. The solution to this differential equation is $A(t)=A(0)e^{\alpha t}$.

The film is unstable to buckling when $\alpha>0$; namely, when $h_f k<\sqrt{\beta}=h_f k_c$. Small wave number perturbations grow since the energy reduction associated with the expansion of the film compensates for the bending energy. For large wave numbers, perturbations decay since, in this case, the energy penalty associated with bending the film dominates the elastic energy release associated with the expansion of the film. This is shown graphically in Fig. 2. Note, the instability condition ($k<k_c$) for buckling (see Fig. 3) is the same as that for a free-standing film subject to a compressive stress. The viscous layer neither prevents the buckling instability nor changes the instability condition for the compressed film. The viscosity of the glass layer only sets the time scale for the growth of the unstable modes.

Figure 2 also shows that the growth rate of the perturbation is sensitive to the values of the normalized glass layer thickness (h_g/h_f), yet the glass layer thickness plays no role in determining the instability condition. The growth rate increases (decreases) with increasing h_g/h_f for wave numbers below (above) the critical value, but saturates in the limit that $h_g/h_f\to\infty$. In this limit ($h_g k\to\infty$), the growth rate α is

$$\alpha=\frac{E}{24\eta(1-\nu^2)}[\beta(kh_f)-(kh_f)^3], \tag{6}$$

while in the small glass layer thickness limit the growth rate reduces to

$$\alpha=\frac{E(kh_g)^3}{36\eta(1-\nu^2)}[\beta(kh_f)-(kh_f)^3]. \tag{7}$$

The perturbation wavelength most likely to be observed in an annealing experiment corresponds to that which grows the fastest. The fastest growing wave number, $k_{\max}$, is found by setting $\partial\alpha/\partial k=0$. This fastest growing mode is shown graphically in Fig. 3. For prescribed misfit strain (β), the fastest growing mode occurs at lower wave numbers as the glass layer thickness is increased. The maximally unstable mode in the limit of small glass layer thickness is $h_f k_{\max}=\sqrt{2\beta/3}$ and the perturbation growth rate is $\alpha=E\beta^3(h_g/h_f)^3[3^5\eta(1-\nu^2)]^{-1}$. In the limit of an infinitely thick glass layer ($h_g\to\infty$) the maximally unstable mode is $h_f k_{\max}=\sqrt{\beta/3}$ and the perturbation growth rate is $\alpha=E\beta^{3/2}[36\sqrt{3}\,\eta(1-\nu^2)]^{-1}$. Finally, we note that in the case of a compressed elastic film on an elastic substrate and relaxation occurring by interfacial diffusion,[6] $h_f k_{\max}=\sqrt{2\beta/3}$, as in the thin glass layer limit. Further calculations (not shown) indicate that the predicted growth rates and maximally unstable wavelength are nearly unchanged on going from a viscous to a viscoelastic layer.[5]

Although the amplitude evolution given by the perturbation analysis $A(t)=A(0)e^{\alpha t}$ is only valid for small times, once the perturbation amplitude becomes large, compared with the film thickness, these results are no longer valid. The real amplitude will not increase without limit since the driving force, the elastic energy stored in the compressed film, will decrease as the buckling amplitude increases. In the long time limit, for a fixed wave number, there will be a thermodynamic, equilibrium amplitude corresponding to a balance

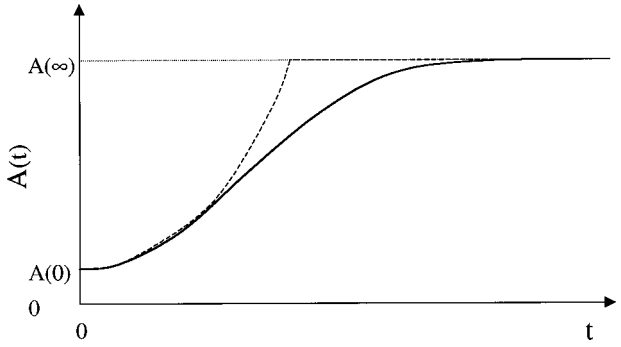

FIG. 4. Schematic illustration of the time (t) dependence of the perturbation amplitude (A). The solid line schematically represents the solution obtained on solving the complete nonlinear elasticity equations for large deflections. The dashed line represents the linearized solution, $A(t) = A(0)\exp[\alpha(k_{\max})t]$. The dotted line shows the equilibrium amplitude of the buckled film [Eq. (8)].

between the release of the compressive stress in the film and the energy of bending the film into the buckled profile.

A generalized expression for the evolution of the thin film profile can be found within the framework of the Foppl–von Karman (FvK) plate analysis.[7] The FvK equations are two coupled partial nonlinear partial differential equations in $w(x,t)$ and the Airy stress function $\chi(x,t)$. For large deflections, the in-plane stress will be reduced upon buckling and will vary spatially along the film (especially when bonded to the viscous substrate). This in-plane stress variation gives rise to a nonlinearity in the governing equations. An additional nonlinearity arises when converting the boundary conditions for the displacements into the corresponding conditions on χ and w. The coupled nonlinear equations can be solved numerically to obtain the full time dependent solution for the amplitude of the perturbation which is valid at all times. For our case, the equilibrium postbuckling elasticity solution can be obtained from the FvK equations in the infinite time limit. The equilibrium amplitude[8,9] is given by

$$A(\infty) = h_f \sqrt{\frac{\beta - \beta_c(k)}{3\beta_c(k)}}, \tag{8}$$

where $\beta_c(k)$ is the critical strain required to buckle the film at a particular wave number, k (see Fig. 3). Assuming that the buckling wave number that survives at late time is the one that grows the fastest ($k = k_{\max}$), Eq. (8) becomes $A(\infty) = h_f\sqrt{2/3}$ for a thick glass layer and $A(\infty) = h_f\sqrt{1/6}$ for a thin glass layer. Figure 4 shows schematically how the amplitude of the film profile will evolve in time for a compressed film on a viscous layer resting on a rigid substrate.

The earlier analysis was based upon the assumption that the buckling wave number that dominates is the one that grows the fastest, i.e., $k_{\max}$. While this is a very good assumption while the buckling is occurring, at late times the amplitude saturates [as per Fig. 4 and Eq. (8)]. Since the absolute minimum elastic energy of the system corresponds to the case where the unstable wave number is as small (i.e., the wavelength is as large) as possible, the wavelength of the buckled film may grow slowly in time. This will commonly correspond to the size of the sample and, hence, very long anneals will be required to achieve the necessary viscous relaxation over this length scale. The final equilibrium state for a finite size sample is a flat, unstressed film lying on a flat glass layer.

The experiments of Hobart *et al.*[2] on $Si_{0.7}Ge_{0.3}$ upon a borophosphorosilicate glass provides data that can be used to check the predictions of the theory. In their experiments, they found a buckling wavelength of approximately 1 μm and a buckling amplitude of 70–80 nm for a film with $\varepsilon_0 = 0.012$ and $h = 34$ nm (30 nm $Si_{0.7}Ge_{0.3}$ film and 4 nm Si underlayer). Assuming that this corresponds to the fastest growing wavelength, the theory presented earlier predicts a wavelength of 0.85 μm and an amplitude $[2A(\infty)]$ of 56 nm. The predictions and experiments are in good agreement. Unfortunately, data showing the kinetics of the evolution of the film were not presented and, hence, it was not possible to directly validate the full range of predictions.

The present analysis provides the theoretical basis for understanding the stability of compressed thin films on viscous substrates (and finite thickness viscous layers resting on rigid substrates) and the basis for the design of processes that are based on viscous relaxation of compressive thin film stresses. Theoretical limits on the absolute stability of these films against buckling and the rates at which instabilities were developed. The analysis, while approximate, is robust and has been validated against the existing experimental data on viscous, compliant substrates.

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