Stability analysis of a viscoelastic model for ion-irradiated silicon

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Abstract

To study the effect of stress within the thin amorphous film generated atop Si irradiated by Ar$^+$, we model the film as a viscoelastic medium into which the ion beam continually injects biaxial compressive stress. We find that at normal incidence, the model predicts a steady compressive stress of a magnitude comparable to experiment. However, linear stability analysis at normal incidence reveals that this mechanism of stress generation is unconditionally stabilizing due to a purely kinematic material flow, depending on none of the material parameters. Thus, despite plausible conjectures in the literature as to its potential role in pattern formation, we conclude that beam stress at normal incidence is unlikely to be a source of instability at any energy, supporting recent theories attributing hexagonal ordered dots to the effects of composition. In addition, we find that the elastic moduli appear in neither the steady film stress nor the leading order smoothening, suggesting that the primary effects of stress can be captured even if elasticity is neglected. This should greatly simplify future analytical studies of highly nonplanar surface evolution, in which the beam-injected stress is considered to be an important effect.

1 Introduction

Pattern formation resulting from uniform ion irradiation of solid surfaces represents a promising potential route to controlled nano-scale surface modification. In particular, the low energy regime (typically $10^2 - 10^4$ eV), where the energy loss is dominated by nuclear collision cascades, has been the topic of continued experimental and theoretical investigations. Due to its simplicity, noble-gas ion irradiation of silicon has been extensively studied as a very promising system for experimental tests of theory: it is a monatomic system amenable to molecular dynamics simulation and its near-surface region is amorphous under ion bombardment, thereby minimizing the potentially confounding effects of disproportionation and crystallographic singularities [1,2].

Despite its attractive attributes, the noble gas / silicon system has proven remarkably finicky, confounding researchers via inter-laboratory irreproducibilities. In particular, for normal-incidence ion-irradiation, researchers in various groups at various times have observed either hexagonal arrays of dots [3], disordered ripple structures [4], combinations of dots and ripples [5], or featureless flat surfaces [6]. The most current physical models of pure materials [7,8,9] – coupling erosion [10], mass redistribution [11], and ion-enhanced viscous flow [12] – have been shown to be maximally stable at normal incidence [9], suggesting that flat surfaces should be generically observed. This has led to speculation that additional physical effects may be generating the observed structures, such as long-range atomic redeposition [13], thin-film stress [4], or the effect of contaminants [14].

There is growing evidence that structures observed on Si under normal-incidence ion irradiation are due to experimental contaminants. On the one hand, it has been shown that after the careful removal of contaminants [4,15] and...
other experimental artifacts [6], formerly patterned surfaces become flat. On the other hand, the controlled addition of contaminants to pure surfaces causes patterns to emerge [16, 17, 14]. Finally, a recent model of concentration effects does admit an instability at normal incidence [18]. These results represent strong evidence for the impurity-driven theory of structure formation.

To isolate impurities as the sole cause of these structures, it is desirable to rule out all other proposed candidates. Very recently, Bradley [19] has shown that redeposition is a nonlinear effect and therefore cannot contribute to linear stability. In this paper, we show that a very general, viscoelastic model of the amorphous surface layer into which a normal-incidence ion beam is continually injecting biaxial stress is morphologically stable against topographical perturbations of all wavelengths. This stands in agreement with the most recent experimental results for this system [6], and provides additional support for the concentration-dependence of observed structures. In addition, we find that the leading-order film dynamics due to beam-injected stress are independent of the elastic constants of the film, suggesting that elasticity may be safely neglected to first approximation.

2 Model

We consider a two-dimensional viscoelastic film of amorphous silicon, irradiated at normal incidence, sitting atop a rigid crystalline substrate. For simplicity, we neglect erosion, so as to focus purely on the effect of stress. We choose a co-ordinate system \((x, z)\) pinned to the film/substrate interface \(z = 0\). Hence, \(x\) is the lateral co-ordinate, and \(z\) is the vertical co-ordinate, with the semi-infinite crystal occupying \(z < 0\). In what follows, \(E\) and \(T\) denote the strain and stress tensors, respectively, while \(E_D\) and \(T_D\) are their deviatoric components:

\[
E_D = E - \frac{1}{3} \text{tr}(E) I
\]

\[
T_D = T - \frac{1}{3} \text{tr}(T) I.
\]

Because we are studying infinitesimal perturbations to a stationary film, we will employ the small-strain approximation:

\[
\frac{DE}{Dt} \approx \frac{1}{2} \left( \nabla v + \nabla v^T \right),
\]

where \(v = (u, w)^T\) is the velocity vector.

The ion irradiation imparts a stress into the film, while simultaneously enhancing the fluidity of the film; hence, a viscoelastic constitutive model is used. In two dimensions, a simple constitutive relation for the film is (see [20, 21, 22, 23]):

\[
\frac{D}{Dt} [E] = \frac{1}{2\eta} T_D + \frac{D}{2G} \frac{D}{Dt} [T_D] + \frac{1}{9B} \frac{D}{Dt} \left[ \text{tr}(T) \right] I + f A \begin{bmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & -2 \end{bmatrix}
\]

1 In principle, there is the possibility that sputtered ions charged with one sign or the other will be sufficiently attracted to the surface (depending on the surrounding field lines) to redeposit at distances large with respect to the instability wavelength. However, due to the concentration of field lines at topographical hilltops, we anticipate that this would be a destabilizing effect proportional to curvature, and so it would not lead to the height-dependent, stabilizing term hypothesized by Facsko et al. [13] and discussed by Davidovitch et al. [7].

2 When erosion is considered, one must adopt a moving frame of reference that follows the eroding interface. In this frame, there is a steady "background" velocity in the vertical (normal) direction. In addition, the stress is zero at the film/substrate interface in the absence of density change, and exponentially approaches the steady state found here. The net effect is that the eroding film has a steady stress of the same form, but of a smaller and position-dependent magnitude, than that found here. The differences are small when the time it takes material to be advected through the amorphous layer from crystalline substrate to free surface is long compared to the Maxwell time, given by the ratio of the shear viscosity to the shear modulus. Under the experimental conditions described here, the advection time is approximately 15 seconds, whereas the Maxwell time is approximately 20 milliseconds.
The first three terms on the right-hand side of Eqn. (1) constitute a standard Maxwell model of viscoelasticity for a two-dimensional material with viscosity $\eta$, shear modulus $G$, and bulk modulus $B$. The fourth term describes the imposition of a stress-free strain by the beam, with $f$ the ion flux, $A$ a measure of strain imparted per ion, and the matrix describing a “pancake strain” – a pure shear consisting of compression in the vertical direction and an equal expansion in the lateral direction.

With stress and strain defined in terms of the velocity field in (1), the bulk governing equations are simply Newton’s second law and the conservation of mass. Assuming that the former simplifies to Stokes flow in a limit of low Reynolds number, we thus have in the bulk

$$\nabla \cdot T = 0$$  \hspace{1cm} (2)
$$\nabla \cdot (\rho \mathbf{v}) = 0,$$  \hspace{1cm} (3)

where $\rho$ is the density. At the boundaries, we have

$$\mathbf{v} = 0 \quad \text{(at } z = 0)$$  \hspace{1cm} (4)
$$\nu_n = \mathbf{v} \cdot \hat{n} \quad \text{(at } z = h(x)),$$  \hspace{1cm} (5)
$$\mathbf{T} \cdot \hat{n} = -\gamma \kappa \hat{n} \quad \text{(at } z = h(x))$$  \hspace{1cm} (6)

Here (4) is the no-slip condition at the film/substrate interface $z = 0$. At the free interface $z = h(x)$, $\hat{n}$ is the surface normal, the kinematic condition (5) relates the velocity $\nu_n$ of the free surface, normal to itself, to the bulk material velocity field $\mathbf{v}$. Finally, condition (6) gives the surface stress in terms of the surface energy $\gamma$ and surface curvature $\kappa$.

3  Analysis

3.1  Steady Solution

We first look for a steady state ($\partial / \partial t \to 0$) consisting of a flat film. Using translational and reflective symmetry in $x$ and $y$, we can limit the steady velocity field $\mathbf{v}_0$ to the form

$$\mathbf{v}_0 (z) = (0, 0, w_0(z))^T.$$  \hspace{1cm} (7)

Then, conservation of mass requires that $w_0(z) = 0$, and so the film is stationary, as we expect. However, the strain and stress associated with this steady state are not determined by the above considerations. These can be obtained as follows. First, from the steady version of the constitutive relation (1), we can write the steady deviatoric stress as

$$T_{D,0} = -2\eta fA \begin{bmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & -2 \end{bmatrix}.$$  \hspace{1cm} (8)

Hence, the steady stress tensor is

$$T_0 = -2\eta fA \begin{bmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & -2 \end{bmatrix} + \frac{1}{3} \text{tr}(T_0) I,$$  \hspace{1cm} (9)
where the trace of the stress tensor (the negative pressure) is unknown. Second, we apply the surface stress condition for a flat surface to obtain a single equation

\[ \text{tr}(T_0) = -12\eta fA, \]  

which solves for the steady stress. Third, the spherical part of the constitutive relation integrates to

\[ \text{tr}(E_0) = \frac{1}{3B} \text{tr}(T_0) = -\frac{4}{B} \eta fA. \]  

Finally, the form for the steady velocity limits the steady strain tensor to the form

\[ E_0 = \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & \frac{\partial w_0}{\partial z} \end{bmatrix}, \]

implying that \( \frac{\partial w_0}{\partial z} = \text{tr}(E_0) = -\frac{4}{B} \eta fA. \) Collecting all of this information, we can express the steady strain and stress as

\[ E_0 = 4 \frac{\eta}{B} fA \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & -1 \end{bmatrix}; \]

\[ T_0 = 6\eta fA \begin{bmatrix} -1 & 0 & 0 \\ 0 & -1 & 0 \\ 0 & 0 & 0 \end{bmatrix}; \]

hence, in the steady state the material is compressively strained vertically by the beam, and compressively stressed laterally. It is notable that the steady stress does not depend on the elastic moduli of the film – only the viscosity.

For one experimental measurement, the steady state exhibits reasonable agreement with experiment. For Si irradiated with Ar\(^+\) at 250 eV with a flux of \( f = 3.5 \times 10^{15} \) ions/(cm\(^2\) sec), a steady stress of 1.4 GPa is observed \[24\]. At this energy and flux, we have previously estimated \( \eta \approx 6.2 \times 10^8 \) Pa sec (see supplement of \[9\]), and although measurements of \( A \) are rare, at 3 keV there has been an estimate of \( A \approx 5 \times 10^{-17} \) cm\(^2\)/ion \[25\]. For these values, we obtain a prediction of \( T_{0,xx} = T_{0,yy} \approx 0.65 \) GPa, which is within about a factor of 2 of the observed value.

### 3.2 Linear Stability

We now study the linear stability of this system under a small perturbation to the film/vapor interface by an infinitesimal normal mode in the \( x \)-direction:

\[ h(x) = h_0 + \varepsilon \exp(ikx + \sigma t) \]  

We consider the plane strain limit of the governing equations, and assume that in the linear regime, the strain and velocity fields will share the same sinusoidal dependence on \( x \) and \( t \); we therefore write

\[ \begin{bmatrix} v \\ E \end{bmatrix} = \begin{bmatrix} v_0 \\ E_0 \end{bmatrix} + \varepsilon \begin{bmatrix} \tilde{v}(z) \\ \tilde{E}(z) \end{bmatrix} e^{ikx + \sigma t}. \]

Upon inserting the ansatz into the governing equations, and keeping only terms to leading order in the infinitesimal parameter \( \varepsilon \), we find that the perturbation \( \tilde{v} = (\tilde{u}, \tilde{w})^T \) to the velocity field is governed by the pair of
ordinary differential equations

\[ \tilde{u}'' - N \tilde{w}' - K \tilde{u} = 0 \]
\[ \tilde{w}'' - M \tilde{u}' - L \tilde{w} = 0 \] (16)

where

\[ K = \frac{4\alpha + 6\beta}{3\alpha} k^2 \]
\[ M = -i \frac{\alpha + 6\beta}{4\alpha + 6\beta} k \]
\[ L = \frac{3\alpha}{4\alpha + 6\beta} k^2 \]
\[ N = -i \frac{\alpha + 6\beta}{3\alpha} k \] (17)

and

\[ \alpha = \frac{2\eta}{1 + \frac{4\sigma}{c}} \quad \beta = \frac{B}{\sigma} \] (18)

These equations can be re-written as a linear system and solved using eigenvalue analysis; the general solution can be expressed as

\[ \begin{bmatrix} \tilde{u} \\ \tilde{w} \end{bmatrix} = \begin{bmatrix} a & c \\ b & d \end{bmatrix} \cosh(kz) + \begin{bmatrix} b \end{bmatrix} \sinh(kz) + \frac{\alpha + 6\beta}{7\alpha + 6\beta} k^2 \begin{bmatrix} b - ic \\ -ia - d \end{bmatrix} \cosh(kz) + \begin{bmatrix} -ib - c \\ (a - id) \end{bmatrix} \sinh(kz) \] (19)

To obtain the four unknowns \{a, b, c, d\}, we must apply the linearized boundary conditions. From Eqn. (4) at \( z = 0 \), we immediately find that \( a = c = 0 \). Turning next to Eqn. (6) at \( z = h \), we find that its linearization is

\[ \tilde{T}_{xx} = \frac{\alpha}{2} (ik\tilde{w} + \tilde{u}') = -6fA\eta ik \]
\[ \tilde{T}_{zz} = \frac{\alpha}{3} (-ik\tilde{u} + 2\tilde{w}') + \beta (ik\tilde{u} + \tilde{w}') = -\gamma k^2 \] (20)

Because \( a = c = 0 \), Eqn. (20) represents a matrix equation for \( b \) and \( d \); solution of this equation yields

\[ b = -\frac{ia k}{\Delta} \left\{ 6fA\eta k [V \cosh(Q) - UQ \sinh(Q)] + \gamma k^2 [-UQ \sinh(Q) + UQ \cosh(Q)] \right\} \]
\[ d = -\frac{ak}{\Delta} \left\{ 6fA\eta k [-UQ \sinh(Q) - UQ \cosh(Q)] + \gamma k^2 [V \cosh(Q) + UQ \sinh(Q)] \right\} \] (21)

where

\[ Q = kh \]
\[ U = \frac{\alpha + 6\beta}{7\alpha + 6\beta} \]
\[ V = \frac{4\alpha + 6\beta}{7\alpha + 6\beta} \]

are common dimensionless groups, and

\[ \Delta = (\alpha k)^2 \left[ V^2 + U \sinh^2(Q) + U^2 Q^2 \right] \] (22)

is the determinant of the matrix associated with equation (20). Finally, inserting the coefficients (21) into (19), we apply the linearized version of the kinematic condition (5).

\[ \sigma = \tilde{w}(h) \] (23)
which provides the implicit dispersion relation between the growth rate $\sigma$ and wavenumber $k$:

\[
\frac{2R}{1+R} \left[ V^2 + U^2 Q^2 + U \sinh^2 (Q) \right] + D \left[ U^2 Q^2 - (V - U) \sinh^2 (Q) \right] + CV Q \left[ \sinh (2Q) - 2UQ \right] = 0
\]  

(24)

Here we have converted to the dimensionless parameters $\{ R, Q, D, C \}$ are given by

\[ R = \frac{\eta G}{\sigma} \] (growth rate)

\[ Q = hk \] (wavenumber)

\[ D = \frac{6fA\eta}{G} \] (Deborah number)

\[ C = \frac{\gamma}{2Gh} \] (Capillary number)

### 3.3 Interpretation

Equation (24) is our central theoretical result, but requires some further examination. Although an explicit dispersion relation is not available, we can perform neutral stability analysis on (24) by setting $\sigma \to 0$ and solving the resulting expression for $D$, which value of $D$ we name $D^*$. In this limit one can show that $U \to 1$, $V \to 1$, $R \to 1$, and the resulting expression for $D^*$ is

\[ D^*(Q) = 2C \left[ 1 - \frac{\sinh (2Q)}{2Q} \right] \]  

(26)

this value of $D$ establishes the neutral stability boundary. For values of $D^*$ in the domain of this function, both stable and unstable wavenumbers $Q$ exist; hence, the extremal values of $D$ serve as boundaries between stable and unstable regions of parameter space. As observed in Figure 4, $D^*(Q)$ is a strictly negative function of $Q$, with a global maximum of $D = 0$ at $Q = 0$, and so the stability of the film depends upon the sign of $D$. By implicitly differentiating (24) in $R$ and $D$, we find that $\frac{\partial R}{\partial D}$ is negative at $R = D = Q = 0$, so that positive $D$ implies negative $R$. Because $D$ is a physical constant and positive by definition, we conclude that the film is stable at all wavelengths. Hence, even though the pancake strain places the film in a state of compressive stress, the film is unconditionally stable to perturbations. Our stability result may be understood intuitively by noting that even though the beam stresses the bulk material below the valleys of a perturbation, the effect on the hilltops of a small perturbation is for them to shorten and widen under the stress-free pancake strain.

Further quantitative understanding is available for the commonly-observed situation in which the film thickness is much smaller than the perturbation wavelength – i.e., that $Q = hk \ll 1$. In the limit of long wavelengths and slow evolution ($Q \ll 1$ and $R \ll 1$), the dispersion relation (24) reduces, keeping the lowest order of $Q$ in each of the coefficients, to

\[ R \approx -\frac{1}{2} D Q^2 - \frac{2}{3} C Q^4 \]  

(27)

or, reverting to dimensional form,

\[ \sigma = -3fA (hk)^2 - \frac{\gamma}{3\eta h} (hk)^4. \]  

(28)

Hence, for common case of long-wavelength perturbations, the leading-order contribution of the pancake strain at normal incidence is a second-order smoothing of perturbations. A very important property of this smoothing is that it depends on none of the bulk material properties of the film – it is a purely kinematic response to the biaxial stress
injected by the beam.

Because neither the steady stress, nor the leading order stability properties of the film depend on the elastic properties of the medium, it is reasonable to consider neglecting elasticity altogether. Hence, we conclude by considering the limit of a film that is purely viscous and incompressible \((G \to \infty \text{ and } B \to \infty)\). In that limit, we recover the (dimensional) result

\[
\sigma(k) = -\frac{6fA(hk)^2 + \frac{k\gamma}{\eta}(2hk - 2hk)}{1 + 2(hk)^2 + \cosh(2hk)}.
\]  

(29)

This is again unconditionally stable, and in the absence of the beam \((f \to 0)\), it reduces to the classic result of Orchard for viscous surface leveling on films of arbitrary thickness \([26]\).

4 Summary

As a model for amorphous ion-irradiated solids, we have studied the dynamics of a thin viscoelastic film subject to continual injection of biaxial stress, and obtained two primary results.

- First, we have shown that biaxial compressive stress injected into an amorphous film by the ion beam is unconditionally stabilizing at normal incidence, and hence that smooth surfaces at low angles should be generic for pure amorphous materials under energetic particle irradiation. Together with growing experimental consensus that normal-incidence patterns only appear when contaminants are present, and Bradley’s recent demonstration that a simple model of concentration effects does admit an instability at normal incidence \([18]\), this strengthens the case that these structures are due entirely to concentration effects.

- Second, we have shown that the leading-order contributions to film dynamics in the small-curvature limit of this

\[3\]Two comments are in order here. First, although these limits appear problematic in the dimensionless parameters \([25]\), they are in fact well-defined if the dispersion relation \([25]\) is first converted to dimensional form. Second, while the incompressible limit is a singular limit at the level of the governing equations, requiring the introduction of a hydrodynamic pressure, it is an ordinary limit at the level of the solution to those equations. Hence, a re-analysis using the incompressible equations is unnecessary.
model are independent of the elastic constants. Rather than being due to elasticity, the steady stress observed in this model is due to viscous resistance to the stress-free strain induced by the beam. This result suggests that elasticity may be safely neglected in future analytical efforts. In particular, as demonstrated by George et al. [25], a purely viscous version of Equation (1) will greatly simplify the analysis of highly-nonplanar surface evolution.

Although our analysis is restricted to one independent spatial dimension and does not account for the advection in a moving reference frame due to sputter erosion, we anticipate that the conclusions drawn here will be no different from a deeper analysis that accounts for these effects.

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