Stability analysis of a viscoelastic model for ion-irradiated silicon

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Recently, elastic stress has been among several mechanisms hypothesized to induce the formation of ordered structures in Si irradiated at normal incidence by energetic ions. To test this hypothesis, we model the thin amorphous film atop ion-irradiated Si as a viscoelastic continuum 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 and molecular dynamics simulation. 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 compressive stress induced by normal-incidence ion bombardment is unlikely to be a source of instability at any energy. In fact, with this result, all hypothesized mechanisms suggested to explain structures on pure materials under normal incidence irradiation have now been overturned, supporting recent theories attributing hexagonal ordered dots to the effects of composition. In addition to this result, 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 supports the basic framework recently adopted by other authors and should allow future analytical studies of highly nonplanar surface evolution, in which the beam-injected stress is considered to be an important effect.

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I. INTRODUCTION

Pattern formation resulting from uniform ion irradiation of solid surfaces represents a promising potential route to controlled nanoscale 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 interlaboratory irreproducibilities. In particular, for normalincidence ion irradiation, researchers in various groups at various times have observed either hexagonal arrays of dots,³ disordered ripple structures,^{[4](#page-5-0)} combinations of dots and ripples, 5 or featureless flat surfaces. 6 However, the most current physical models of pure materials, $7-9$ coupling erosion,^{[10](#page-5-0)} mass redistribution, $11,12$ and ion-enhanced viscous flow, $13,14$ have been shown to be maximally stable at normal incidence, 9 suggesting that flat surfaces should be generically observed. This disconnect between theory and experiment has led to speculation that additional physical effects may be generating the observed structures, such as long-range atomic redeposition, 15 or elastic stress, 4 under a hypothesized mechanism analogous to the well-known Asaro-Tiller-Grinfeld (ATG) instability¹⁶⁻¹⁸ in solid films subjected to a mismatch strain with the underlying substrate.

More recently, however, there has been growing evidence that structures observed on Si under normal-incidence ion irradiation are due to neither of these effects, but rather to experimental contaminants.^{[19](#page-5-0)} On the one hand, it has been shown that after the careful removal of contaminants^{[4,20](#page-5-0)} 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.^{19,21,22} Finally, a recent model of concentration effects does admit an instability at normal incidence. 23 23 23

Although the experimental evidence for contaminants is compelling, it is desirable to achieve closure by ruling out previously proposed candidates, which are still discussed frequently within the community. Very recently, Bradley has shown that redeposition is a nonlinear effect and therefore cannot contribute to linear stability, 24 thus rejecting the redeposition hypothesis. In this paper, we investigate whether or not an instability similar to ATG could be present in irradiated solids. Modeling the amorphous layer as a general, viscoelastic film into which a normal-incidence ion beam is continually injecting biaxial stress, we find that the film is unconditionally stable against topographical perturbations of all wavelengths. It therefore provides an analytical foundation for rejecting the elastic stress hypothesis. Together, the rejection of these alternate hypotheses provides additional support for the concentration dependence of observed structures. In addition, we also find that the leading-order film dynamics due to beam-injected stress are independent of the elastic constants of the film, suggesting that elastic effects may be safely neglected to first approximation.

II. MODEL

We consider a crystalline silicon target under normalincidence irradiation by argon ions in the eV to low-keV range of energies. Under this irradiation, a ∼3–10-nm film of silicon becomes amorphous; this film is the object of our study, and we model it as a homogeneous viscoelastic medium. For simplicity, we neglect erosion, so as to focus purely on the effect of stress. We choose a coordinate system (x, y, z) pinned to the the film/substrate interface $z = 0$, with the film occupying the region $0 \le z \le h$ and the semi-infinite crystal occupying $z < 0$. In what follows, $\mathbf{u} = (u_1, u_2, u_3)^T$ and $\mathbf{v} = (v_1, v_2, v_3)^T$ denote the displacements and velocities, respectively, **E** and **T** denote the strain and stress tensors, respectively, and **E***^D* and **T***^D* are their deviatoric components:

$$
\mathbf{E}_D = \mathbf{E} - \frac{1}{3} \text{tr}(\mathbf{E}) \mathbf{I}, \quad \mathbf{T}_D = \mathbf{T} - \frac{1}{3} \text{tr}(\mathbf{T}) \mathbf{I}.
$$

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

$$
\mathbf{E} \approx \frac{1}{2} (\nabla \mathbf{u} + \nabla \mathbf{u}^{\mathrm{T}}), \quad \frac{D \mathbf{E}}{Dt} \approx \frac{1}{2} (\nabla \mathbf{v} + \nabla \mathbf{v}^{\mathrm{T}}). \tag{1}
$$

As with any continuous mechanical medium, the governing equations within the interior of the film 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

$$
\nabla \cdot \mathbf{T} = 0,\tag{2}
$$

$$
\nabla \cdot (\rho \mathbf{v}) = 0,\tag{3}
$$

where ρ is the density. At the boundaries of the film, we have

$$
\mathbf{v} = 0 \quad (\text{at } z = 0), \tag{4}
$$

$$
v_{n} = \mathbf{v} \cdot \hat{\mathbf{n}} \quad [\text{at } z = h(x, y)], \tag{5}
$$

$$
\mathbf{T} \cdot \hat{\mathbf{n}} = -\gamma \kappa \hat{\mathbf{n}} \quad [\text{at } z = h(x, y)]. \tag{6}
$$

Here (4) is the no-slip condition at the film/crystal interface $z = 0$ (the crystal is treated as a rigid body). At the free interface $z = h(x, y)$, $\hat{\mathbf{n}}$ is the surface normal, and the kinematic condition (5) relates the velocity v_n of the free surface, normal to itself, to the bulk material velocity field **v**. Finally, condition (6) gives the surface stress in terms of the surface energy *γ* and surface curvature *κ*.

It remains to relate the stress and the strain via a constitutive relationship and to describe the effect of the beam within the film. Recently, a model has been advanced in which the film obeys the Navier-Stokes equations, and the effect of the beam is modeled as an "effective body force":^{14,25}

$$
\mathbf{b} = \nabla \cdot \mathbf{T}^{S} = \mathbf{f}_{E} \Psi (\theta - \gamma), \qquad (7)
$$

which acts on the entirety of the amorphous film. Although the incoming ions undoubtedly exert a force on the film as they are slowed down by the film, that force is vanishingly small, and Eq. (7) is not interpreted as representing any physical force. Instead, **f***^E* is proposed to represent "the coarse-grained information about the effect of the residual stress created in the target^{["14](#page-5-0)} and $\Psi(\theta - \gamma)$ is proposed to encode dependence upon the local angle of incidence $\theta - \gamma$. Though unusual in the sense that a "body force" acting throughout the film would traditionally be completely independent of the configuration of the nearest patch of surface (surface dependencies normally appearing as boundary conditions), this is an interesting effort to encompass the many complexities of the ion irradiation process within a simple, intuitive form.

We shall here adopt different models for both the film and the action of beam therein. Because we wish to investigate the relative importance of elastic and viscous effects and specifically to test whether elastic effects could cause an instability, we require a more general constitutive law, and we use as a starting point a linear Maxwell viscoelastic relation to describe the film itself. As for the effect of the beam, we prefer to use a model more closely, if still loosely, tied to the microscopic picture associated with single-ion impacts. At this scale, molecular dynamics simulation has shown that each impact significantly redistributes the target silicon atoms to new locations, $9,12,26$ $9,12,26$ gradually increasing the magnitude of a compressive stress to a saturated state, 27 which is also observed experimentally[.28](#page-6-0) Each impact thus induces a direct deformation of the material, suggesting that the effect of the beam be incorporated directly into the constitutive relationship between stress and strain in a way that depends linearly on the total fluence.

A model with exactly these properties has already been developed to describe high-energy ion irradiation in the regime of electronic stopping, where the stress is generated by a rapid thermal cycling[.29–32](#page-6-0) This *physical mechanism* is, of course, not directly applicable in the nuclear stopping regime. However, anisotropic plastic flow has been observed experimentally even in the nuclear-stopping regime, 33 and the *mathematical form* of the governing equations has been successfully applied to describe various phenomena at low energies.^{[34,35](#page-6-0)} In addition, we will here demonstrate that, without specifying a mechanism at all, simple symmetry arguments allow the derivation of this form, suggesting it is, in fact, generic. Thus, we adapt from Ref. [32](#page-6-0) the constitutive relation

$$
\frac{D}{Dt}[\mathbf{E}] = \frac{1}{2\eta}\mathbf{T}_D + \frac{1}{2G}\frac{D}{Dt}[\mathbf{T}_D] + \frac{1}{9B}\frac{D}{Dt}[\text{tr}(\mathbf{T})]\mathbf{I} + fA\mathbf{D}.
$$
\n(8)

The first three terms on the right-hand side of Eq. (8) constitute a standard Maxwell model of viscoelasticity for a two-dimensional material with viscosity *η*, shear modulus *G*, and bulk modulus *B*; these provide the minimal framework required to investigate the relative importance of viscous to elastic effects. The fourth term describes the imposition of a stress-free rate of strain by the beam depending linearly on the ion flux *f* , with *A* being a measure of strain imparted per ion and the tensor **D** describing the nature of that strain. Hence, this way of describing stress is compatible both with the direct imposition of strain by ion impacts and with the linear growth of that strain in the fluence.

Starting merely from the *observation* of a steady stress during low-energy irradiation^{28,34–37} and without directly appealing to any particular physical mechanism, we now proceed to make three reasonable restrictions on the form of **D**. First, for normal-incidence irradiation, we expect *translational, reflective, rotational, and chiral* symmetry in *x* and *y*. The translational symmetry rules out any dependence on these variables, the reflective and chiral symmetries require that the off-diagonal members of **D** vanish, and the rotational symmetry requires that $D_{xx} = D_{yy}$. Second, because the number of implanted argon atoms saturates at very early times, we expect **D** to be traceless in any steady state. Third, merely for convenience, we take **D** to be uniform throughout the film (i.e., without a dependence on *z*). These considerations alone are sufficient to produce a strain tensor of the form

$$
\mathbf{D} = \begin{bmatrix} v & 0 & 0 \\ 0 & v & 0 \\ 0 & 0 & -2v \end{bmatrix}; \tag{9}
$$

then, because **D** is already multiplied by the parameter *A* measuring induced strain per ion, we take $\nu = 1$ without loss of generality. We thus arrive at a constitutive law identical to that employed for high-energy irradiation, without invoking any of the physics of that regime (which do not apply here). Indeed, the nature of the arguments used to obtain Eq. (9) suggest that it is, in fact, generic.

III. ANALYSIS

A. Steady solution

We first look for a steady state (*∂/∂t* → 0) consisting of a flat film. Using translational and reflective symmetry in *x* and *y*, we can limit the steady velocity field v_0 to the form

$$
\mathbf{v}_0(z) = (0, 0, w_0(z))^T.
$$
 (10)

Then, conservation of mass in the steady state 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 [\(8\),](#page-1-0) we can write the steady deviatoric stress as

$$
\mathbf{T}_{D,0} = -2\eta f A \begin{bmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & -2 \end{bmatrix} . \tag{11}
$$

Hence, the steady stress tensor is

$$
\mathbf{T}_0 = -2\eta f A \begin{bmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & -2 \end{bmatrix} + \frac{1}{3} \text{tr}(\mathbf{T}_0) \mathbf{I}, \quad (12)
$$

where the trace of the stress tensor (the *negative pressure*) is unknown. Second, we apply the surface stress condition [\(6\)](#page-1-0) for a flat surface to obtain a single equation,

$$
tr(T_0) = -12\eta f A, \qquad (13)
$$

which solves for the steady stress. Third, the by taking the trace of Eq. (8) and integrating in time, we obtain from the isotropic part of the constitutive relation

$$
tr(E_0) = \frac{1}{3B} tr(T_0) = -4\frac{\eta}{B} f A.
$$
 (14)

Finally, the form (10) for the steady velocity limits the steady strain tensor to the form

$$
\mathbf{E}_0 = \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & \frac{\partial w_0}{\partial z} \end{bmatrix},
$$
(15)

implying that $\frac{\partial w_0}{\partial z}$ = tr (**E**₀) = −4 $\frac{\eta}{B} f A$. Collecting all of this information, we can express the steady strain and stress as

$$
\mathbf{E}_0 = 4\frac{\eta}{B} f A \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & -1 \end{bmatrix},
$$

\n
$$
\mathbf{T}_0 = 6\eta f A \begin{bmatrix} -1 & 0 & 0 \\ 0 & -1 & 0 \\ 0 & 0 & 0 \end{bmatrix};
$$
 (16)

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.

We here pause briefly to compare the steady-state predictions (16) to experiment. For Si irradiated with Ar^+ at 250 eV with a flux of $f = 3.5 \times 10^{15}$ ions/(cm² s), a steady stress of 1.4 GPa is observed. 28 At this energy and flux, we have previously estimated $\eta \approx 6.2 \times 10^8$ Pa s (Ref. [9\)](#page-5-0), and although measurements of *A* are rare, at 3 keV there has been an estimate of $A \approx 5 \times 10^{-17}$ cm²/ion.³⁵ Using this value of *A* directly, we obtain a prediction of $T_{0,xx} = T_{0,yy} \approx 0.65 \text{ GPa}$, which is within about a factor of 2 of the observed value. Without an estimate of *A* at 250 eV, it is impossible to make a better prediction, but the mechanism gives a reasonable estimate of the observed stress.

B. 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*; because of the symmetries in the system we may, without loss of generality, orient this mode in the *x* direction:

$$
h(x) = h_0 + \varepsilon \exp(ikx + \sigma t). \tag{17}
$$

Because of the infinite extent of the film in the *y* direction, it is appropriate to consider the *plane-strain* limit of the governing equations and to assume that in the linear regime, the velocity and stress fields will share the same sinusoidal dependence on *x* and *t*; we therefore write for the displacements and velocities

$$
\begin{bmatrix} \mathbf{u} \\ \mathbf{v} \end{bmatrix} = \begin{bmatrix} \mathbf{u}_0 \\ \mathbf{v}_0 \end{bmatrix} + \varepsilon \begin{bmatrix} \tilde{\mathbf{u}}(z) \\ \tilde{\mathbf{v}}(z) \end{bmatrix} \exp(ikx + \sigma t). \tag{18}
$$

From the ansatz (18) , the the perturbed strain field $\mathbf{E} =$ $\mathbf{E}_0 + \mathbf{\tilde{E}}(z)$ is obtained from the definitions [\(1\),](#page-1-0) whereupon the perturbed stress field $T = T_0 + \tilde{T}(z)$ is obtained from the constitutive relation [\(8\).](#page-1-0) Finally, upon inserting this expression for the stress into the governing equations (2) and (3) and keeping only terms to leading order in the infinitesimal parameter *ε*, we find that the perturbation $\tilde{\mathbf{v}} = (\tilde{v}_1, 0, \tilde{v}_3)^T$ to the velocity field is governed by the pair of ordinary differential equations

$$
\tilde{v}_1'' - N\tilde{v}_3' - K\tilde{v}_1 = 0, \quad \tilde{v}_3'' - M\tilde{v}_1' - L\tilde{v}_3 = 0,\tag{19}
$$

where

$$
K = \frac{4\alpha + 6\beta}{3\alpha}k^2, \quad M = -i\frac{\alpha + 6\beta}{4\alpha + 6\beta}k,
$$

$$
L = \frac{3\alpha}{4\alpha + 6\beta}k^2, \quad N = -i\frac{\alpha + 6\beta}{3\alpha}k,
$$
 (20)

and

$$
\alpha = \frac{2\eta}{1 + \frac{\eta \sigma}{G}}, \quad \beta = \frac{B}{\sigma}.
$$
 (21)

In the Appendix we show that Eqs. (19) can be rewritten as a linear system and solved using eigenvalue analysis; a convenient form for the general solution is

$$
\begin{bmatrix} \tilde{u} \\ \tilde{w} \end{bmatrix} = \begin{bmatrix} a \\ c \end{bmatrix} \cosh(kz) + \begin{bmatrix} b \\ d \end{bmatrix} \sinh(kz) + \frac{\alpha + 6\beta}{7\alpha + 6\beta} kz
$$

$$
\times \begin{bmatrix} (b - ic)\cosh(kz) + (a - id)\sinh(kz) \\ (-ia - d)\cosh(kz) + (-ib - c)\sinh(kz) \end{bmatrix}.
$$
(22)

To obtain the four unknowns {*a, b, c, d*}, we must apply the linearized boundary conditions. From Eq. [\(4\)](#page-1-0) at $z = 0$, we immediately find that $a = c = 0$. Turning next to Eq. [\(6\)](#page-1-0) at $z = h$, we find that its linearization is

$$
\tilde{T}_{xz} = \frac{\alpha}{2} (ik\tilde{w} + \tilde{u}') = -6f A \eta ik,
$$
\n
$$
\tilde{T}_{zz} = \frac{\alpha}{3} (-ik\tilde{u} + 2\tilde{w}') + \beta (ik\tilde{u} + \tilde{w}') = -\gamma k^2.
$$
\n(23)

Because $a = c = 0$, Eq. (23) represents a matrix equation for *b* and *d*; solution of this equation yields

$$
b = -\frac{iak}{\Delta} \{6f A\eta k[V \cosh(Q) - UQ \sinh(Q)]
$$

+ $\gamma k^2 [-(V - U) \sinh(Q) + UQ \cosh(Q)]\},$

$$
d = -\frac{ak}{\Delta} \{6f A\eta k[-(V - U) \sinh(Q) - UQ \cosh(Q)]
$$

+ $\gamma k^2 [V \cosh(Q) + UQ \sinh(Q)]\},$ (24)

where

$$
Q = kh, \quad U = \frac{\alpha + 6\beta}{7\alpha + 6\beta}, \quad V = \frac{4\alpha + 6\beta}{7\alpha + 6\beta}
$$

are common dimensionless groups, and

$$
\Delta = (\alpha k)^{2} [V^{2} + U \sinh^{2}(Q) + U^{2} Q^{2}]
$$
 (25)

is the determinant of the matrix associated with Eq. (23). Finally, inserting the coefficients (24) into (22), we apply the linearized version of the kinematic condition [\(5\),](#page-1-0)

$$
\sigma = \tilde{w}(h),\tag{26}
$$

which provides the implicit dispersion relation between the growth rate σ and wave number k :

$$
\frac{2R}{1+R}[V^2 + U^2 Q^2 + U \sinh^2(Q)]
$$

+ $D[U^2 Q^2 - (V - U) \sinh^2(Q)]$
+ $CV Q[\sinh(2Q) - 2U Q] = 0.$ (27)

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

$$
R = \frac{\eta}{G}\sigma \quad \text{(growth rate)},
$$

\n
$$
Q = hk \quad \text{(wave number)},
$$

\n
$$
D = \frac{6fA\eta}{G} \quad \text{(Deborah number)},
$$

\n
$$
C = \frac{\gamma}{2Gh} \quad \text{(capillary number)}.
$$
 (28)

C. Interpretation

Equation (27) is our central theoretical result, but being highly implicit, it requires some further examination. Although an explicit dispersion relation is not available, we can perform neutral stability analysis on Eq. (27) 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 $U \to 1$, $V \to 1$, $R \rightarrow 1$, and the resulting expression for *D*^{*} is

$$
D^*(Q) = 2C \left[1 - \frac{\sinh(2Q)}{2Q} \right];\tag{29}
$$

this value of *D* establishes the *neutral stability boundary.* For values of *D*[∗] in the *domain* of this function, both stable and unstable wave numbers *Q* exist; hence, the extremal values of *D* serve as boundaries between stable and unstable regions of parameter space. As observed in Fig. 1, $D^*(Q)$ 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 (27) 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 strain [\(9\)](#page-2-0) 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

FIG. 1. (Color online) Neutral stability curve *D*[∗](*Q*), from Eq. (29).

the hilltops of a small perturbation is for them to shorten and widen under the stress-free 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 [\(27\)](#page-3-0) reduces, keeping the lowest order of *Q* in each of the coefficients, to

$$
R \approx -\frac{1}{2}D Q^2 - \frac{2}{3}C Q^4 \tag{30}
$$

or, reverting to dimensional form,

$$
\sigma = -3fA(hk)^{2} - \frac{\gamma}{3\eta h}(hk)^{4}.
$$
 (31)

Hence, for the common case of long-wavelength perturbations, the leading-order contribution of the induced strain [\(9\)](#page-2-0) 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. Hence, especially for problems exhibiting the limiting behavior $hk \ll 1$, it is reasonable to consider neglecting elasticity entirely and treating the film as a purely viscous material.

IV. 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.

(i) First, we have shown that biaxial compressive stress injected into an amorphous film by the ion beam is unconditionally stabilizing at normal incidence; hence, no analog of the Asaro-Tiller-Grinfeld mechanism is present in stressed viscoelastic films. Together with recent results showing that long-range redeposition has been shown to be a purely nonlinear effect 24 and that the net effect of erosion and redistribution in the "prompt regime" is stable at normal incidence,^{[9](#page-5-0)} there now remain *no* hypothesized mechanisms predicting instability at normal incidence, suggesting that smooth surfaces at low angles should be generic for pure amorphous materials under energetic particle irradiation. Together with growing experimental consensus that normalincidence 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, 23 23 23 this strengthens the case that these structures are due entirely to concentration effects.

(ii) Second, we have shown that the leading-order contributions to film dynamics in the small-curvature limit of this 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, supporting the basic framework behind recent contributions from other authors.¹⁴ In fact, a version of our constitutive law (8) , with elastic terms

neglected entirely, could in fact provide a physical origin for the "effective body force" proposed therein.

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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APPENDIX: ADDITIONAL SOLUTION DETAILS

We here present a more detailed solution of the linear second-order system (19) , which we rewrite for convenience:

$$
\tilde{v}_1'' - N\tilde{v}_3' - K\tilde{v}_1 = 0, \quad \tilde{v}_3'' - M\tilde{v}_1' - L\tilde{v}_3 = 0.
$$
 (A1)

We first recast Eq. $(A1)$ as a first-order dynamical system,

$$
\frac{d\zeta}{dz} = \mathbf{A}\zeta,\tag{A2}
$$

where $\zeta = [\tilde{v}_1, \tilde{v}_3, \tilde{v}'_1, \tilde{v}'_3]$, and

$$
\mathbf{A} = \begin{bmatrix} 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 1 \\ K & 0 & 0 & N \\ 0 & L & M & 0 \end{bmatrix} .
$$
 (A3)

a. Solution. A short calculation using the definitions [\(20\)](#page-3-0) reveals that the matrix **A** in Eq. (A3) has two eigenvalues, $\lambda_1 = -k$ and $\lambda_2 = k$, each having multiplicity 2. We will consider these simultaneously. Solving $(A \pm kI)$ $v_{1,2} = 0$ gives one eigenvector of Eq. $(A3)$ for each eigenvalue and a corresponding solution to Eq. (A2):

$$
\mathbf{v}_{1,2} = \begin{bmatrix} \pm kN \\ (K - k^2) \\ k^2N \\ \pm k(K - k^2) \end{bmatrix}, \quad \zeta_{1,2}(z) = \mathbf{v}_{1,2} e^{\mp kz}. \quad (A4)
$$

Now, the system $(A \pm kI) v = 0$ has only one nontrivial solution, and hence each eigenvalue has only one eigenvector. Nevertheless, following Ref. [38,](#page-6-0) two additional solutions $\zeta_{3,4}(z)$ associated with the eigenvalues $\lambda = \pm k$ may be found, in the form

$$
\zeta_{3,4}(z) = (\mathbf{v}_{3,4} + z\mathbf{v}_{1,2})e^{\mp kz},\tag{A5}
$$

where $v_{3,4}$ are a pair of a generalized eigenvectors of **A** satisfying

$$
(A \pm kI) v_{3,4} = v_{1,2}.
$$
 (A6)

However, instead of solving the 4 \times 4 system [\(A6\)](#page-4-0) for the eigenvectors $\mathbf{v}_{3,4}$ of A, we will now take a shortcut. Recalling that the four-component system $(A2)$ is just a means to the solution of the two-component system $(A1)$, we first extract from Eq. $(A4)$ the associated pair of solutions for $(\tilde{v}_1, \tilde{v}_3)$:

$$
\begin{bmatrix} \tilde{v}_1 \\ \tilde{v}_3 \end{bmatrix}_{1,2} = \begin{bmatrix} \pm Nk \\ (K - k^2) \end{bmatrix} e^{\mp kz}.
$$

From this first solution, appealing to Eq. [\(A5\),](#page-4-0) we immediately look for a second solution pair of the form

$$
\begin{bmatrix} \tilde{v}_1 \\ \tilde{v}_3 \end{bmatrix}_{3,4} = \left\{ \begin{bmatrix} c_1 \\ c_2 \end{bmatrix} + z \begin{bmatrix} \pm Nk \\ (K - k^2) \end{bmatrix} \right\} e^{\mp kz}.
$$

Inserting this into [\(A2\),](#page-4-0) we solve the resulting 2×2 system for c_1 and c_2 , obtaining the second solution pair

$$
\begin{bmatrix} \tilde{v}_1 \\ \tilde{v}_3 \end{bmatrix}_{3,4} = \left\{ \begin{bmatrix} -N \\ \pm 2k \end{bmatrix} + z \begin{bmatrix} \pm Nk \\ (K - k^2) \end{bmatrix} \right\} e^{\mp kz}.
$$

b. Change of basis. Dropping the \pm notation, the four solutions just obtained represent a general solution of the form

$$
\begin{bmatrix} \tilde{v}_1 \\ \tilde{v}_3 \end{bmatrix} = \tilde{a} \begin{bmatrix} Nk \\ K - k^2 \end{bmatrix} e^{-kz} + \tilde{b} \begin{bmatrix} -Nk \\ K - k^2 \end{bmatrix} e^{kz} + \tilde{c} \begin{bmatrix} -N + Nkz \\ 2k + (K - k^2)z \end{bmatrix} e^{-kz} + \tilde{d} \begin{bmatrix} -N - Nkz \\ -2k + (K - k^2)z \end{bmatrix} e^{kz}.
$$
 (A7)

We convert to hyperbolic functions by introducing new constants,

 $\hat{a} = \tilde{a} + \tilde{b}, \quad \hat{b} = \tilde{a} - \tilde{b}, \quad \hat{c} = \tilde{c} + \tilde{d}, \quad \hat{d} = \tilde{c} - \tilde{d};$

this gives a new general solution of the form

$$
\begin{bmatrix} \tilde{v}_1 \\ \tilde{v}_3 \end{bmatrix} = \begin{bmatrix} Nk\hat{b} - N\hat{c} \\ (K - k^2)\hat{a} + 2k\hat{d} \end{bmatrix} \cosh(kz) + \begin{bmatrix} N\hat{d} - Nk\hat{a} \\ -(K - k^2)\hat{b} - 2k\hat{c} \end{bmatrix} \sinh(kz) + z \begin{bmatrix} Nk(\hat{d}\cosh(kz) - \hat{c}\sinh(kz)) \\ (K - k^2)(\hat{c}\cosh(kz) - \hat{d}\sinh(kz)) \end{bmatrix}.
$$

Finally, replacing all entries in the top line with simple constants via

$$
a = Nk\hat{b} - N\hat{c}, \quad b = N\hat{d} - Nk\hat{a}, \quad c = (K - k^2)\hat{a} + 2k\hat{d}, \quad d = -(K - k^2)\hat{b} - 2k\hat{c},
$$

we solve for \hat{c} and \hat{d} to obtain the convenient form of the solution given in Eq. [\(22\)](#page-3-0) in the main text.

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