

Fluidity and morphological stability of an amorphous thin film with radiation-induced defect kinetics

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It is common to model ion-irradiated amorphous thin films as if they were highly viscous fluids. In such models, one is frequently concerned with the ion-enhanced fluidity, a measure of the ability of the free interface to relax surface energy. Motivated by usual fluid dynamics problems, the ion-enhanced fluidity is near universally treated as a constant throughout the amorphous layer. However, for an irradiated thin film, the fluidity is ultimately caused by radiation-induced defect kinetics within the film, leading to regions with greater or lesser fluidity, and sensitive dependence on ion energy, species, flux, irradiation angle, temperature, and other experimental parameters. Here we develop and analyze a model of radiation-induced defect kinetics coupled to the continuum equations of a viscous thin film where sputtering-induced surface destabilization is neglected. Using realistic parameter values, we show that defect kinetics can meaningfully alter theoretical predictions of surface relaxation and ion-enhanced fluidity. Implications for aligning theoretical and experimental work, especially for surface nanopatterning of silicon induced by low-energy argon irradiation, are discussed.

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I. INTRODUCTION

Broad-beam low-energy ion bombardment of group IV semiconductors, such as silicon and germanium, leads to the destruction of the crystal lattice and the formation of an amorphous layer whose thickness is typically on the order of a few nanometers [1], and often the spontaneous self-organization of the irradiated surface into patterns [2–6]. Initial theoretical models aiming to describe this phenomenon focused on the sputter erosion of near-surface atoms [7] and later on the redistribution of atoms not sputtered away [8–11].

However, since the late 1990s, it has been proposed that stress accumulation and relaxation within the thin amorphous layer are important for determining the dynamics of semiconductor surfaces under irradiation, including pattern formation [1,3–6,12], or even the dominant mechanism driving pattern formation (see, e.g., [13–17]). With this viscous characterization of the film in mind, many contemporary models of the dynamics of these amorphous thin films posit that they behave as fluids with extremely high viscosity η and aim to understand them using techniques from fluid mechanics, especially hydrodynamic stability theory [5,13–25] or pattern formation theory [26,27]. We note that this class of viscous-flow models suggests an alternative explanation for pattern formation which contrasts with those models which consider

primarily erosion and redistribution (e.g., [7,9,10,28–30]) or kinetic surface processes [31–33]. In some cases, good agreement between viscous-flow-type models and experimental observations has been seen even from models excluding erosion and redistribution entirely (see, e.g., [16,17]). However, as noted in [5], caution is required: The nonlinear partial differential equations (PDEs) obtained from analyses of both viscous models and erosive-redistributive models share many of the same terms [5,9,22,33–35]. From such nonlinear PDEs, pattern formation behavior occurs generically. Hence, despite the apparent ability of recent models to reproduce experimental observations when parameters are chosen appropriately, it remains unclear how to obtain theoretical estimates of parameters such that new experimental results may be anticipated.

Moreover, and crucial to the present work, the analogy between classical fluid dynamics and irradiated thin films is imperfect. Stress accumulation and relaxation in the irradiated thin film are largely determined by implanted ions and the kinetics of flow defects (Frenkel pairs) created during irradiation [3,36–39]. These flow defects produce an ion-enhanced fluidity η^{-1} , which allows surface energy relaxation [40]. Exact values of η^{-1} for experimental systems of interest have been subject to much speculation due to the central importance that η^{-1} holds in modeling surface evolution in viscous models [5,10,20,39,41]. For lack of a comprehensive first-principles model, it has been commonly used as a *de facto* fit parameter [5,10,17,42], and both linear and nonlinear scalings of η^{-1} with ion flux and energy have been discussed [10,39,41]. When the angle dependence of η^{-1} has been considered, it has been assumed to follow $\cos(\theta)$ [5,20].

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In modeling surface evolution for irradiated semiconductors, a near-universal assumption is that η^{-1} is constant throughout the film, as is typically the case in traditional fluid mechanics problems, leading to surface energy relaxation according to the analysis of [40]. However, one does not generally expect that the distribution of defects, and therefore the ion-enhanced fluidity, should be constant throughout an irradiated film (see, e.g., [17,38,43]). Moreover, defect production within the amorphous layer shares its physical origin with sputter erosion: Both are fundamentally caused by energy deposition and the displacement of atoms and modeling one without the other, particularly within viscous-flow models, is a possible cause for concern, or at least a simplification whose innocuousness should be confirmed. In order to better understand the effect of defect kinetics on the morphological stability of the irradiated thin film, we do not consider the local sputtering rates, which lead to the Bradley-Harper instability. This is in following with the existing literature where theoretical models are commonly studied one mechanism at a time and then their linear superpositions are considered for broader comparison with experimental data (see, e.g., [20,21,24]).

Adding to the danger of treating the fluidity η^{-1} as spatially uniform, it has now been seen in several other contexts that spatial inhomogeneity of model parameters can introduce significant differences in model predictions of irradiated surface evolution: Spatially inhomogeneous energy distribution produces the well-known Bradley-Harper instability [7,30], local differences in density produce a stabilizing effect [21,24], and tying local density changes directly to ion implantation can greatly strengthen or weaken that stabilizing effect [44].

To understand how the defect kinetics determine the amorphous layer's ion-enhanced fluidity and induce a complex dependence of the fluidity on ion flux, energy, film temperature, and the distribution of defects within the amorphous layer, we study a generalization of a model due to [38]. This model was originally used to simulate defect kinetics resulting from normal-incidence irradiation of a flat substrate and explained residual stresses and thermal effects in a series of wafer-curvature experiments on irradiated copper, which does not become amorphous. Here we adapt the defect kinetics model from [38] to the case of off-normal incidence and couple it to a continuum model of a viscous fluid with a free surface. We use the resulting model to address two related questions. (i) When defect kinetics in the style of [38,39] are explicitly included in free-surface continuum models of an amorphous thin film, do any new phenomena emerge, particularly, new morphological instabilities or qualitative changes in viscous surface relaxation? (ii) Can a continuum model of the type studied here lead to quantitatively reasonable predictions in how the fluidity should change with flux, ion energy, irradiation angle, and other model parameters?

Throughout the history of theoretically modeling ion-induced pattern formation, multiple surface relaxation mechanisms have been proposed, used, and later refuted. The initial Bradley-Harper model used surface diffusion as a surface relaxation mechanism [7] and later removed it following [1]. An effective surface diffusion mechanism was proposed [28,29] and subsequently refuted [5,30] before broad agreement was reached that viscous surface relaxation was a better

explanation [1,5,20,45,46]. However, given the difficulty of obtaining a single unified model for ion-induced nanopatterning [5,6], it is natural to revisit prevailing modeling assumptions in search of important physics that those assumptions may have obscured: A central question is how far the viscous characterization of the irradiated thin film may be pushed.

The paper is organized as follows. First, we discuss the continuum model studied in the present work, which is motivated by background on both viscous thin films and a pre-existing model for radiation-induced defect kinetics. Second, we state the mathematical problem of interest: the steady-state equations, the derivation of equations for the growth of a single Fourier mode, a simplifying limit, and some details of numerical implementation. Finally, we consider the results from the model. We (i) compare linear-regime surface relaxation for thin films of constant fluidity [40] with thin films of spatially varying fluidity resulting from defect kinetics, (ii) comment on the use of an approximately $\cos(\theta)$ angle dependence of the mean fluidity $\eta^{-1}(\theta)$ [20], and (iii) explore the flux and energy dependence of $\eta^{-1}(\theta)$, with an eye towards validating commonly used analytical approximations which do not explicitly consider the defect kinetics [5,10,20]. All code needed to reproduce our results is publicly available [47].

II. MODEL

To describe the dynamics of the first few nanometers of the irradiated surface, we follow the approach found in the substantial literature on viscous-flow models of ion-induced nanopatterning [1,12–18,21–25,48] where this heavily damaged region is treated as a fluid of extremely high viscosity η . It is to this viscous-type model that we connect a separate model of defect kinetics [38]. For simplicity, we will restrict attention to the x (projected downbeam) and z (depthwise) axes, following the conventions of, e.g., [13–16]. Generalizing the model to three spatial coordinates is straightforward.

A. Conservation laws

Assuming an isothermal viscous fluid, the constitutive law

$$\mathbf{T} = -p(x, z, t)\mathbf{I} + 2\eta(x, z, t)\dot{\mathbf{E}} \quad (1)$$

is applied, with $\mathbf{T}$ the Cauchy stress tensor, $\mathbf{I}$ the identity tensor, p the hydrostatic pressure, and $\dot{\mathbf{E}} = \frac{1}{2}(\nabla \vec{v} + \nabla \vec{v}^T)$ the usual strain rate tensor. Here $\eta(x, z, t)$ is the local viscosity, an important departure from existing work. Conservation of mass and momentum are applied in differential form as

$$\begin{aligned} \frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \vec{v}) &= 0, \\ \nabla \cdot \mathbf{T} &= \vec{0}, \end{aligned} \quad (2)$$

respectively, where $\rho = \rho(x, z, t)$ is the local mass density and $\vec{v}$ is the local Eulerian velocity field. Above, conservation of momentum is written in the limit of vanishingly small Reynolds number ($\text{Re} \rightarrow 0$), as is standard for these systems (see, e.g., [13–18]). With Eq. (1), momentum conservation specializes to

$$-\nabla p + 2\nabla \cdot (\eta \dot{\mathbf{E}}) = \vec{0}. \quad (3)$$

These standard continuum equations for a viscous fluid are then coupled to the defect kinetics in the amorphous bulk.

B. Model of defect kinetics

We model the defect kinetics as a generalization of [38], where attention was restricted to normal-incidence irradiation and viscosity was not considered due to the experimental system of interest. The concentration fields of vacancies C_V and interstitials C_I , which have units in nm^{-3} , are governed by the nonlinear reaction-diffusion equations

$$\begin{aligned}\frac{\partial C_V}{\partial t} &= fY_V I(x, z; \theta) + D_V \nabla^2 C_V \\ &\quad - \frac{D_V C_V V_V}{k_B T} \frac{1}{3} \nabla^2 \text{tr}(\mathbf{T}) - 4\pi \lambda C_I C_V (D_I + D_V), \\ \frac{\partial C_I}{\partial t} &= fY_I I(x, z; \theta) + D_I \nabla^2 C_I \\ &\quad - \frac{D_I C_I V_I}{k_B T} \frac{1}{3} \nabla^2 \text{tr}(\mathbf{T}) - 4\pi \lambda C_I C_V (D_I + D_V),\end{aligned}\quad (4)$$

with f the nominal flux of ions and Y_V and Y_I defect production rates per unit energy for vacancies and interstitials, respectively. Defect production varies spatially throughout the amorphous layer according to the distribution of deposited energy, $I(x, z; \theta)$ [36,49,50]. Here θ is the angle of irradiation against a macroscopically flat surface, with $\theta = 0^\circ$ normal-incidence irradiation and $\theta \rightarrow 90^\circ$ grazing-incidence irradiation; D_I and D_V are diffusivities of interstitials and vacancies, respectively; V_I and V_V are the relaxation volumes of interstitials and vacancies [38]; k_B is the Boltzmann constant; T is the temperature in the thin film, which can be estimated theoretically [51,52] and is assumed constant in space and time; and λ is the mean distance over which an interstitial and a vacancy can recombine.

With such a complex model, explanations for each term are warranted. On the right-hand side of Eq. (4), termwise explanations are as follows. (i) The terms $fY_V I(x, z; \theta)$ and $fY_I I(x, z; \theta)$ provide the pointwise rate at which defects (Frenkel pairs) are produced. The parameters Y_V and Y_I are extractable from binary collision approximation (BCA) simulations (e.g., SRIM [49]) or estimated via the Kinchin-Pease formula [36]. In particular, $I(x, z; \theta)$ is the local rate of energy deposition, and defect production is proportional to this quantity. This term is the same as in the model of [38]. (ii) The terms $D_V \nabla^2 C_V$ and $D_I \nabla^2 C_I$ model the diffusion of defects away from areas of high concentration. While a more detailed model might express this diffusion in terms of the chemical potential energy, modeling as pure diffusion is typical and was sufficient for [38], whose work we follow here. (iii) The terms $-\frac{D_V C_V V_V}{k_B T} \frac{1}{3} \nabla^2 \text{tr}(\mathbf{T})$ and $-\frac{D_I C_I V_I}{k_B T} \frac{1}{3} \nabla^2 \text{tr}(\mathbf{T})$ describe the diffusion of defects by the stress gradient and are the most significant departure of the present treatment of defect kinetics from that of [38]. In [38] the corresponding terms are $\frac{D_V C_V V_V}{k_B T} \frac{\partial^2 \sigma}{\partial z^2}$ and $\frac{D_I C_I V_I}{k_B T} \frac{\partial^2 \sigma}{\partial z^2}$, where $\sigma = -\frac{1}{3} \frac{E}{1-\nu} [C_{\text{Ar}}(z, t) V_{\text{Ar}} + C_I(z, t) V_I + C_V(z, t) V_V]$ is the local isotropic stress with the assumption of an elastic substrate, with E the elastic modulus, ν Poisson's ratio, C_{Ar} the local concentration of implanted Ar^+ , and V_{Ar} the relaxation

volume of Ar^+ in the film. In the present work we have ignored the addition of Ar^+ to the film, as discussed below in Sec. II F. Unlike [38], we are interested in the growth rates of perturbations about the x axis, which requires generalization beyond the plane-stress condition while retaining the original physical meaning. To this end, we replace diffusion strictly along the z axis with biaxial diffusion. In [38] the isotropic stress is given by σ ; more generally, the isotropic stress is computed as $\frac{1}{3} \text{tr}(\mathbf{T})$, where $\mathbf{T}$ is the stress tensor given in Eq. (1) and tr is the trace. We note that if we had instead considered an elastic solid, as in [38], then the two expressions would agree in the case of an unperturbed free surface. Further discussion of the coefficients $\frac{D_V C_V V_V}{k_B T}$ and $\frac{D_I C_I V_I}{k_B T}$ and of stress-driven defect diffusion can be found in [53,54]. (iv) The terms $-4\pi \lambda C_I C_V (D_I + D_V)$ describe annihilation of defects due to recombination of vacancies and interstitials. The coefficient $4\pi \lambda (D_I + D_V)$ is the Smoluchowski rate constant.

Ultimately, and while acknowledging several modeling simplifications, we regard this model's plausibility and suitability for the present work as reinforced by its apparent explanatory power, as discussed in [38]. A similar model has also proven useful in explaining experimental observations of irradiation-induced stresses becoming "frozen in" [39].

C. Equations of state and fluidity

For closure, we require an equation of state. Somewhat generalizing [21,23,24], we apply a simple quasi-incompressibility equation of state

$$\rho(x, z, t) = \frac{m_0}{V_0 + C_I V_I + C_V V_V}, \quad (5)$$

where m_0 is atomic mass and V_0 is atomic volume. This says that density changes can only result from local defects and are not explicitly due to pressure. Despite its simplicity, similar equations of state have been successfully used elsewhere [21,23–25,48,55–57] as an approximation. Note that identifying relative volumization $\Delta = C_I \frac{V_I}{V_0} + C_V \frac{V_V}{V_0}$ and basic density $\rho^* = \frac{m_0}{V_0}$ recovers the form

$$\rho(x, z, t) = \frac{\rho^*}{1 + \Delta}, \quad (6)$$

which has been established elsewhere [21,24]. As a description of the ion-enhanced fluidity, we suppose

$$\frac{1}{\eta} = \frac{1}{\eta^*} + \left(\frac{C_I}{\eta_I} + \frac{C_V}{\eta_V} \right) \quad (7)$$

or, equivalently, the viscosity $\eta = [\frac{1}{\eta^*} + (\frac{C_I}{\eta_I} + \frac{C_V}{\eta_V})]^{-1}$. This makes the assumption, which has been used elsewhere [10,39], that the local ion-enhanced fluidity is linear in the number of flow defects, with η_I^{-1} the contribution to fluidity by interstitials and η_V^{-1} the contribution to fluidity by vacancies. The parameter $1/\eta^*$ is a baseline fluidity in the absence of any defects, which is assumed to be very small.

D. Modeling defect production

In the governing equations of the defect fields, we take the flux-diluted energy deposition [3,5,7,30,58,59] through a free

surface $h(X, t)$,

$$I(x, z, t) = \int_{-\infty}^{\infty} \frac{\cos(\theta) + h_x \sin(\theta)}{\sqrt{1 + h_x^2}} E(x, z; X, h(X, t)) dX, \quad (8)$$

where

$$E(x, z; X, h(X, t)) = \frac{E_0}{2\pi\alpha\beta} \exp\left(-\frac{\{(x - X) \sin(\theta) - [z - h(X, t)] \cos(\theta) - a\}^2}{2\alpha^2} - \frac{\{(x - X) \cos(\theta) + [z - h(X, t)] \sin(\theta)\}^2}{2\beta^2}\right) \quad (9)$$

is the energy deposited at a given location in the film, which results in the creation of Frenkel pairs. Elsewhere, it has been shown that $E(x, z; X, h(X, t))$ is reasonably well approximated as a bivariate Gaussian ellipsoid in downbeam-crossbeam coordinates [3,7,30,58–61]. Here a is the downbeam depth where energy deposition is greatest, corresponding to the downbeam depth that ions penetrate before initiating the collision cascade, α is the downbeam standard deviation, and β is the crossbeam standard deviation.

In the interior, this energy deposition leads to two important phenomena: (i) sputtering of atoms that receive enough energy to exit through the free interface (which leads to the classical Bradley-Harper mechanism) and (ii) the creation of Frenkel pairs. The nonuniform sputtering rates through the surface would be incorporated into the $V(\theta)$ term if we included the dependence on local slope h_x . However, because the Bradley-Harper mechanism is already well understood, we focus instead on the defect-producing effects of energy deposition, taking $V(\theta)$ constant across the free surface. In the limit of small surface perturbations and vanishing cross terms, it is typical to consider a composite growth rate Σ as the linear superposition of the growth rates associated with several distinct models, as in, e.g., [20].

E. Boundary conditions

As the surface is irradiated, material is sputtered away from the free interface [58], and newly exposed crystalline substrate can be amorphized. Hence the present model describes a thin film translating in the direction of the ion beam, as in [21,23–25], where a translating reference frame is introduced. The ongoing erosion of the free interface leads to the modified kinematic equation [21] [Eq. (10b)] and an apparent nonzero velocity at the amorphous-crystalline interface [Eq. (11a)] [21,23–25]. At the free surface $z = h(x, t)$,

$$\mathbf{T} \cdot \hat{\mathbf{n}} = \gamma \kappa \hat{\mathbf{n}}, \quad (10a)$$

$$v_I = \vec{v} \cdot \hat{\mathbf{n}} - V(\theta) \frac{\rho^*}{\rho}, \quad (10b)$$

where γ is surface energy and κ is the mean curvature; in one dimension, $\kappa = \frac{h_{xx}}{(1+h_x^2)^{3/2}}$. In addition, $\hat{\mathbf{n}}$ is the outward-normal unit vector from the free surface, v_I is the outward-normal interfacial speed (i.e., $\frac{h_t}{\sqrt{1+h_x^2}}$ in one dimension), and $V(\theta)$ is the speed of downward translation (in laboratory coordinates) of the film due to erosion [7,30,58,59]. Equation (10a) is stress balanced at the interface and the second is the kinematic

equation modified to account for ongoing sputtering [21]. At the lower interface $z = g(x, t)$,

$$\vec{v} = V(\theta) \hat{\mathbf{k}}, \quad (11a)$$

$$\Delta = \frac{\partial \Delta}{\partial \hat{n}} = \frac{\partial^2 \Delta}{\partial \hat{n}^2} = 0, \quad (11b)$$

where $\Delta = C_I \frac{V_I}{V_0} + C_V \frac{V_V}{V_0}$, as above, and $\hat{\mathbf{k}}$ is a unit vector along the z axis. These boundary conditions for C_I and C_V are chosen to describe a sharp interface between the amorphous and crystalline phases. In principle, more sophisticated boundary conditions could be chosen, but we regard these as sufficient for an initial study. The relationship between $g(x, t)$ and $h(x, t)$ is characterized by the interface relation provided by [25], where $g(x, t) = h(x - x_0(\theta), t) - h_0(\theta)$ and

$$\begin{aligned} h_0(\theta) &= a \cos(\theta) + 2\sqrt{\alpha^2 \cos^2(\theta) + \beta^2 \sin^2(\theta)}, \\ x_0(\theta) &= a \sin(\theta) + \frac{2(\alpha^2 - \beta^2) \sin(\theta) \cos(\theta)}{\sqrt{\alpha^2 \cos^2(\theta) + \beta^2 \sin^2(\theta)}}. \end{aligned} \quad (12)$$

In order to focus on the influence of the defect kinetics, the effect of phase change at the amorphous-crystalline interface [25] is ignored.

F. Model of defect kinetics

Some comments on this model are in order. While embracing the complexity of including defect kinetics in a model of a viscous thin film, three important simplifications are still made for tractability. (a) We suppose that the primary effect of implanted ions is to produce Frenkel pairs, while neglecting implantation of noble gas ions and their interactions with the amorphous layer, which were explicitly considered in [38]. This simplification is made (i) in order to leave our statement of mass conservation unchanged, (ii) to avoid the need to model bubble formation and other effects [38,62], and (iii) to facilitate a relatively simple first study. However, by neglecting ion implantation, we sacrifice the ability to seek quantitative agreement with ion-induced stresses [38,43]. (b) We assume that there is no meaningful thermal gradient in the film. In principle, considering the thermal gradient would mean that we would also need to include spatially inhomogeneous diffusivities D_I and D_V , a major increase in complexity on top of an already-complex model that we will defer to future work. Moreover, it has been argued elsewhere that a thin film with thickness on the order of a few nanometers should be unable to sustain a significant thermal gradient [16]. (c) We do not take into account removal of defects by

sputtering. We assume that the recombination of Frenkel pairs is the only means of eliminating them, as in [39].

III. ANALYSIS

A. Linear stability analysis in Fourier modes

We are interested in (i) the steady-state form of $\eta^{-1}(\theta)$ and (ii) the effect of defect dynamics on linear-regime surface relaxation, which motivates a linear stability analysis of the continuum system described above. Our linear stability analysis follows the style of [27], which we briefly summarize as follows.

For Eqs. (1)–(11) we restrict attention to the x and z axes and compute the steady-state solution. Steady-state solutions should exhibit time invariance ($\frac{\partial}{\partial t} \rightarrow 0$). We also assume translation invariance along the extended axis ($\frac{\partial}{\partial x} \rightarrow 0$). Having obtained this translation-invariant steady-state solution, to be defined $\tilde{\Psi}_0(z) = [p_0(z), u_0(z), w_0(z), C_{V,0}(z), C_{I,0}(z)]^T$, we seek the system's linear response to vanishingly small perturbations of the steady state via a two-term asymptotic expansion $\tilde{\Psi}(x, z, t) = \tilde{\Psi}_0(z) + \epsilon \tilde{\Psi}_1(x, z, t)$, where $\epsilon \approx 0$ and $\tilde{\Psi}_1(x, z, t) = [p_1(x, z), u_1(x, z), w_1(x, z), C_{V,1}(x, z), C_{I,1}(x, z)]^T$ must be determined. To this end, one considers periodic boundaries on the extended x axis, which motivates the use of the ansatz e^{ikx} for the x dependence of $\tilde{\Psi}_1$. The resulting partial differential equations are separable in t so that one anticipates temporal scaling $e^{\Sigma t}$. Together, this motivates the ansatz $\tilde{\Psi}(x, z, t) = \tilde{\Psi}_0(z) + \epsilon \tilde{\Psi}_1(z) e^{\Sigma t + ikx}$, where $\tilde{\Psi}_1(z) = [\tilde{p}_1(z), \tilde{u}_1(z), \tilde{w}_1(z), \tilde{C}_{V,1}(z), \tilde{C}_{I,1}(z)]^T$. The interfaces are similarly linearized as $h(x, t) = h_0(\theta) + \epsilon \tilde{h}_1 e^{\Sigma t + ikx}$ and $g(x, t) = \epsilon \tilde{h}_1 e^{\Sigma t + ik[x - x_0(\theta)]}$, that is, one expects a corrugated lower (amorphous-crystalline) interface $z = g$ that is a phase-shifted and vertically displaced copy of the upper (free) interface $z = h$ [25,44]. The assumption that the amorphous-crystalline interface $z = g$ is a simple transformation of $z = h$ is nontrivial, and this point has been explored in greater detail elsewhere [25,44].

Following this, all t dependence is subsumed by Σ , and all x dependence is subsumed by k . One solves the resulting system of ordinary differential equations and boundary conditions as a nonlinear eigenvalue problem for $\tilde{\Psi}_1(z)$ and the growth rate Σ [26,27,63]. Hence Σ provides the rate at which an idealized sinusoidal perturbation of wave number k to the free interface $z = h$ grows in time. This growth rate provides two natural points of comparison with experimental observations. First, the experimentally observed critical angle θ_c beyond which pattern formation initiates at the free interface $z = h$ should correspond to the least θ for which $\Sigma > 0$ for some wave number k . Second, for $\theta > \theta_c$ and short irradiation times, one expects that the wave number of experimentally observed ripples should agree with the fastest-growing wave number k predicted by the theoretical Σ . (See, e.g. [13–18,21–25] for many analyses of this type for viscous-flow models, [7,9–11,30,64] for analyses of pattern-forming models not of the viscous type, and [3–6] for general discussion.)

Within the analysis described above, one assumes translation invariance with respect to x of the steady state, as

is typical (see, e.g., [13–18,21–25]). This assumption, while restrictive, is not wholly unjustified, even if the justification is somewhat heuristic. We first note that an unirradiated wafer is expected to be spatially uniform in its properties. From the linearized form of Eq. (8), it appears that the distribution of ions, energy, defect production, or any other quantity correlated with (4) is itself translation invariant with respect to x when it results from irradiation through a flat free interface $z = h$. Since all transport within the amorphous layer is expected to originate from (4) or other isotropic mechanisms like diffusion, it is unclear whence the x dependence would arise. Nonetheless, a more cautious analysis may be eventually warranted.

To obtain the systems of differential equations constituting the mathematical problem of the present work, we require a linearization of Eq. (8). Following [44], its linearization about a perturbed interface $I(x, z, t) = I_0(z) + \epsilon I_1(z) \exp(\Sigma t + ikx)$, suitable to linear stability analysis for a single Fourier mode of wave number k , is computed. We use the approximation of [44] here as

$$I_0(z) = \frac{\cos(\theta)}{\sqrt{2\pi(\alpha^2 c^2 + \beta^2 s^2)}} \exp\left(-\frac{(z - h_0 + ac)^2}{2(\alpha^2 c^2 + \beta^2 s^2)}\right) \quad (13)$$

and

$$I_1(z) = \frac{\cos(\theta)}{2\sqrt{\pi A \alpha^3 \beta^3}} \exp\left(\frac{\tilde{B}^2(z)}{4A} - C(z)\right) \times \left(c_1(z) - c_2 \frac{\tilde{B}(z)}{2A}\right) \tilde{h}_1, \quad (14)$$

where

$$\begin{aligned} A &= \frac{\beta^2 s^2 + \alpha^2 c^2}{2\alpha^2 \beta^2}, \quad B(z) = \frac{(z - h_0)sc(\alpha^2 - \beta^2) - a\beta^2 s}{\alpha^2 \beta^2}, \\ \tilde{B}(z) &= \frac{(z - h_0)sc(\alpha^2 - \beta^2) - a\beta^2 s}{\alpha^2 \beta^2} + ik, \\ C(z) &= (z - h_0)^2 \frac{\beta^2 c^2 + \alpha^2 s^2}{2\alpha^2 \beta^2} + \frac{a(z - h_0)c}{\alpha^2} + \frac{a^2}{2\alpha^2}, \\ c_1(z) &= (z - h_0)[\alpha^2 \sin^2(\theta) + \beta^2 \cos^2(\theta)] \\ &\quad + \cos(\theta)a\beta^2 + ik\alpha^2 \beta^2 \tan(\theta), \\ c_2 &= \cos(\theta) \sin(\theta)(\alpha^2 - \beta^2), \end{aligned} \quad (15)$$

and $c = \cos(\theta)$ and $s = \sin(\theta)$ throughout. The above $I_0(z)$ and $I_1(z)$ provide the local rates of energy deposition into the film. These are converted to pointwise rates of defect production using the Kinchin-Pease relation [36,38] via the terms $fY_V I(x, z; \theta)$ and $fY_I I(x, z; \theta)$ in Eqs. (4).

B. Steady-state equations in a simplifying limit

The full steady-state equations are shown in the Appendix, but they elude analytical solution. Moreover, the equations for defect concentrations C_I and C_V produce a pair of coupled third-order equations that are difficult to solve, even using numerical methods. However, for some systems of interest, like amorphous silicon, the diffusion rate of vacancies, D_V , may be two or more orders of magnitude larger than that of interstitials, D_I [65,66].

This suggests the use of the $\frac{D_I}{D_V} \rightarrow 0$ limit, which enables a pseudo-steady-state approximation commonly used in reaction kinetics. This simplification, described in the Appendix, produces a single nonlinear third-order ordinary differential equation for steady state C_{V0} ,

$$fY_V I_0(z) + D_V C_{V,0zz} + \frac{D_V}{3} \left(\frac{V_V}{k_B T} \right) C_{V,0} p_{0zz} - fY_I I_0(z) = 0, \quad (16)$$

when we express p_0 , ρ_0 , and η_0 as

$$\begin{aligned} p_0 &= -2V(\theta)\eta_0 \frac{\rho_{0z}}{\rho_0^2}, \\ \rho_0 &= \left(1 + C_{V0} \frac{V_V}{V_0} + \frac{fY_I I_0 V_I}{4\pi\lambda C_{V0} D_V V_0} \right)^{-1}, \\ \eta_0 &= \left(\frac{1}{\eta^*} + \frac{C_{V0}}{\eta_V} + \frac{fY_I I_0}{4\pi\lambda C_{V0} D_V \eta_I} \right)^{-1} \end{aligned} \quad (17)$$

so that Eq. (16) can be written in terms of $C_{V0}(z)$, its derivatives, and assigned parameters. For readability, we record this lengthy equation in the Appendix.

Equations (11), again in the $\frac{D_I}{D_V} \rightarrow 0$ limit, lead to the following boundary conditions at the amorphous-crystalline interface $z = 0$:

$$\begin{aligned} C_{V0} &= \sqrt{\frac{-fY_I V_I}{4\pi\lambda D_V V_V}} \sqrt{I_0(z)}, \\ C_{V0z} &= \sqrt{\frac{-fY_I V_I}{4\pi\lambda D_V V_V}} \frac{I_0'(z)}{\sqrt{I_0(z)}}, \\ C_{V0zz} &= \sqrt{\frac{-fY_I V_I}{4\pi\lambda D_V V_V}} \left(\frac{I_0''(z)}{\sqrt{I_0(z)}} - \frac{1}{2} \frac{[I_0'(z)]^2}{I_0(z)^{3/2}} \right). \end{aligned} \quad (18)$$

Above, we have observed that p_0 , ρ_0 , and η_0 are entirely expressible in terms of $C_{V0}(z)$. In the Appendix we also show that $w_0 = \frac{V(\theta)\rho^*}{\rho_0}$ and $u_0 = 0$. Hence the solution for $C_{V0}(z)$ fully determines the steady state. The $O(\epsilon)$ equations for the growth rates Σ are much more complicated, and their simplifications under the pseudo-steady-state approximation are less dramatic. We defer them to the Appendix for brevity.

C. Linear stability analysis in Fourier modes

Similar to [51,52], we estimate the temperature of the irradiated wafer using the point model

$$\frac{dT}{dt} = \frac{1}{\rho_c C_v h_0} [fE_{\text{ion}} - \sigma_{\text{SB}} \epsilon_{\text{em}} (T^4 - T_{\text{env}}^4)], \quad (19)$$

where C_v is the specific heat, E_{ion} is the nominal ion energy, $\sigma_{\text{SB}} \approx 3.54 \times 10^{-7} \text{ eV s}^{-1} \text{ nm}^{-2} \text{ K}^{-4}$ is the Stefan-Boltzmann constant, T is the wafer temperature, and T_{env} is the temperature of the environment. This model simply states that the temperature change over time is proportional to the energy flux. For simplicity, we assume (i) that the only source of energy is the kinetic energy of incoming ions, fE_{ion} , and (ii) that the only way that energy is lost is by thermal radiation, $\sigma_{\text{SB}} \epsilon_{\text{em}} (T^4 - T_{\text{env}}^4)$, which is the Stefan-Boltzmann law. This leads to a predicted steady-state temperature $T =$

$(T_{\text{env}}^4 + \frac{fE_{\text{ion}}}{\sigma_{\text{SB}} \epsilon_{\text{em}}})^{1/4}$, which we use throughout the present work. We use $\epsilon_{\text{em}} = 0.75$ as an estimate for the effective emissivity of a typical irradiated Si wafer in an open-back wafer holder [51]. We take $T_{\text{env}} = 294 \text{ K}$ unless stated otherwise. These parameter values lead to reasonable agreement with heating observed during ion bombardment in [39,67–69] and elsewhere and are sufficient for our present purposes.

D. Estimating translation speed $V(\theta)$

We require an estimate of the speed of the film's downward (i.e., into-the-substrate) translation due to erosion, $V(\theta)$ [21,23–25]. Rather than performing a large number of simulations using, e.g., [49,50] or other BCA tools, we use the semiempirical Yamamura approximation of sputter yield $Y(\theta)$ [70,71],

$$Y(\theta) = Y(0) \sec^{f_Y}(\theta) \exp\{-\Sigma_Y [\sec(\theta) - 1]\}, \quad (20)$$

which has been commonly used (see, for example, [34,72,73]). Above, $f_Y = f_S [1 + 2.5(\frac{1-\xi}{\xi})]$, $\xi = 1 - \sqrt{\frac{E_{\text{th}}}{E_{\text{ion}}}}$, $\Sigma_Y = f_Y \cos(\theta_{\text{opt}})$, $\theta_{\text{opt}} = 90^\circ - 286\psi(E_{\text{ion}})^{0.45}$, and $\psi(E_{\text{ion}}) = \frac{\psi(1 \text{ eV})}{\sqrt{E_{\text{ion}}}}$. The parameters E_{th} , f_S , and $\psi(1 \text{ eV})$ are ion- and target-dependent quantities that can be looked up in the tables of [70]. For Ar^+ -irradiated Si, $E_{\text{th}} = 33 \text{ eV}$, $f_S = 1.85$, and $\psi(1 \text{ eV}) = 0.0895$ [70]. To compute the normal-incidence sputter yield $Y(0)$ for any particular E_{ion} , we again use the formulas provided in [70], which we do not record here for brevity. Then $V(\theta) = f \cos(\theta) Y(\theta) V_0$ is easily computed for Ar^+ -irradiated Si at any flux, angle, and energy of interest. We remind the reader that because θ is the beam angle against a macroscopically flat surface, $V(\theta)$ is constant across the surface, as in, e.g., [21,23].

E. Scaling of a , α , β , Y_V , and Y_I with E_{ion}

Elsewhere [74], it has been observed that ion implantation parameters a , α , and β scale roughly with $\sqrt{E_{\text{ion}}}$ for a fixed ion-target combination at low energies and have negligible angle dependence [60,61] for irradiation angles not “too close” to grazing. In Sec. IV we make use of this simple scaling argument to obtain results for Ar^+ -irradiated Si at different energies by scaling a , α , and β obtained for 1-keV Ar^+ -irradiated Si using SRIM [49,50]. To scale per-ion defect production rates Y_V and Y_I with energy, we apply the usual Kinchin-Pease relationship [10,36,75] $Y_V = \frac{0.8E}{E_D}$ and $Y_I = \frac{0.8E}{E_D}$, where $E_D = 15 \text{ eV}$ is the displacement energy for a Si atom.

F. Numerical implementation

The nonlinear system of boundary value problems of Eqs. (16)–(18) is formidable; we make no serious attempt at solving it using analytical techniques. Instead, we turn to numerical solution using Matlab's `bvp5c`, first numerically solving (16)–(18) and then passing steady-state solutions to a second solver for the eigenvalue problem represented by the $O(\epsilon)$ equations discussed in the Appendix. This produces a full set of solutions $w_0(z)$, $u_0(z)$, $p_0(z)$, $\rho_0(z)$, $\eta_0(z)$, $C_{V0}(z)$, and $C_{I0}(z)$ in the steady state and $\tilde{w}_1(z)$, $\tilde{u}_1(z)$, $\tilde{p}_1(z)$, $\tilde{\rho}_1(z)$, $\tilde{\eta}_1(z)$, $\tilde{C}_{V1}(z)$, $\tilde{C}_{I1}(z)$, and Σ for the $O(\epsilon)$ equations. To avoid

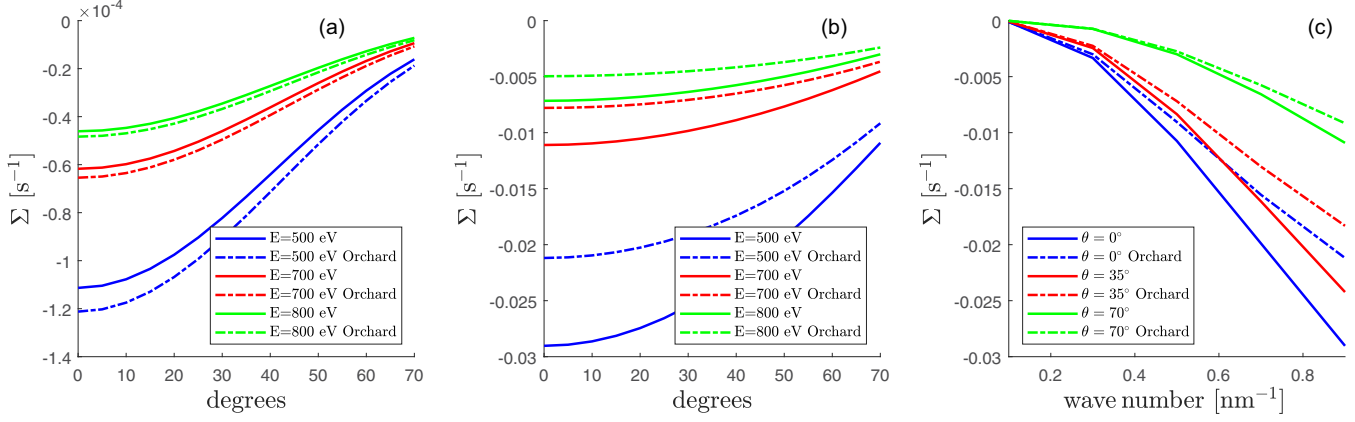


FIG. 1. Comparison of linear growth rate Σ due to Orchard [40] (dashed lines) and as computed in the present work (solid lines) for selected energies, wave numbers, and irradiation angle: $f = 2$ ions/nm²/s and (a) $k = 0.1$ nm⁻¹, (b) $k = 0.9$ nm⁻¹, and (c) $E = 500$ eV. There is broad agreement between the two, which worsens somewhat at higher wave numbers and lower fluxes.

loss of numerical precision, all stated parameter values are converted to units of Å, ns, K, and Gamu (i.e., 10⁹ amu) in Matlab.

IV. RESULTS

The model (1)–(11) contains too many adjustable parameters for an exhaustive survey of parametric dependence. To reduce the number of free parameters, we model the irradiation of Si by Ar⁺, where many parameter values have already been estimated. Accordingly, we assign relaxation volumes $V_I = V_0 = -V_v$ and $V_0 = 0.02$ nm³ and atomic mass $m_0 = 28.09$ amu, corresponding to Si. Since interstitials and vacancies occur in pairs [36], we assign defect yields for vacancies and interstitials $Y_V = Y_I$. We also assign $\lambda = 0.237$ nm, representing a typical nearest-neighbor distance between silicon atoms in a crystalline structure. As in [38], we find that the choice of λ is not a strong influence on our results.

Here parameter estimation is a significant issue, and obtaining reasonable estimates of model parameters is an area of ongoing interest (see, e.g., [39,65,66,76,77]). For lack of high-precision parameter estimates, we consider a range of plausible values, aiming to study the qualitative behavior of the present model. We assign baseline viscosity $\eta^* = 10^5$ GPa⁻¹ s⁻¹, exceeding experimental estimates by two or more orders of magnitude (see, e.g., [20,41,69]). We also assign the fluidity-per-defect parameters $\eta_I^{-1} = \eta_V^{-1} = 2.39$ nm³ GPa⁻¹ s⁻¹, as estimated in [39], where Ar⁺-irradiated Si was studied. To compute temperature-dependent diffusivities D_V and D_I , we apply Arrhenius relations [77] $D_V(T) = D_{V0} e^{-Q_r/k_B T}$ and $D_I(T) = D_{I0} e^{-Q_r/k_B T}$, where temperature T is computed as described in Sec. II. We assign isoconfigurational activation enthalpy $Q_r = 2$ eV, within the range of values discussed in, e.g., [65,77]. We choose plausible coefficients $D_{I0} = 10^{25}$ nm²/ns and $D_{V0} = 10^{27}$ nm²/ns so that $D_I \approx 10^{-8}$ nm²/ns and $D_V \approx 10^{-6}$ nm²/ns at room temperature, within the range of values from [39,65,66,76]. Surface energy γ is assigned as 1.36 J/m² for Si, as in [78,79]. We will also consider $D_{I0} = 10^{23}$ nm²/ns and $D_{V0} = 10^{25}$ nm²/ns to better understand the influence of lower diffusivities.

A. Linear stability results

We compare the numerical results for linear growth rate Σ of the present model with the classical Orchard result [40] for that of a fluid of uniform η^{-1} , which reads

$$\Sigma = -\frac{\gamma \eta^{-1}}{2h_0(\theta)} \frac{Q[\sinh(2Q) - 2Q]}{1 + 2Q^2 + \cosh(2Q)}, \quad (21)$$

where $Q = kh_0(\theta)$. For the purposes of comparing the form of Eq. (21), where η^{-1} is assumed uniform, with the growth rate due to the present model, where η^{-1} is not, we assign the uniform fluidity η^{-1} in (21) as $\langle \eta^{-1} \rangle$, the mean fluidity across the film depth, computed using the present model. This allows for a fair, if somewhat simplified, quantitative comparison between predicted relaxation rates using [40] and those of the present model. In Fig. 1 we compare linear growth rates Σ due to Orchard with those of the present work for a fixed flux f and a few wave numbers k , ion energies E , and angles of incidence θ , with other parameter values assigned as discussed above. In Fig. 2 we produce results for the same set of parameters, except with the basic diffusivities D_{V0} and D_{I0} assigned two orders of magnitude lower: $D_{I0} = 10^{23}$ nm²/ns and $D_{V0} = 10^{25}$ nm²/ns instead of $D_{I0} = 10^{25}$ nm²/ns and $D_{V0} = 10^{27}$ nm²/ns. Here a few observations can be made. (i) The linear growth rate of the present work exhibits the same dependence on θ and k as in the work of Orchard [Eq. (21)] up to the assignment of η^{-1} . At higher energies, the agreement is stronger; we attribute this to thermally enhanced diffusion tending to uniformize the distribution of defects, leading to a more uniform fluidity. (ii) Differences in linear growth rates Σ are more pronounced at lower energies and at higher wave numbers. The intuitive explanation for the energy dependence rests on noting that higher energies lead to greater temperatures, which enhance diffusion, and lead to more uniform η^{-1} across the film. For the wave-number dependence, one notes that the effect of geometric flux dilution [5,44] on the distribution of deposited energy in the amorphous layer is least for flat surfaces ($k \rightarrow 0$) and increases with k . However, for most wave numbers experimentally observed (below around 0.4 nm⁻¹, corresponding to wave numbers larger than 15 nm [14,15,17,20,68]), the difference is on the order of

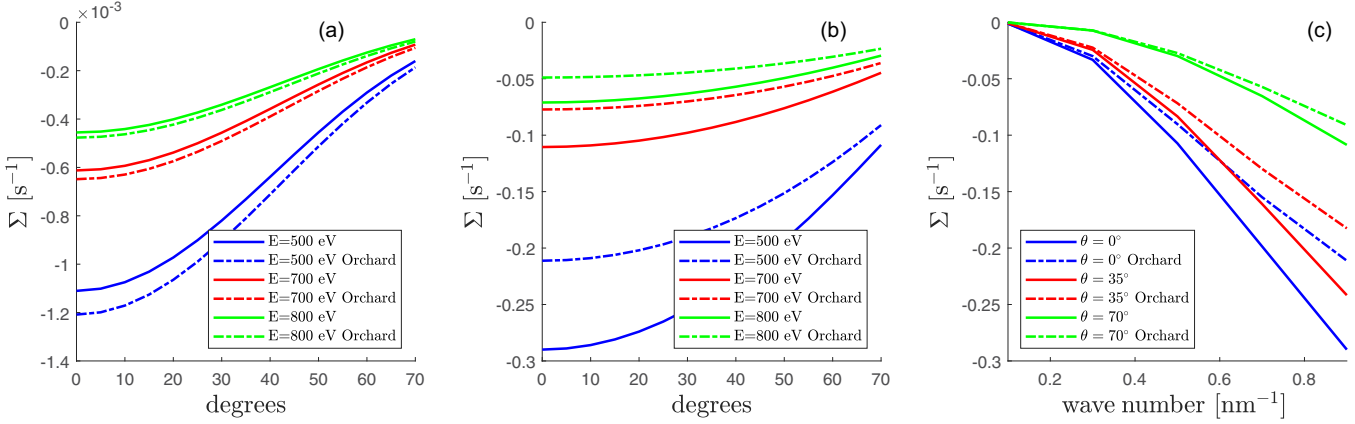


FIG. 2. Comparison of linear growth rate Σ due to Orchard [40] (dashed lines) and as computed in the present work (solid lines), as in Fig. 1, while using D_{A0} and D_{B0} two orders of magnitude lower. The curves agree with those shown in Fig. 1 up to a scaling factor of 10.

10%. (iii) The effect of decreasing diffusivities by two orders of magnitude is simply to reduce the linear growth rate of the present model by one order of magnitude. This observation is immediately useful; despite the difficulty surrounding parameter estimation for models of irradiated thin films [5], any qualitative results that we obtain here should survive refinements of diffusivity values. However, the present model, with D_{V0} and D_{I0} tuned to produce reasonable agreement with [66] at room temperature, leads to a contribution to the linear growth rate due to viscous surface energy relaxation of the same order of magnitude as experimentally observed in [20]. This also lends some confidence that the diffusivities used here are reasonable.

Hence the form of Eq. (21) is essentially correct, supposing that the value of $\langle \eta^{-1} \rangle$ is known. We do not find evidence within the present model that defect kinetics alone may produce their own morphological instability.

B. Results for the fluidity

When modeling irradiated surfaces as viscous thin films, an estimate of η^{-1} is necessary to obtain predictions of angle-dependence wavelengths, bifurcation angle, roughening, and other common points of comparison between theory and experiment [13–17]. Its parametric dependence on flux f , nominal beam energy E , and irradiation angle θ has been a subject of interest and speculation. Suggestions for flux dependence have included $\eta^{-1}(\theta)/\eta^{-1}(0) \propto f \cos(\theta)$, as in [20]; $\eta^{-1}(\theta)/\eta^{-1}(0)$ constant, as in most of the theoretical literature (see, e.g., [15–17,22]); and $\eta^{-1}(\theta)/\eta^{-1}(0) \propto \sqrt{f}$, implied by [39]. The fluidity η^{-1} has also been broadly assumed to scale with $\frac{E}{h_0}$ [20], which would suggest a roughly $\sqrt{E}$ dependence for constant f and θ [74].

In Fig. 3 we show the angle, flux, and energy dependence of $\langle \eta^{-1} \rangle$ under various experimental conditions. Several aspects of the data are noteworthy. (i) For the assigned parameters D_{V0} , D_{I0} , and Q_r , increased flux can lead to reductions in fluidity when the increased heating produces much faster annealing of defects. However, when temperature is controlled (here either to room temperature or to 100°C), clear flux scalings can be identified. In particular, we observe a $\sqrt{f}$ scaling of $\langle \eta^{-1} \rangle$ at fixed energy and irradiation angle,

consistent with [39]. (ii) Across fluxes and energies, an angle dependence close to $\sqrt{\cos(\theta)}$, rather than $\cos(\theta)$, is observed. Taken alongside the above observation about f , this suggests a flux-diluted scaling like $\sqrt{f \cos(\theta)}$ for constant energy, regardless of heating. (iii) When temperature is controlled, energy dependence is far weaker than previously believed, showing an $E^{1/4}$ scaling at constant flux and irradiation angle. Otherwise, increased energy leads to increased heating, which can anneal away defects by enhancing diffusion and recombination. This result is again consistent with the fluidity scaling implied by [39] (see also [48]).

V. DISCUSSION

A. Conclusions

In this work we have developed and studied a model that combines a common modeling simplification, i.e., treating the amorphous layer as a viscous thin film, with a model for defect kinetics, where the local defect concentration determines local fluidity. Radiation-induced heating was taken into account by adjusting the diffusivity rates of the defects. In an effort to minimize free parameters and obtain physically meaningful results, we have used plausible parameter values due to [51,52,66,76–79], while acknowledging that many are difficult to obtain and remain uncertain. Nonetheless, our central qualitative results persist despite some uncertainty in the exact parameters. We have also explicitly excluded the Bradley-Harper instability [7,30] in order to focus instead on how flow defects contribute to surface stability.

We have obtained two sets of results: one pertaining directly to the mean ion-enhanced fluidity $\langle \eta^{-1} \rangle$ and its angle, flux, and energy dependence in the steady state and one pertaining to the linear stability effects of viscous surface relaxation in the presence of spatially inhomogeneous defects. Our main findings are summarized as follows. (i) The form (21) is consistent with surface relaxation of an amorphous thin film even in the presence of defects, provided that η^{-1} can be assigned. In particular, differences between (21) and the decay rate of surface perturbations predicted by the present work disappear in the limit of small wave number k . Hence the long-wave approximation of (21) is validated

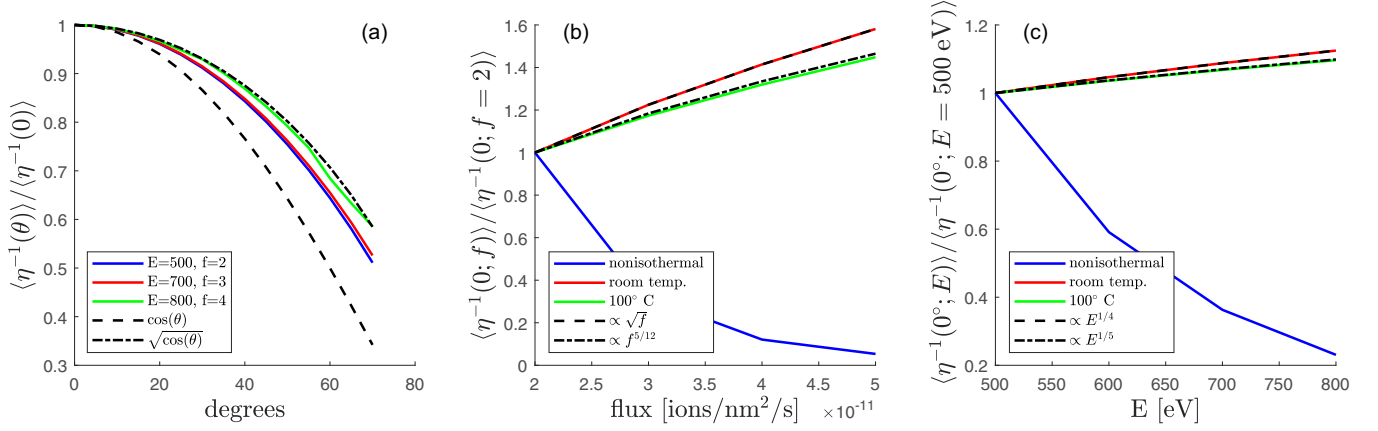


FIG. 3. (a) Angle dependence of η^{-1} (nonisothermal) for selected energies and fluxes. For comparison, $\cos(\theta)$ and $\sqrt{\cos(\theta)}$ are plotted. The data are evidently consistent with a $\sqrt{\cos(\theta)}$ dependence at these energies and fluxes. (b) Flux dependence under selected thermal conditions: $\theta = 0^\circ$ and $E = 500$ eV. When temperature is controlled, and especially near room temperature, η^{-1} scales approximately as $\sqrt{f}$. (c) Energy dependence of η^{-1} for $\theta = 0^\circ$ and $f = 2$ ions/nm²/s. When temperature is controlled, and especially near room temperature, η^{-1} scales with $E^{1/4}$.

for seeking theoretical predictions of bifurcation angles. (ii) We find that depth-averaged fluidity $\langle \eta^{-1} \rangle$ near room temperature is expected to scale with flux as $\sqrt{f}$, with angle as $\sqrt{\cos(\theta)}$, and with energy as $E^{1/4}$. This is in contrast with the scaling arguments that have been advanced elsewhere for η^{-1} [10,20,41,42,80], where the defect kinetics were not explicitly considered. Notably, the $\sqrt{f}$ scaling of η^{-1} is consistent with the model and experimental work of [39]. Alongside the observation that, roughly, $h_0(\theta) \propto E^{1/2}$ [74], this suggests a scaling law consistent with $\sqrt{\frac{f \cos(\theta) E(\theta)}{h_0(\theta)}}$, an intriguing modification of the scaling arguments that have been considered elsewhere [10,20,41,42,80]. (iii) Using plausible values of Q_r , D_V , D_I , and ϵ_{em} (isoconfigurational activation enthalpy, diffusivity of vacancies, diffusivity of interstitials, and emissivity, respectively), thermal effects can lead to a loss of fluidity as f and E are increased, even at temperatures typically reached during irradiation. This would be expected to exert a strong influence on pattern formation in typical experimental systems.

Hence, to the two questions posed in the Introduction, our findings are as follows. (i) We do not find evidence of new morphological instabilities induced by a nonuniform fluidity within the amorphous layer. However, numerical results suggest that the contribution to the linear growth rate Σ of viscous surface energy relaxation scales differently with wave number k than is suggested by [40], and the effect is modified by nominal beam energy. For example, at 500 eV nominal beam energy and a flux of 2 ions/nm²/s, a single Fourier mode of wave number $k = 0.9$ nm⁻¹ is predicted to relax at a rate roughly 50% greater than if fluidity were constant throughout the film. This could significantly alter theoretical predictions and model validation against experimental observations and represents an avenue for further study. (ii) With plausible parameter values, explicitly modeling the defect kinetics within the amorphous layer leads to the prediction that sufficiently high ion flux f (though within the range of typical experiments [14,17,42,68]) can lead to a loss of fluidity due to thermally enhanced recombination of Frenkel pairs, which

would induce a novel flux dependence not anticipated by other theoretical studies [17,20]. This suggests that temperature control during ion bombardment experiments intended for comparison with theoretical models is an important and overlooked consideration. The functional form of η^{-1} arising from the numerical model is also different than what has been previously considered [20].

While the present results are sensitive to the diffusion rates of vacancies and interstitials and especially to how the diffusion rates respond to temperature changes, these findings are significant for efforts to model ion-induced pattern formation on semiconductor surfaces like silicon and germanium. In particular, we have shown that the defect kinetics within the amorphous thin film cannot in general be ignored for the purposes of modeling pattern formation, especially when temperature is not controlled during irradiation.

B. Comparison with experimental results

The present theoretical work focuses on a smoothening mechanism, that is, viscous relaxation of surface energy, rather than a pattern-forming mechanism. This naturally complicates comparison with experimental observations, where ripple wavelengths at early irradiation times are a typical benchmark. However, the present work does exhibit consistency with experimental observations. First, we note that the $\sqrt{f}$ scaling of η^{-1} with flux f is consistent with [39], where several theoretical models were considered alongside stress measurements from irradiation experiments (Ar⁺-irradiated Si at various energies and fluxes). Second, we have computed growth rates Σ for 500-eV Ar⁺ irradiation of Si at 2 ions/nm²/s that are roughly the same order of magnitude as those obtained in [20] for 1-keV Ar⁺ irradiation of Si at a lower flux (see our Fig. 1). Despite uncertainty in the parameters Q_r , D_{V0} , D_{I0} , ϵ_{em} , and λ (isoconfigurational activation enthalpy, diffusivity of vacancies, diffusivity of interstitials, emissivity, and recombination range, respectively) used to obtain Fig. 1, it lends some confidence to the present modeling

approach that plausible parameters produce plausible agreement with the experiments of [20].

Hence we have presented no comparison with experimental results of ripple patterning because (i) the present work contains no mechanism for surface destabilization and (ii) there exist multiple contenders for a theoretical explanation for surface destabilization within the family of viscous models attempting to describe nanopatterning under irradiation (see, e.g., [14,16,22]). Choosing one particular mechanism for the present work and surveying experimental results would have led to a study of a much different character and focus than the current one. In particular, it would require either an exploration of all candidates for an explanatory model of surface destabilization or some arbitrary choice to be made, the conclusions of which paper would then be easily characterized as incomplete. We have instead chosen to focus on the qualitative theoretical results pertaining directly to surface relaxation.

C. Future work

This work illustrates important connections between defect modeling and the idealization of amorphous thin films as fluids and exposes the potential for sensitivity of irradiation thin film evolution to the details of the underlying defect kinetics, especially because fluidity can evidently decrease with flux and energy if thermally enhanced recombination of defects is strong enough, which would be expected to influence observed ripple patterns. This may be particularly important when experimental systems reach high temperatures, such as in [68]. The present work highlights the importance of estimating Q_r , D_{V0} , and D_{I0} at energies relevant to low-energy irradiation experiments. Because these are only three free parameters, they could in principle be used to fit a large experimental data set across fluxes and energies. These and other implications await further exploration.

In this work we have neglected two important effects, i.e., sputter removal of defects and ion implantation, which would be needed to develop a fully quantitative model capable of predicting stresses and fluidities across fluxes, angles, energies, and temperatures, using only a small set of fit parameters. This is a clear direction for future development. Implications for ion-induced pattern formation in the presence of ion-induced stresses, as modeled by an extension of the present work, should also be studied. The present model appears to be incapable of producing morphological instabilities regardless of parameter choice. However, this work is predicated on a near-complete acceptance of the viscous characterization of the irradiated thin film, except, notably, for the defect kinetics. It is possible that a future model which incorporates other granularlike features of the thin film will indeed predict a new morphological instability. Reconciling the viscouslike and granularlike features of the irradiated thin film system and thereby bridging the erosive-redistributive and viscous-flow classes of models remains an important future direction.

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DATA AVAILABILITY

The data that support the findings of this article are openly available [47]; embargo periods may apply.

APPENDIX: LINEARIZATION DETAILS

Here we record the details of linearization of Eqs. (1)–(11).

1. Steady-state equations

Conservation of mass and momentum become

$$\begin{aligned}\frac{\partial}{\partial z}(\rho_0 w_0) &= 0, \\ \frac{\partial}{\partial z}(\eta_0 u_{0,z}) &= 0, \\ -p_{0,z} + 2\frac{\partial}{\partial z}(\eta_0 w_{0,z}) &= 0.\end{aligned}\quad (\text{A1})$$

The steady-state equations for the defect kinetics are

$$\begin{aligned}fY_V I_0(z) + D_V \frac{\partial^2}{\partial z^2} C_{V,0} + \frac{1}{3} \left(\frac{D_V V_V}{k_B T} \right) C_{V,0} p_{0zz} \\ - 4\pi \lambda C_{I,0} C_{V,0} (D_I + D_V) &= 0, \\ fY_I I_0(z) + D_I \frac{\partial^2}{\partial z^2} C_{I,0} + \frac{1}{3} \left(\frac{D_I V_I}{k_B T} \right) C_{I,0} p_{0zz} \\ - 4\pi \lambda C_{I,0} C_{V,0} (D_I + D_V) &= 0.\end{aligned}\quad (\text{A2})$$

At lower interface $z = 0$,

$$\begin{aligned}u_0 &= 0, \\ w_0 &= V(\theta), \\ \Delta_0 = \frac{\partial \Delta_0}{\partial z} = \frac{\partial^2 \Delta_0}{\partial z^2} &= 0.\end{aligned}\quad (\text{A3})$$

At free interface $z = h_0(\theta)$,

$$\begin{aligned}0 &= w_0 - V(\theta) \frac{\rho^*}{\rho_0}, \\ \eta_0 u_{0,z} &= 0, \\ -p_0 + 2\eta_0 w_{0,z} &= 0.\end{aligned}\quad (\text{A4})$$

2. Note on the steady-state equations

Although the steady-state equations appear to be analytically intractable, two useful insights are readily available. First, observe that $\rho_0 w_0 = c_1$, $\eta_0 u_{0,z} = c_2$, $-p_0 + 2\eta_0 w_{0,z} = c_3$, and the boundary condition at $z = h_0(\theta)$ implies $c_3 = 0$. Also, $w_0 = \frac{c_1}{\rho_0}$, implying that $w_{0z} = -\frac{c_1 \rho_{0z}}{\rho_0^2}$, and the boundary conditions at $z = 0$ imply that $c_1 = V(\theta) \rho^*$. Then the steady pressure $p_0 = -2V(\theta) \rho^* \frac{\eta_0 \rho_{0z}}{\rho_0^2}$, and η_0 and ρ_0 are defined entirely in terms of C_{V0} and C_{I0} . Moreover, the steady-state equations for C_{I0} and C_{V0} are independent of the others. Hence the steady state is entirely governed by the defect fields. This reflects the primacy of the defect kinetics in understanding the thin film dynamics. Second, observe that $u_{0z} = \frac{c_2}{\eta_0}$. Then

$u_0 = \int_0^z \frac{c_2}{\eta_0} dz + c_4$. However, at $z = 0$, $u_0 = 0$, so $c_4 = 0$, and $\eta_0 u_{0,z} = 0$ at $z = h_0(\theta)$ implies that $c_2 = 0$. Hence $u_0 = 0$. Physically, this says that the prescribed defect kinetics cannot

produce a shear flow of the type expected from [16] and related work. This observation also permits simplification of the linearized equations.

3. Simplification of steady-state equations with $\frac{D_I}{D_V} \rightarrow 0$

The steady-state equations, as written, frustrate typical analytical and numerical techniques. In particular, the presence of p_{0zz} in the defect kinetics equations leads to two third-order ordinary differential equations (ODEs) where C_{I0}''' and C_{V0}''' cannot be separated algebraically, as would be required for a numerical solution using bvp5c or other prepackaged numerical solvers. However, in silicon, self-diffusion of interstitials is orders of magnitude slower than that of vacancies, that is, $\frac{D_I}{D_V} \approx 0$ [65,66]. Then we identify a characteristic rate $\frac{D_V h_0}{V_V}$ (with units s^{-1}). Dividing both sides by this, we obtain the dimensionless equations

$$\begin{aligned} \frac{fY_I I_0(z)V_V}{D_V h_0} + \frac{V_V}{h_0} C_{V,0zz} + \frac{1}{3} \frac{V_V}{h_0} \left(\frac{V_V}{k_B T} \right) C_{V,0} p_{0zz} - 4\pi \lambda C_{V0} C_{I0} \frac{V_V}{h_0} \left(1 + \frac{D_I}{D_V} \right) &= 0, \\ \frac{fY_I I_0(z)V_V}{D_V h_0} + \left(\frac{D_I}{D_V} \right) \frac{V_V}{h_0} C_{I,0zz} + \frac{1}{3} \frac{D_I}{D_V} \frac{V_V}{h_0} \left(\frac{V_I}{k_B T} \right) C_{I,0} p_{0zz} - 4\pi \lambda C_{V0} C_{I0} \frac{V_V}{h_0} \left(1 + \frac{D_I}{D_V} \right) &= 0. \end{aligned} \quad (A5)$$

To leading order in $\frac{D_I}{D_V}$, we obtain

$$\frac{fY_I I_0(z)V_V}{D_V h_0} + \frac{V_V}{h_0} C_{V,0zz} + \frac{1}{3} \frac{V_V}{h_0} \left(\frac{V_V}{k_B T} \right) C_{V,0} p_{0zz} - 4\pi \lambda C_{V0} C_{I0} \frac{V_V}{h_0} = 0, \quad (A6a)$$

$$\frac{fY_I I_0(z)V_V}{D_V h_0} - 4\pi \lambda C_{V0} C_{I0} \frac{V_V}{h_0} = 0, \quad (A6b)$$

with the latter leading immediately to

$$C_{I0} = \frac{fY_I I_0(z)}{4\pi \lambda C_{V0} D_V} \quad (A7)$$

to leading order in the diffusivity ratio. This approximation greatly simplifies the problem at hand. With these simplifications, we express p_{0zz} in terms of C_{V0} , and Eq. (A6a) yields a nonlinear third-order ODE, and the three boundary conditions for Δ_0 at $z = 0$, when expressed in terms of C_{V0} , lead to a solvable problem whose solution fully determines the other steady-state quantities u_0 , w_0 , ρ_0 , η_0 , p_0 , and C_{V0} .

4. Steady-state equation for C_{V0}

Here we record the third-order nonlinear ordinary differential equation providing the steady state of C_{V0} , which was suppressed in the main text for brevity,

$$C_{V0zzz} = - \frac{3k_B T (D_V C_{V0zz} + \frac{D_V V_V C_{V0} \Phi}{3k_B T}) (\frac{1}{\eta^*} + \frac{C_{V0}}{\eta_V} + \frac{Y_I f I_0}{4D_V \lambda \eta_I \pi C_{V0}})}{2D_V V_V V(\theta) C_{V0} (\frac{V_V}{V_0} - \frac{V_I Y_I f I_0}{4D_V \lambda V_0 \pi C_{V0}^2})}, \quad (A8)$$

where Φ is

$$\begin{aligned} & 2V(\theta) \left(\frac{V_I Y_I f I_{0zzz}}{4D_V \lambda V_0 \pi C_{V0}} - \frac{3V_I Y_I f C_{V0z} I_{0zz}}{4D_V \lambda V_0 \pi C_{V0}^2} - \frac{3V_I Y_I f I_{0z} C_{V0zz}}{4D_V \lambda V_0 \pi C_{V0}^2} - \frac{3V_I Y_I f I_0 C_{V0z}^3}{2D_V \lambda V_0 \pi C_{V0}^3} + \frac{3V_I Y_I f C_{V0z}^2 I_{0z}}{2D_V \lambda V_0 \pi C_{V0}^3} + \frac{3V_I Y_I f I_0 C_{V0z} C_{V0zz}}{2D_V \lambda V_0 \pi C_{V0}^3} \right) \\ & \quad - \frac{\frac{1}{\eta^*} + \frac{C_{V0}}{\eta_V} + \frac{Y_I f I_0}{4D_V \lambda \eta_I \pi C_{V0}}}{4V(\theta) \left(\frac{C_{V0z}}{\eta_V} + \frac{Y_I f I_{0z}}{4D_V \lambda \eta_I \pi C_{V0}} - \frac{Y_I f I_0 C_{V0z}}{4D_V \lambda \eta_I \pi C_{V0}^2} \right) \left(\frac{V_V C_{V0zz}}{V_0} + \frac{V_I Y_I f I_{0zz}}{4D_V \lambda V_0 \pi C_{V0}} + \frac{V_I Y_I f I_0 C_{V0z}^2}{2D_V \lambda V_0 \pi C_{V0}^3} - \frac{V_I Y_I f I_0 C_{V0zz}}{4D_V \lambda V_0 \pi C_{V0}^3} - \frac{V_I Y_I f C_{V0z} I_{0z}}{2D_V \lambda V_0 \pi C_{V0}^3} \right)} \\ & \quad + \frac{4V(\theta) \left(\frac{V_V C_{V0z}}{V_0} + \frac{V_I Y_I f I_{0z}}{4D_V \lambda V_0 \pi C_{V0}} - \frac{V_I Y_I f I_0 C_{V0z}}{4D_V \lambda V_0 \pi C_{V0}^2} \right) \left(\frac{C_{V0z}}{\eta_V} + \frac{Y_I f I_{0z}}{4D_V \lambda \eta_I \pi C_{V0}} - \frac{Y_I f I_0 C_{V0z}}{4D_V \lambda \eta_I \pi C_{V0}^2} \right)^2}{\left(\frac{1}{\eta^*} + \frac{C_{V0}}{\eta_V} + \frac{Y_I f I_0}{4D_V \lambda \eta_I \pi C_{V0}} \right)^3} \\ & \quad - \frac{2V(\theta) \left(\frac{V_V C_{V0z}}{V_0} + \frac{V_I Y_I f I_{0z}}{4D_V \lambda V_0 \pi C_{V0}} - \frac{V_I Y_I f I_0 C_{V0z}}{4D_V \lambda V_0 \pi C_{V0}^2} \right) \left(\frac{C_{V0zz}}{\eta_V} + \frac{Y_I f I_{0zz}}{4D_V \lambda \eta_I \pi C_{V0}} - \frac{Y_I f C_{V0z} I_{0z}}{2D_V \lambda \eta_I \pi C_{V0}^2} + \frac{Y_I f I_0 C_{V0z}^2}{2D_V \lambda \eta_I \pi C_{V0}^3} - \frac{Y_I f I_0 C_{V0zz}}{4D_V \lambda \eta_I \pi C_{V0}^3} \right)}{\left(\frac{1}{\eta^*} + \frac{C_{V0}}{\eta_V} + \frac{Y_I f I_0}{4D_V \lambda \eta_I \pi C_{V0}} \right)^2}. \end{aligned}$$

Hence $C_{V0}(z)$ is fully determined by system parameters and the derivatives of $I_0(z)$.

5. Linearized equations with $\frac{D_V}{D_I} \rightarrow 0$

In the bulk, conservation of mass and momentum are

$$\begin{aligned}\rho_{1t} + \rho_0 u_{1x} + \frac{\partial}{\partial z}(\rho_0 w_1) + \frac{\partial}{\partial z}(\rho_1 w_0) &= 0, \\ -p_{1x} + 2\eta_0 u_{1xx} + \frac{\partial}{\partial z}[\eta_0(u_{1z} + w_{1x})] &= 0, \\ -p_{1z} + \eta_0(w_{1xx} + u_{1xz}) + 2\frac{\partial}{\partial z}(\eta_0 w_{1z}) + 2\frac{\partial}{\partial z}(\eta_1 w_{0z}) &= 0\end{aligned}\quad (\text{A9})$$

and the reaction-diffusion equations for the defects are

$$\begin{aligned}C_{V,1t} &= fY_V I_1(x, z) + D_V \nabla^2 C_{V,1} - \frac{1}{3} \left(\frac{D_V V_V}{k_B T} [C_{V,1} \nabla^2 (-2p_0 + 2\eta_0 w_{0z}) \right. \\ &\quad \left. + C_{V,0} \nabla^2 (-2p_1 + 2\eta_0(u_{1x} + w_{1z}) + 2\eta_1(w_{0z})) \right] - 4\pi\lambda D_V (C_{V0} C_{I1} + C_{V1} C_{I0}), \\ C_{I,1t} &= fY_I I_1(x, z) - 4\pi\lambda D_V (C_{V0} C_{I1} + C_{V1} C_{I0}).\end{aligned}\quad (\text{A10})$$

At lower interface $z = 0$,

$$\begin{aligned}u_1 &= 0, \\ w_{0z} g_1 + w_1 &= 0, \\ \Delta_{0z} g_1 + \Delta_1 &= 0, \\ \Delta_{0zz} g_1 + \Delta_{1z} &= 0, \\ \Delta_{0zzz} g_1 + \Delta_{1zz} &= 0.\end{aligned}\quad (\text{A11})$$

At free interface $z = h_0$,

$$\begin{aligned}h_{1t} &= w_1 + h_1 w_{0z} + V(\theta) \frac{\rho^*}{\rho_0^2} (\rho_{0z} h_1 + \rho_1), \\ h_{1x} p_0 + \eta_0(u_{1z} + w_{1x}) &= 0, \\ h_1 \frac{\partial}{\partial z} (-p_0 + 2\eta_0 w_{0z}) + -p_1 + 2(\eta_0 w_{1z} + \eta_1 w_{0z}) &= -\gamma h_{1xx}.\end{aligned}\quad (\text{A12})$$

Throughout the above,

$$\begin{aligned}\rho_0 &= \frac{m_0}{V_0 + C_{I,0} V_I + C_{V,0} V_V}, \quad \rho_1 = -\frac{m_0(C_{I,1} V_I + C_{V,1} V_V)}{(V_0 + C_{I,0} V_I + C_{V,0} V_V)^2}, \\ \eta_0 &= \frac{1}{\frac{1}{\eta^*} + \frac{C_{I,0}}{\eta_I} + \frac{C_{V,0}}{\eta_V}}, \quad \eta_1 = -\frac{\frac{C_{I,1}}{\eta_I} + \frac{C_{V,1}}{\eta_V}}{\left(\frac{1}{\eta^*} + \frac{C_{I,0}}{\eta_I} + \frac{C_{V,0}}{\eta_V}\right)^2}.\end{aligned}\quad (\text{A13})$$

6. Linearized equations for normal modes with $\frac{D_V}{D_I} \rightarrow 0$

Following the application of the linear stability ansatz to Eqs. (1)–(11), we obtain the following set of boundary value problems for eigenvalue Σ , which will determine the stability of Fourier modes of wave number k . In the bulk, we obtain the following ODEs for mass conservation and momentum conservation:

$$\Sigma \tilde{\rho}_1 + ik\rho_0 \tilde{u}_1 + \rho_{0z} \tilde{w}_1 + \tilde{w}_{1z} \rho_0 + \tilde{\rho}_{1z} w_0 + w_{0z} \tilde{\rho}_1 = 0, \quad (\text{A14a})$$

$$-ik\tilde{p}_1 - 2k^2\eta_0 \tilde{u}_1 + \eta_{0z}(\tilde{u}_{1z} + ik\tilde{w}_1) + \eta_0(\tilde{u}_{1zz} + ik\tilde{w}_{1z}) = 0, \quad (\text{A14b})$$

$$-\tilde{p}_{1z} + \eta_0(-k^2 \tilde{w}_1 + ik\tilde{u}_{1z}) + 2(\eta_{0z} \tilde{w}_{1z} + \eta_0 \tilde{w}_{1zz}) + 2(\tilde{\eta}_{1z} w_{0z} + \tilde{\eta}_1 w_{0zz}) = 0. \quad (\text{A14c})$$

We also obtain the following ODEs for the reaction, advection, and diffusion of defects within the amorphous bulk:

$$\begin{aligned}\Sigma \tilde{C}_{V,1} &= fY_V \tilde{P}_1 + D_V (-k^2 \tilde{C}_{V,1} + \tilde{C}_{V,1zz}) - \frac{1}{3} \frac{D_V V_V}{k_B T} (-\tilde{C}_{V,1} p_{0zz} + C_{V,0} \{-2(-k^2 \tilde{p}_1 + \tilde{p}_{1zz}) \\ &\quad + 2[(ik\tilde{u}_1 + \tilde{w}_{1z})\eta_{0zz} + \eta_0(-ik^3 \tilde{u}_1 + ik\tilde{u}_{1zz} - k^2 \tilde{w}_{1z} + \tilde{w}_{1zzz}) + 2\eta_{0z}(ik\tilde{u}_{1z} + \tilde{w}_{1zz})] \\ &\quad + 2[w_{0z}(-k^2 \tilde{\eta}_1 + \tilde{\eta}_{1zz}) + \tilde{\eta}_1 w_{0zz} + \tilde{\eta}_{1z} w_{0zz}]\}) - 4\pi\lambda D_V (C_{V,0} \tilde{C}_{I,1} + \tilde{C}_{V,1} C_{I,0})\end{aligned}\quad (\text{A15})$$

for the interstitials and

$$\Sigma \tilde{C}_{I,1} = fY_I \tilde{P}_1 - 4\pi\lambda D_V (C_{V,0} \tilde{C}_{I,1} + \tilde{C}_{V,1} C_{I,0}) \quad (\text{A16})$$

for the vacancies. These equations for the interior are accompanied by the following boundary conditions. At $z = 0$,

$$\begin{aligned} \tilde{u}_1 &= 0, \\ w_{0z} \tilde{g}_1 + \tilde{w}_1 &= 0, \\ \Delta_{0z} \tilde{g}_1 + \tilde{\Delta}_1 &= 0, \\ \Delta_{0zz} \tilde{g}_1 + \tilde{\Delta}_{1z} &= 0, \\ \Delta_{0zzz} \tilde{g}_1 + \tilde{\Delta}_{1zz} &= 0. \end{aligned} \quad (\text{A17})$$

To facilitate numerical solution, we note that the latter three boundary conditions above can also be expressed as

$$\begin{aligned} \tilde{C}_{V1} &= -\left(C_{V0z} + C_{I0z} \frac{V_I}{V_V}\right) \tilde{g}_1 - \tilde{C}_{I1} \frac{V_I}{V_V}, \\ \tilde{C}_{V1z} &= -\left(C_{V0zz} + C_{I0zz} \frac{V_I}{V_V}\right) \tilde{g}_1 - \tilde{C}_{I1z} \frac{V_I}{V_V}, \\ \tilde{C}_{V1zz} &= -\left(C_{V0zzz} + C_{I0zzz} \frac{V_I}{V_V}\right) \tilde{g}_1 - \tilde{C}_{I1zz} \frac{V_I}{V_V}. \end{aligned} \quad (\text{A18})$$

At $z = h_0$,

$$\begin{aligned} \Sigma \tilde{h}_1 &= \tilde{w}_1 + \tilde{h}_1 w_{0z} + V(\theta) \frac{\rho^*}{\rho_0^2} (\rho_{0z} \tilde{h}_1 + \tilde{\rho}_1), \\ ik \tilde{h}_1 p_0 + \eta_0 (\tilde{u}_{1z} + ik \tilde{w}_1) &= 0, \\ \tilde{h}_1 [-p_{0z} + 2(\eta_{0z} w_{0z} + \eta_0 w_{0zz})] - \tilde{p}_1 + 2(\eta_0 \tilde{w}_{1z} + \tilde{\eta}_1 w_{0z}) &= \gamma k^2 \tilde{h}_1. \end{aligned} \quad (\text{A19})$$

Throughout the above,

$$\tilde{\rho}_1 = -\frac{m_0 (\tilde{C}_{I,1} V_I + C_{V,1} V_V)}{(V_0 + C_{I,0} V_I + C_{V,0} V_V)^2}, \quad \tilde{\eta}_1 = -\frac{\frac{\tilde{C}_{I,1}}{\eta_I} + \frac{\tilde{C}_{V,1}}{\eta_V}}{\left(\frac{1}{\eta^*} + \frac{C_{I,0}}{\eta_I} + \frac{C_{V,0}}{\eta_V}\right)^2}, \quad (\text{A20})$$

where we again note that C_{V0} and C_{V1} are fully determined by C_{I0} and C_{I1} according to Eqs. (A7) and (A16), respectively. We also apply the phase shift that relates the interfaces,

$$\tilde{g}_1 = \tilde{h}_1 \exp[-ikx_0(\theta)], \quad (\text{A21})$$

where $x_0(\theta)$ is as described in Eq. (12). This assignment is described in more detail in [25].

7. Note on solution technique

Solving a nonlinear eigenvalue problem with Matlab's `bvp5c` requires that there be one more boundary condition than the number of ODEs when all of the bulk equations are expressed as a system of first-order ODEs. The presence of eight boundary conditions then requires the expression of the equations for the bulk as a system of seven first-order ODEs.

To obtain these, we use Eq. (A14a) to express $\tilde{w}_{1z}$ in terms of $\tilde{\rho}_{1z}$ and $\tilde{u}_1$ ($\tilde{\rho}_1$ can be expressed in terms of $\tilde{C}_I$ and $\tilde{C}_V$); use Eq. (A14b) as an equation for $\tilde{u}_{1zz}$; use Eq. (A14c) as an equation for $\tilde{p}_{1z}$, where the $\tilde{w}_{1zz}$ is expressible in terms of $\tilde{C}_V$, $\tilde{C}_{Vz}$, $\tilde{C}_{Vzz}$, $\tilde{u}_1$, and $\tilde{u}_{1z}$ using Eq. (A14a), as described above; and use Eq. (A15) as a third-order ODE for $\tilde{C}_V$, where $\tilde{p}_{1zz}$, $\tilde{w}_{1zzz}$, $\tilde{u}_{1z}$, $\tilde{u}_{1zz}$, $\tilde{\eta}_{1z}$, and $\tilde{\eta}_{1zz}$ can be expressed in terms of lower-order derivatives using the other substitutions described above.

In the resulting system of ODEs, each unknown function's highest derivative occurs exactly once, and the resulting equations are expressible as a system of first-order ODEs, as `bvp5c` requires. The manipulations described above were performed using Matlab's Symbolic Math Toolbox. We do not record the resulting equations which were used with `bvp5c` due to their extreme length.

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