Viscous Thin-film Models of Nanoscale Self-organization Under Ion Bombardment

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VISCOUS THIN-FILM MODELS OF NANOSCALE SELF-ORGANIZATION UNDER ION BOMBARDMENT

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VISCOUS THIN-FILM MODELS OF NANOSCALE SELF-ORGANIZATION
UNDER ION BOMBARDMENT

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For decades, it has been observed that broad-beam irradiation of semiconductor surfaces can lead to spontaneous self-organization into highly regular patterns, sometimes at length scales of only a few nanometers. Initial theory was largely based on erosion and redistribution of material occurring on fast time scales, which are able to produce good agreement with certain aspects of surface evolution. However, further experimental and theoretical work eventually led to the realization that numerous effects are active in the irradiated target, including stresses associated with ion-implantation and the accumulation of damage leading to the development of a disordered, amorphous layer atop the substrate. It was also shown that relaxation of this amorphous layer proceeds in a manner closer to viscous flow rather than surface diffusion on a crystal lattice.

Observing the viscous character of the amorphous layer, it is natural to consider whether stress-based continuum models might help explain pattern formation under ion bombardment and the observations described above. Indeed, there are early indications from the experimental literature that this may be the case, and, at low energies (∼1keV), at least one experimental-theoretical study has shown that they may even dominate erosive and redistributive effects in their contribution to surface evolution.
In this thesis, we develop a continuum model based on viscous thin-film flow and ion-induced stresses within the amorphous layer. This model is a composite of, and significant generalization of, a previously-studied “anisotropic plastic flow” (APF) mechanism and a previously-studied “ion-induced isotropic swelling” (IIS) mechanism. Previous work has shown that, with certain simplifying assumptions about the amorphous-crystalline interface and spatial homogeneity of anisotropic plastic flow, this mechanism produces an instability capable of predicting pattern formation beginning at 45° angle of incidence against the macroscopically-flat substrate, consistent with some experimental systems. Under similar simplifying assumptions, ion-induced swelling has been shown to be capable of suppressing pattern formation. Our generalizations allow the use of simulation data to inform both linear and nonlinear surface evolution due to the spatial localization of APF and IIS to certain regions of the bulk, improved treatment of the amorphous-crystalline geometry, and boundary conditions suitable to the physical systems of interest. We are then able to provide insight into several phenomena that have previously been difficult to explain, but seem to emerge naturally from a more detailed treatment of the physical system.
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5.3. Fitting the model growth rate (Equation (5.37)) to $\theta_c$ across 250eV-2keV $\text{Ar}^+$, $\text{Kr}^+$, $\text{Xe}^+ \rightarrow \text{Si}$, and $\text{Kr}^+$, $\text{Xe}^+ \rightarrow \text{Ge}$ using two free parameters, $\hat{K}_I$ and $\hat{K}_D$, per target. For Si, we obtain $l^2 \approx 7.6, \hat{K}_D = 4.442 \times 10^{-5}, \hat{K}_I = 1.046 \times 10^{-4}$. For Ge, we obtain $l^2 \approx 4.3, \hat{K}_D = 3.7585 \times 10^{-4}, \hat{K}_I = 8.028 \times 10^{-4}$. **For Si targets** we have: 1keV $\text{Ar}^+$ [58, 33], 500eV $\text{Ar}^+$ [33], 250eV $\text{Ar}^+$ [43], 2keV $\text{Kr}^+$ [83], 1.2keV $\text{Kr}^+$ [3], 1keV $\text{Kr}^+$ [47], 1keV $\text{Xe}^+$ [33], 500eV $\text{Xe}^+$ [33]. **For Ge targets** we have: 1.2keV $\text{Kr}^+$ [57], 1keV $\text{Kr}^+$ [84], 500eV $\text{Kr}^+$ [55], 250eV $\text{Kr}^+$ [55], 2keV $\text{Xe}^+$ [57], 1.2keV $\text{Xe}^+$ [57], 600eV $\text{Xe}^+$ [57]. Dots denote experimental data, and dashed lines are model predictions. ..............................

5.4. Here, we compare predictions of $\lambda(\theta)$ from two experimental studies to those obtained from our theoretical model using the values of $fA_I$ and $fA_D$ inferred from fitting $\theta_c$ across our 15 systems of interest simultaneously. **Left:** comparison with data from [68]. **Right:** comparison with data from [33]. ..............................

5.5. Here, we consider $\theta_c$ vs beam energy for several different values of $\frac{\rho_a}{\rho_c}$ where we consider $\hat{K}_D$ taken as the fitted value. For these parameter values, the region around 1.2keV beam energy coincides with a very sharp increase in $\theta_c$ as experimentally observed. These values of $\hat{K}_I$ and $\hat{K}_D$ coincide with the region in which the effect of the boundary amorphization term can dominate stress effects. Also note that these values of $\frac{\rho_a}{\rho_c}$ are well-within experimentally-determined values, and that the lowest value of $\theta_c$ is around 30°. For each of the values of $\frac{\rho_a}{\rho_c}$ considered, it is possible to obtain similar “jumps” in $\theta_c$ for a corrected value of $\hat{K}_D$. ..............................

5.6. Here, we consider $\text{Ne}^+$, $\text{Ar}^+ \rightarrow \text{Ge}$ for 250eV-2keV; these systems are not observed to form patterns, at least up to about 75° [57]. Our model incorrectly predicts that these systems are pattern-forming. We discuss this inconsistency below. ..............................
5.7. Here, we have attempted to fit the model with the same scaling argument but without spatially-resolved IIS— that is, using the model of Chapter 4. We fit the model (Equation (5.37)) to $\theta_c$ across 250eV-2keV $\text{Ar}^+$, $\text{Kr}^+$, $\text{Xe}^+ \rightarrow \text{Si}$, and $\text{Kr}^+$, $\text{Xe}^+ \rightarrow \text{Ge}$ using two free parameters, $\hat{K}_I$ and $\hat{K}_D$, per target. For Si, we obtain $l^2 \approx 10.09, \hat{K}_D = 5.474 \times 10^{-6}, \hat{K}_I = 3.0769 \times 10^{-6}$. For Ge, we obtain $l^2 \approx 12.83, \hat{K}_D = 7.659 \times 10^{-6}, \hat{K}_I = 4.351 \times 10^{-6}$. Dots denote experimental data, and dashed lines are model predictions.

6.1. Simulation of nonlinear evolution due to Equations (6.75)-(6.77) in the fictitious case that $q_0 = 1, h_0 = 3\text{nm}, x_0 = 1\text{nm}, fA = .1^1_s, \gamma = 1.36\frac{1}{\text{m}^2}, \eta = .6 \text{GPa-s}$. Horizontal and vertical length scales above are in nm. Initial conditions are Gaussian white noise with mean 0, standard deviation $\frac{1}{10}$. We have taken discretization $\Delta x = \frac{1}{2}, \Delta t = \frac{1}{100}$.

A.1. The A-dependent, angle-independent coefficient from (a) the real part and (b) the imaginary part of the long-wave dispersion relation, from equations (A.44) and (A.45).

B.1. Schematic depicting the construction of depth-dependence at a point $S$ in the film depth in Cartesian $x, z$ coordinates by mapping the influence of an ion that enters the surface at the point $Q$ to the point $S$. In the limit of small slopes, this geometric argument leads to analytically-tractable forms.
Dedicated to my family, the search for understanding, and human progress.
Chapter 1

Introduction

It has been known since at least the 1960s that some materials may be irradiated by an ion beam in an energy range of 100eV to 10keV in order to produce nanoscale patterns of various kinds [1]. Examples include ripples, hexagonal arrays of dots, and continuous transitions between the two [2, 3] where both may appear at once. These structures can range in scale from approximately 5-20nm [2]. Because ion beams are already ubiquitous in industrial settings, these early findings inspired great interest in developing a comprehensive theory of nanostructuring. It was thought that the tendency for surfaces to spontaneously self-organize into coherent patterns could be controlled and applied in a mass-manufacturing context, facilitating the production of cheap and high-quality nano- and meta- materials. Rather than precisely engineering a structure part-by-part, these materials could be “grown” through a “bottom-up” approach, requiring only coarse control of the system at scales many orders of magnitude greater than that of the resulting nanostructures. However, despite decades of effort, such a comprehensive theory has remained elusive [4, 5, 6], and the dream of “bottom-up” nano-engineering has gone (lamentably) unrealized at the time of writing.

The lack of a universal theory aside, some things have come to be understood, and it has proven useful to consider mechanisms acting on two time scales: the “prompt regime” associated with atomic displacements caused by individual ion impacts (on the order of $\sim 10^{-9}$ seconds), and the “gradual regime” associated with the buildup and relaxation of stress (on the order of $\sim 10^2$ seconds or longer). In the “prompt regime”, some target atoms may be permanently sputtered away from the target during each impact, and the resulting surface erosion rates may vary across a given film topography, resulting in the well-known Bradley-Harper instability [7, 8, 9, 10]. Conversely, other target atoms are redistributed
to new locations, which can either stabilize or destabilize the surface depending on the ion incidence angle [11, 12, 13, 14]. A recent, integrated approach to handling both erosion and redistribution simultaneously is the crater function framework [15, 16, 17, 18, 19], a long-wave approximation of the surface that utilizes simulation data to collect information about the “craters” caused by individual ion impacts, which may then be used to study aggregate behavior. Several variations of this framework are currently in use [17, 20, 21, 22, 23], including specialized Python packages [24]. However, there still exist technical problems with this approach, such as the apparent disagreement between crater function-based predictions [18, 24] and experimental observations. While other approaches claim superior agreement [20, 21, 22, 23], some of the modifications that result in this apparent agreement have been disputed [6]. We will elaborate on this point somewhat in the Discussion of Chapter 4.

For most of the 1960s through the early 2000s, the dominant hypothesis was that pattern formation was due primarily to erosion [7, 8, 9, 25, 6], with surface diffusion acting as the regularization mechanism prohibiting the selection of arbitrarily-small wavelengths (which would violate the continuum hypothesis) [4, 6]. However, Umbach [26] showed that regularization by viscous surface leveling [27] was more consistent with experimental observations, at least near room temperature and for amorphizable materials. This finding was later reinforced by [28]. Other regularization mechanisms were proposed in the intervening time, including athermal, effective surface diffusion [29], which was later rejected [25, 6].

The “dominant erosion” hypothesis was drawn into question by [14, 18], which presented evidence that mass redistribution of non-sputtered matter could dominate the instability due to erosion. This led to a re-imagining of the theory and proposed modifications [20, 21, 22, 23] to the crater function framework [17, 6]. Later, the importance of erosion at low energies was questioned altogether [30, 31, 32, 33] as attention turned to the effects of ion-induced stresses on pattern formation, which evolve slowly.

Within this “gradual regime,” it has been established that some materials, upon sustained exposure to ion bombardment, may develop a thin, amorphous layer near the surface, which can be modeled as a highly viscous fluid [26]. That the gradual regime is slow should
not be taken to imply that it is less important in determining surface evolution than the prompt regime phenomena. Indeed, within the gradual regime, such mechanisms as stress buildup and relaxation, surface diffusion, and viscous flow may occur [34, 35, 36, 37, 38, 39, 40, 41, 42, 43, 44, 45, 46, 47, 48]. Many other theoretical mechanisms have been suggested to act in this regime to influence pattern formation, such as effective body forces [31, 49], anisotropic plastic flow [32], and isotropic swelling [48, 50]. Because these mechanisms are slower, smaller in length-scale and active within the amorphous bulk, they may be more difficult to experimentally observe when compared with prompt-regime surface phenomena. Nonetheless, they are increasingly viewed as important to fully understand, as agreement between theory and experiment appears to require the superset of various mechanisms [28].

Cataloging all such mechanisms remains a rich source of theoretical-experimental collaboration. As outlined in [51], it is of great theoretical interest to see how much can be explained on the basis of stress, particularly since patterns are observed to form even in energetic regimes where prompt-regime phenomena may be negligible [52]. This may have practical implications for engineering problems, such as in the design of nuclear reactor walls; see the brief discussion in [18]. As an aside, we note that the work of [53] proposed that the amorphous layer could act as the “stage” for stress-induced pattern formation nearly a decade before other groups, and suggested a continuum treatment of the amorphous bulk before [26]; unfortunately, this work, interesting in its own right and of importance to the history of this field, appears to have been largely overlooked and is only infrequently cited.

Despite advances in modeling irradiated surfaces, no unifying theory explaining all observed parametric dependencies exists. This is not surprising, given the extreme number of mechanisms involved; see [4, 5, 6, 51] for an extensive discussion. However, linear stability analysis in Fourier modes has proven to be a very useful tool in bridging theory and experiment, and a rich literature surrounding this approach to pattern formation already exists [54, 5, 6, 51]. A typical experiment produces observations on the presence or absence of patterns, as well as the wavelength of patterns, across a variety of experimental parameters such as the target species, ion species, ion energy, and ion incidence angle [43, 14, 55]. Sim-
ilarly, a linear stability analysis in normal modes predicts whether any wavenumbers have positive growth rate (leading to the presence of patterns), and which wavenumbers grow the fastest (leading to wavelength selection). More sophisticated experimental study using, e.g., Grazing Incidence Small-Angle X-ray Scattering (GISAXS) can additionally provide the real-time growth rate of surface undulations as a function of wavenumber, and predictions on these quantities, too, are provided by linear stability analysis [43, 14, 45, 28, 47].

Unfortunately, even predicting the presence or absence of patterns across parameter values has proven challenging. For instance, although very many ion, target, and energy combinations have been observed to transition from flat to patterned surfaces when the ion incidence exceeds a critical angle $\theta_c$, the value of this angle varies from system to system, with observations ranging from as low as $30^\circ$ [56] to as high as about $70^\circ$ [57]. In some systems, no patterns are observed for any tested incidence angle [57, 22], although grazing incidence is frequently excluded from study. No unified theoretical treatment has been able to satisfactorily explain these varying outcomes, with proposed explanations coming from both the erosive-distributive [22] and stress-based [33] families of models.

Experimental results for 250eV-1keV Ar$^+$ on Si yield $\theta_c \approx 45^\circ$ [43], whereas heavier ions such as Xe$^+$ and Kr$^+$ on Si at similar energies result in $\theta_c \approx 60^\circ$ [47]. Other values have been obtained experimentally for different ion, energy and target combinations; Reference [33] contains a convenient list. In contrast, most of the prevailing theoretical analyses of the hydrodynamic type have all predicted $\theta_c = 45^\circ$ as an apparently-universal feature [31, 58, 32], with the exception of [33], which we address in the Discussion of Chapter 3. Clearly, such a discrepancy between theory and experiment requires an amended theory.

In the direction of explaining the nonuniversality of $\theta_c$, there has been recent consideration of some avenues for the introduction of material-specificity within the hydrodynamic family of models: in particular, depth-dependence in combination with phenomenological modeling of ion-induced stress is purported to lead to good fits with MD simulation data, both in the linear and nonlinear regimes [33, 49]. On the other hand, because there are so many competing mechanisms at work within nanostructure-forming systems, development
of theory is highly nuanced and must be approached cautiously. Indeed, while a proposed mechanism, or its mathematical form as appears in, e.g., an evolution equation, may apparently lead to good predictions within one regime, it is possible that this is not due to having modeled the correct physics, but, rather, having found a mechanism that has similar mathematical properties as the correct physics within the studied regime, but not others. In fact, this has already happened in determining the physical regularization mechanism for ion-sputtered surfaces [6, 26, 28], as both viscous relaxation and surface diffusion scale similarly for long wave perturbations; a distinction between the two only becomes apparent when considering higher wavenumbers, as in [28]. (We also note that there was a brief time that an “athermal effective surface diffusion” (or “effective smoothing”) was proposed as a regularization mechanism, see Reference [10]; this hypothesis was later refuted in [25], and we refer the reader to the discussion there.)

In this thesis, we develop a continuum model based on viscous thin-film flow and ion-induced stresses within the amorphous layer; we will neglect erosive and redistributive effects (1) for simplicity, as it is still unclear to what extent these effects contribute to surface stability— see the discussion in [28, 6, 51]; (2) because it is of interest to see to what extent stress alone might govern the system, without erosion and redistribution as confounding factors [28, 51]; and (3) because the present work is to be considered an exercise in plausibility, rather than a comprehensive theoretical framework— although, as we will see in Chapter 5, the present work may be able to explain quite a lot.

This model is a composite of, and significant generalization of, an “anisotropic plastic flow” (APF) mechanism and an “ion-induced isotropic swelling” (IIS) mechanism, each of which had been studied previously but in isolation and in simplified cases: (1) simplified assumptions about the geometry of the amorphous-crystalline interface, (2) simplified assumptions about the bulk distribution of the proposed phenomena, and (3) boundary conditions suitable for a more typical fluid mechanics problem, rather than one which exhibits a crystalline-to-amorphous phase-change. With these simplifications, previous work has shown that anisotropic plastic flow leads to an instability capable of predicting pattern
formation beginning at 45° angle of incidence, consistent with some experimental systems, but disagreeing with others (as discussed above). However, this does not lead us to abandon the anisotropic plastic flow model in favor of its “competitors” ([31, 33, 49]) in the “effective body force” family. Elsewhere, anisotropic plastic flow has suggested (1) that beyond about 45°, in-plane stresses should exhibit a compressive-to-tensile transition, as is observed experimentally [47]; (2) possible stress-based explanations for nanopore closure [59]; and (3) explanations for the deformation of colloidal silica ellipsoids [37, 38, 39]. Furthermore, the anisotropic plastic flow model, known elsewhere (especially at higher energies) as “ion-beam hammering” [60, 61] has been used widely, with good explanations based on a microscopic “melt-cycle” in the MeV range [60, 61, 62, 6], although these proposed “melt-cycles” do not occur at the low energies of interest in the present work.

Under similar simplifying assumptions as described above, ion-induced swelling (volumization, or loss of density) has been shown to be capable of suppressing pattern formation due to modification of the flow field within the amorphous bulk [48, 50]. While the existence of radiation-induced swelling has been known for decades [63, 64, 65, 66, 67, 62], it had not been previously considered in the context of thin-film linear stability. This additional stabilization effect is apparently capable of modifying the critical angle for the onset of pattern formation [50], potentially offering an explanation for its non-universality.

Our generalizations of these two models will allow the use of simulation data to inform both linear and nonlinear surface evolution due to the spatial localization of APF and IIS to certain regions of the bulk, improved treatment of the amorphous-crystalline geometry, and boundary conditions suitable to the physical systems of interest. We are then able to provide insight into several phenomena that have previously been difficult to explain—in particular, the critical angle, wavelength selection, the sudden, strong suppression of patterns around 1.3keV Ar⁺ → Si. We are also able to suggest a physical basis for the ion-induced swelling mechanism that had been treated on a phenomenological, or mechanism-agnostic, basis previously.
We make four main contributions to the literature (and a few smaller ones throughout). First, through a much-generalized analysis, we show the previously-unappreciated extreme importance of the relationship between the free interface and the amorphous-crystalline interface, which are connected through the collision cascade, and their effect on critical angle and wavelength selection. This, alone, is capable of inducing material and energy specificity, providing an avenue for the observed nonuniversality of the critical angle and wavelength. The generality of this analysis will also provide a staging point for the work that follows, as it introduces the possibility of conducting the analysis with arbitrarily-varying stresses and interfaces first, and only later specifying them.

Second, we offer a treatment of the amorphous-crystalline interface that directly includes simulation data, and we apply a phase-change boundary condition to consider the effect of the crystalline-to-amorphous transition at the translating lower boundary. We show that this improved treatment of the amorphous-crystalline interface is capable of explaining the strong suppression of patterns for \( \text{Ar}^+ \rightarrow \text{Si} \) in the 1keV-40keV range, and quantitatively predicts strong stabilization near 1keV using physically-realistic parameters.

Third, we abandon the assumption of spatially-homogeneous isotropic swelling. Instead, we hypothesize that the effect may be connected directly and quantitatively to distributions of ions, energy, or other quantities induced by the collision cascade. We provide general expressions for the linear growth rate in terms of arbitrary, spatially-localized isotropic swelling and consider a plausible scaling argument for each mechanism. With the hypothesis that isotropic swelling is connected to the ion distribution, we are then able to perform a nonlinear fit of the model to critical angles for all pattern-forming systems from 250eV-2keV for \( \text{Ar}^+ , \text{Kr}^+ , \text{Xe}^+ \rightarrow \text{Si} , \text{Ge} \) with two free parameters per target; we account for 8 experimental systems for Si across 250eV-2keV and three ions, and 7 experimental systems for Ge across 250eV-2keV and two ions. We find that the resulting parameters that fit the critical angles also lead to strong agreement with wavelengths, despite the critical angle and wavelength being independent quantities. This suggests the validity of the model and reasonable parameter estimates. We are also able to show that spatially-localizing anisotropic plastic flow
to some component of the collision cascade does not lead to improved agreement— i.e., that anisotropic plastic flow is best-treated as a constant throughout the film. This suggests that the underlying cause of anisotropic plastic flow within the nuclear stopping regime is somehow nonlocal. This points to further directions in parameter estimation and mechanism identification, which we discuss.

Fourth, we derive the weakly-nonlinear evolution equations for the composite model, perform simple simulations for simplified cases, and consider the implications of the amorphous thin-film stresses for a multiscale problem. These nonlinear equations are developed with the spatial-localization of anisotropic plastic flow and isotropic swelling in mind, as well as the interfacial description that we have shown leads to good agreement between theory and experiment in the linear regime. This lays the foundation for advances in connecting simulation data directly to nonlinear evolution due to thin-film stresses in a manner analogous to the “crater function framework” of the erosive-redistributive approach.

Finally, we give brief consideration to some directions for future work in both the linear and nonlinear regimes of pattern formation.
In recent years, many ion-induced nanopatterning experiments have been conducted on Si (due to its relevance to the semiconductor industry) at irradiation energies of 1keV or below (due to the ready availability of ion guns designed for this energy range). For this material at these energies, it is observed that as the ion-incidence angle $\theta$ increases from $0^\circ$ (normal incidence) to $90^\circ$ (grazing incidence), the patterns formed transition from flat surfaces, to ripples with wave vector parallel to the ion beam, to ripples with wave vector perpendicular to the ion beam, and that these transitions occur independently of ion energy [43, 68, 45]. Most of the theoretical models described in the Introduction exhibit similar behavior. Importantly, however, this behavior does not seem to be universal, and some researchers have reported a complete absence of patterns for some ion/target/energy combinations – we are motivated in particular by the experiments of (a) Teichmann et al. [57], who observed no patterns at any studied angle ($\leq 85^\circ$) for Ne$^+$→Ge or Ar$^+$→Ge between 400 and 2000 eV, and (b) Hofsäss et al. [22], who observed no patterns at any studied angle for Ar$^+$→Ge between 2 keV and 5 keV. These observations suggest the presence of an additional, unconditionally-stabilizing mechanism that becomes important in these regions of parameter space; however, this is puzzling, as all of mechanisms studied elsewhere are destabilizing for at least some incidence angles.

Recent experimental studies have suggested that impact-induced stresses may be the single greatest contributor to the transition from smooth surfaces to ripples at an incidence of around 45 degrees and an energy near 1 keV [28]. This suggests asking whether some novel stress-related mechanism could exist that is unconditionally stable, explaining the suppression of ripple patterns discussed above. In particular, whereas existing stress-based
pattern formation models have employed the convenient incompressibility condition at low energies [31, 58, 69, 32, 58, 33], researchers have long observed radiation-induced swelling at higher energies, induced by a variety of atomistic mechanisms [63, 70, 71, 72, 73, 65, 67, 74]. Accordingly, recent work by our group explored replacing the incompressibility condition with a simple mathematical model of isotropic swelling, and found that swelling does, indeed, supply the needed surface stabilization at all angles of incidence [48]. However, due to the mathematical complexities of the model, analytical solutions could only be obtained in the limit of very small swelling rates, whereas the observed pattern suppression would presumably occur above some critical value of the swelling rate.

In this work, we generalize the prior results to arbitrary swelling rates using new analytical and numerical approaches, closing the gap between theory and observation. After re-casting the system of equations resulting from linear stability analysis to a single equation, and developing a finite difference scheme for its solution, we compute the dispersion relation at arbitrary swelling rates, wave numbers and angles of incidence. We show that the observed angle-independent stabilization is not an artifact of the small swelling rate assumption used in [48]. Indeed, we demonstrate that the overall stability structure in the full parameter space is largely independent of swelling rates despite several features unique to the nonlinear regime (including a fascinating exchange of stabilities within a multi-branch dispersion relation). With this knowledge, we then turn to several relevant experimental systems, and discuss how the presence of swelling in the model can help us to understand those observations. With this work, therefore, we have significantly strengthened the hypothesis of a needed swelling-induced stabilization mechanism, highlighting the potential importance of this effect in future work, and an urgent need for more experimental data on swelling rates.

2.1. Preliminaries

We will use the same model as in [48]; here we will briefly summarize that model and some preliminary results obtained from its analysis.
2.1.1. Model

We consider a thin, crystalline material irradiated from above by an ion beam, with the z-axis oriented normal to the surface. The accumulation of radiation damage leads to the amorphization of the top few nanometers of the target, and this amorphous thin film is described using continuum equations for viscous fluid. Our goal is to solve equations in two dimensions for velocity \( \vec{v}(x, z) = u(x, z) \hat{i} + w(x, z) \hat{j} \), the density \( \rho(x, z) \), pressure \( p(x, z) \), and an age-tracking variable \( a(x, z) \) within a thin film bounded above by an amorphous-vacuum boundary \( z = h_2(x, t) \) and below by a crystalline-amorphous boundary \( z = h_1(x, t) \). This geometry is illustrated in the schematic in Figure 1.

![Schematic](image)

Figure 2.1. Schematic depicting ion bombardment at an incidence angle of \( \theta \), the resulting formation of an amorphous layer with thickness determined by the ion penetration depth, and an pictorial illustration of the proposed swelling mechanism. Note that for off-normal incidence, the bottom boundary \( z = h_1(x, t) \) may not align with the top boundary \( z = h_2(x, t) \).

**Bulk equations.** In the bulk, the standard equations for conservation mass and momentum, respectively, are

\[
\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \vec{v}) = 0 \tag{2.1}
\]

and

\[
- \nabla p + \eta(\nabla \cdot (\nabla \vec{v}) + \nabla (\nabla \cdot \vec{v})) = 0 \tag{2.2}
\]
where $\eta$ is the viscosity, and we have used the viscous limit of Stokes flow, which is justifiable due to the dominance of “creep” forces over inertial forces in the bulk. We note that the density is not constant, and therefore the incompressibility condition $\nabla \cdot \vec{v}$ does not apply. As a result, we must also supply an equation of state to relate the irradiation-induced damage to the density. As a simple model appropriate to a first study, we choose the equation

$$\rho = \frac{\rho_0^*}{1 + \alpha a},$$

(2.3)

where $\rho_0^*$ is the original density of the crystalline solid, $\alpha$ is a constant expansion rate, and $a$ is an “age” variable that tracks the length of time a parcel of material has existed within the irradiated film, according to a simple forced advection equation

$$\frac{\partial a}{\partial t} + \vec{v} \cdot \nabla a = 1.$$  

(2.4)

in which the 1 on the right hand side provides the steady increase in age as time progresses. Because Equation (2.3) lacks a pressure term, it can be thought of as a “quasi-incompressibility” condition – i.e. the density is allowed to change in response to changes in age, but not to changes in pressure. This preserves some of the analytical convenience of incompressibility, while allowing density changes due to irradiation.

It is worth re-emphasizing from Ref. [48] that our model of swelling is mechanism-agnostic. In other words, whichever specific atomistic mechanism causes the increase in volume over time (i.e. defects and accumulated damage, vacancies coalescing into voids, implanted ions forming bubbles, etc.), that process appears in Equation 2.3 only indirectly by way of the age variable. In this mathematical simplification, swelling simply proceeds linearly in time (i.e. proportional to accumulated fluence) once a parcel of material becomes amorphized at the bottom of the film, and proceeds until the parcel is removed by sputtering at the top of the film.

**Boundary conditions.** At the crystalline-amorphous (bottom) boundary $z = h_1(x,t)$, the standard no-slip and no-penetration conditions are applied simultaneously as
\[ \vec{v} = \vec{0}. \]  

(2.5)

At the amorphous-vacuum (top) boundary \( z = h_2(x,t) \), we have the following two boundary conditions:

\[ v\hat{n} = \vec{v} \cdot \hat{n} - \frac{V \rho^*}{\rho} \]  

(2.6)

and

\[ [T] \cdot \hat{n} = -\gamma \kappa \hat{n} \]  

(2.7)

where equation (2.6) is a modified kinematic condition that incorporates the sputtering of material at the free interface (\( V \) is the surface sputter velocity) [48]. Finally, equation (2.7) is a standard conservation of momentum applied at the interface, \([ \cdot ]\) denotes the jump across the interface, and the stress tensor is that of a standard Newtonian fluid,

\[ T = -pI + \eta(\nabla \vec{v} + \nabla \vec{v}^T), \]  

(2.8)

where bulk viscosity has been neglected, as is typical in continuum models of ion-irradiated nanopattern formation [31, 32, 33, 48] and in some other contexts, including the viscous leveling term frequently used as a regularizing mechanism [27]. Because the effect of surface tension in pattern formation is well-established [26], we here take \( \gamma = 0 \) to focus entirely on the effect of the swelling mechanism, as was done in [48].

In principle, this could lead to the neglect of cross-terms that originate from the interactions of surface tension and ion-induced swelling. However, it has been seen elsewhere [28] that it is often sufficient to consider a linear combination of the dispersion relations resulting from mechanisms which have been studied in isolation. Chapter 3 will also provide an exploration of the case that some swelling and anisotropic plastic flow interact, and we will show that elimination of interaction terms may be justifiable. Finally, we note that in [32], when the anisotropic plastic flow mechanism was considered alongside the viscous leveling (surface tension) effect, the final, full-spectrum linear dispersion relation was a simple super-
position, lacking cross-terms. A more comprehensive treatment of this mechanism would, ideally, leave $\gamma \neq 0$.

### 2.1.2. Previous results

In [48], the above model is first non-dimensionalized and translated into a frame of reference moving downward with the eroding surface at speed $V$. (In this frame of reference, material appears to pass upward through the crystalline/amorphous boundary, proceed to swell as it rises through the film, and finally be sputtered away once it reaches the free surface.) Next, steady-state solutions are computed exhibiting reflection, rotation, and translation invariance. Finally, the non-dimensionalized governing equations are linearized about the steady-state solution in normal modes with wavenumber $k$, leading to the following system of linearized, single-mode equations:

\begin{align}
\Sigma \rho_1 - \frac{A}{\psi^3} w_1 + \frac{1}{\psi} (iQ u_1 + u_1') + \psi \rho_1 + A \frac{1}{\psi} \rho_1 &= 0 \\
\Sigma a_1 + \psi a_1' + A \frac{1}{\psi} w_1 &= 0 \\
\psi \rho_1 + \frac{1}{\psi} a_1 &= 0 \\
-2Q^2 u_1 + u_1'' + iQ u_1' &= iQ p_1 \\
-Q^2 w_1 + 2w_1'' + iQ u_1' &= p_1'
\end{align}

with the following boundary conditions at $z=0$:

\begin{align}
a_1 &= -h_{11} A \\
u_1 &= 0 \\
w_1 &= -h_{11} A
\end{align}
and the following boundary conditions at $z=1$:

$$w_1 = \Sigma h_{21} - (1 + 2A) \rho_1$$  \hspace{1cm} (2.17)

$$u'_1 + iQ w_1 = -2iQ \frac{A}{\sqrt{1 + 2A}} h_{21}$$  \hspace{1cm} (2.18)

$$-p_1 + 2w'_1 = 0$$  \hspace{1cm} (2.19)

In the above, a subscript 1 denotes that the quantity has been linearized in normal (Fourier) modes and occurs at $O(\epsilon)$, retaining spatial dependence only on $z$. Hence $w_1$ occurs via the expansion $w(x,z,t) \rightarrow w_0(z) + \epsilon w_1(z) \exp(\sigma t + ikx)$, and likewise for the other quantities.

Here and throughout this Chapter, $Q = kh_0$ is the dimensionless wave number of a perturbation, $\Sigma = \sigma h_0$ is the dimensionless growth rate of that perturbation, $A = \alpha h_0/V$ is the dimensionless swelling rate of the film, and $\psi = \sqrt{1 + 2Az}$. Physically, the parameter $A$ quantifies the relative increase in volume of a parcel of material by the time it is sputtered away, as discussed above (i.e., $A = 0.1$ reflects an increase of 10% in the volume of a parcel of matter by the time it is sputtered away, and $A = 1$ reflects a doubling in the volume).

As discussed in [48], Equations (2.9)-(2.19) are seemingly not amenable to an exact analytical solution. However, a second linearization in the small-$A$ limit leads to solvable equations, which yield the small-$A$ dispersion relation

$$\Sigma = \left[ \left( 1 - \frac{\cosh(Q) + Q \sinh(Q)}{Q^2 + \cosh^2(Q)} \right) \frac{h_{11}}{h_{21}} - \frac{Q^2}{Q^2 + \cosh^2(Q)} \right] A + O(A^2).$$  \hspace{1cm} (2.20)

where $h_{11}$ and $h_{21}$ are the lower and upper interfaces linearized in normal modes, respectively. This dispersion relation contains a pocket of stable wavenumbers approximately between $Q = 0$ and $Q = 1.5$ for all angles of incidence. A plot of (2.20) at normal incidence is reproduced in Figure 2.2 (a).

Although this result holds for “small enough” swelling rates where the second linearization $A \ll 1$ is valid, we note that if isotropic swelling is hypothesized to suppress ripple formation under some set of conditions, the swelling rate must presumably be “large enough” to do so. This necessarily requires the consideration of larger swelling rates. We therefore now turn

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our attention to the problem of equations (2.9) - (2.19) for arbitrary values of $A$, with two main goals in mind: first, to develop a numerical method with which we can conduct a full modal analysis for arbitrary values of $Q$; and second, to study the dependence of pattern formation on the full $(Q,A,\theta)$ space and compare with the small-$A$ results.

2.2. Numerical methods

In this section, we describe in detail an approach to determine the linear dispersion relation numerically for arbitrary non-dimensional swelling rates $A$. We will restrict our attention to the parameter range $A \in [0, 1]$, which should be sufficient to capture most physical systems, even those such as irradiated Ge that exhibit extreme amounts of volumization.

2.2.1. Reformulation in density

We return to the full linearized equations, (2.9)-(2.19), whose dispersion relation we seek for all of $(Q,A,\theta)$ parameter space. As written, (2.9)-(2.19) is a system of five ODEs with six boundary conditions, one of which (the kinematic condition, (2.17)) must be reserved in order to determine $\Sigma$. However, in doing this, we would have no boundary condition available for the solution of $\rho_1$. Indeed, the boundary conditions at $z=1$ all involve multiple unknowns, complicating numerical solution. One remedy is to simply reduce the system into the density, which yields a single fifth-order ordinary differential equation with six boundary conditions. Although this would appear to be too many boundary condition for a fifth-order ordinary differential equation, the reservation of the linearized kinematic condition as a compatibility condition will allow us to solve for $\sigma$ as an eigenvalue; that is, there will exist solutions satisfying all six boundary conditions for appropriate values of $\sigma$, the growth rate which we wish to determine.

The resulting equation is amenable to solution by simple finite differences, and may also serve as a staging point for an exact analytical solution in the future. The dimensional reduction is performed as follows:

1. Solve for $a_1$ in terms of $\rho_1$ in equation (2.11).
2. Substitute the above result into equation (2.10) so that \( w_1 \) is expressed in terms of \( \rho_1 \).

3. Substitute the expression for \( w_1 \) in terms of \( \rho_1 \) into equation (2.9) and solve for \( u_1 \) in terms of \( \rho_1 \).

4. Substitute expressions for \( w_1 \) and \( u_1 \) in terms of \( \rho_1 \) into equations (2.12) and (2.13).

5. Differentiate the new equation (2.12) with respect to \( z \) and multiply the new equation (2.13) by a factor of \( iQ \).

6. Set these new equations (2.12) and (2.13) equal to each other to yield the fifth-order ordinary differential equation ("ODE") with variable coefficients.

7. Apply the implied relationships between unknowns to the boundary conditions.

The result of these operations is the ODE in the bulk

\[
(Q^4\Sigma + 2AQ^4(4\Sigma z + \psi) + A^4(16Q^4z^3\psi + \Sigma(3 - 4Q^2z^2)) + 8A^3Q^2z(3Q^2z\psi + \Sigma(-3 + 4Q^2z^2)) + 6A^2((-Q^2)\Sigma + 2Q^4z(2\Sigma z + \psi)))\rho_1 - \psi^2((-Q^4)\psi - 6A(-2Q^2\Sigma + Q^4z\psi) + A^2Q^2(48\Sigma z + \psi(31 - 12Q^2z^2)) + 2A^3(Q^2z\psi(31 - 4Q^2z^2) + 5\psi) + A^2(-9\Sigma + 4Q^2\Sigma z^2 + 20Q^2z^2))\rho_1'' + (2Q^2\psi + A(-12\Sigma + 8Q^2z^2)) + A^2(-24\Sigma z + \psi(-63 + 8Q^2z^2))\rho_1^{(3)} - \psi^2((\Sigma + 2A\Sigma z + 18A\psi)\rho_1^{(4)} + \psi^3\rho_1^{(5)})) = 0,
\]

where \( \rho_1^{(j)} \) denotes the \( j \)th derivative in \( z \). We note that the above is linear and homogeneous, along with the following boundary conditions at \( z=0 \):

\[
Ah_{11} - \rho_1 = 0 \quad (2.22)
\]

\[
3A^2\rho_1 + 6A\rho_1' + 3A\rho_1\Sigma + \rho_1'' + \Sigma\rho_1' = 0 \quad (2.23)
\]

\[
A^2h_{11} + 2A\rho_1 + \rho_1' + \rho_1\Sigma = 0 \quad (2.24)
\]

and the following boundary conditions at \( z=1 \):

\[
\frac{\psi^2(2A\rho_1' + \psi\rho_1\Sigma + 3A\rho_1 + \rho_1')}{A} - h_{21}\Sigma = 0 \quad (2.25)
\]
\[
\frac{2A^2 h_{21} Q^2}{\psi} + 4A^2 \rho_1^{(3)} + 20A^2 \rho_1'' + \frac{2A^2 \Sigma \rho_1'}{\psi} + 15A^2 \rho_1' + \frac{3A^2 \rho_1 \Sigma}{\psi} + \psi^2 Q^2 (2A (\rho_1' + \rho_1) + \rho_1') + \\
\psi^2 Q^2 \rho_1 \Sigma + 4A \rho_1^{(3)} + 2A \psi \Sigma \rho_1'' + \psi \Sigma \rho_1'' + 10A \rho_1'' + 5A \psi \Sigma \rho_1' + \frac{A \Sigma \rho_1'}{\psi} + \rho_1^{(3)} = 0
\]

(2.26)

\[
A^3 (4Q^2 (6\psi \rho_1'' + 6(3\psi + \Sigma) \rho_1' + \rho_1 (5\psi + 9\Sigma)) - 2\psi (35\rho_1'' + 4(\rho_1^{(4)} + 7\rho_1^{(3)})) + \Sigma (3\rho_1 - 2(4\rho_1^{(3)} + 18\rho_1'' + 9\rho_1') + A^2 (2Q^2 (18(\psi \rho_1'' + (2\psi + \Sigma) \rho_1') + \rho_1 (5\psi + 18\Sigma)) - \psi (12\rho_1^{(4)} + 56\rho_1^{(3)} + 35\rho_1') - 3\Sigma (4\rho_1^{(3)} + 12\rho_1'' + 3\rho_1') + 3Q^2 (\psi \rho_1'' + \Sigma \rho_1') + A (9Q^2 (2\psi (\rho_1'' + \rho_1') + \Sigma (2\rho_1' + \rho_1)) - \\
2\psi (3\rho_1^{(4)} + 7\rho_1^{(3)}) - \Sigma (2\rho_1^{(3)} + 3\rho_1'')) - \psi \rho_1^{(4)} - \rho_1^{(3)} \Sigma = 0
\]

(2.27)

Hence we have converted a system of five differential equations in five unknowns with six boundary conditions into a single differential equation in one unknown with six boundary conditions. Although, superficially, the form of the expressions is substantially worsened by bringing them into a single unknown, finite difference methods for a single ODE are often very convenient to write and implement.

2.2.2. Numerical algorithm

Mathematically, (2.21)-(2.27) represent an eigenvalue problem for the growth rate \(\Sigma\). In cases where such equations are solvable in closed form, it is typical to reserve one boundary condition (most naturally the kinematic condition, where \(\Sigma\) appears upon application of the linear stability ansatz), solve the system of differential equations with the remaining boundary conditions and \(\Sigma\) remaining an arbitrary parameter up until the end, and then to substitute the computed solution into the reserved boundary condition to determine \(\Sigma\). The parameter \(\Sigma\) is then known in terms of the other system parameters, possibly as an implicit function. It is this approach that we seek to recreate numerically in a least-squares sense. Accordingly, we reserve (2.25), the kinematic condition, from which we define

\[
\Pi(\Sigma; Q, \theta, A) = \frac{\psi^2 (2A \rho_1' + \psi \rho_1 \Sigma + 3A \rho_1 + \rho_1')}{A} - h_{21} \Sigma
\]

(2.28)
which is just the left side of (2.25), and whose $\Sigma$ roots we wish to compute for a given $(Q, \theta)$ pair. In general, $\Sigma$ is complex-valued, and so $\Pi$ will also be complex valued. To obtain the $\Sigma$ roots, we therefore seek to minimize an objective function

$$\Phi(\Sigma; Q, \theta, A) = |\Pi|^2 = \text{Re}(\Pi)^2 + \text{Im}(\Pi)^2$$

which is simply the squared modulus of $\Pi$. Numerically, we designate $\Sigma (Q, \theta)$ to be a root if $\Phi(\Sigma; Q, \theta, A) < \varepsilon$. For convenience, we used Matlab’s `fminsearch` function to determine the $\Sigma$ roots, and our overall method may thus be summarized as follows:

1. Set values of $A, Q, \theta$ for which we desire $\Sigma$.

2. Set initial guess for $\text{Re}[\Sigma]$ and $\text{Im}[\Sigma]$.

3. Solve BVP (2.21)-(2.27) using finite differences.

4. Evaluate $\Phi(\Sigma; Q, \theta, A)$.

5. **If** $|\Psi| < \varepsilon$, assign $\Sigma(Q, \theta, A)$ and **exit**

6. **Else**, adjust $\text{Re}[\Sigma]$ and $\text{Im}[\Sigma]$, **go to 3**.

In order to sweep over the minimization process, we must solve the system (2.21)-(2.27) at each iteration. We use a typical second-order central difference scheme to approximate the first through fifth order derivatives, and ghost points on each end of the domain to facilitate enforcement of the boundary conditions.

“Ghost points” are nonphysical extensions of the domain of solution which allow the application of, for example, central finite differences at boundaries where one or more of the points required for the stencil would be undefined. By considering what the value of the ghost point *would be* if the domain were continued, we can express the value at the out-of-domain grid point in terms of in-domain grid points simply by solving for them algebraically using the finite difference approximations of the boundary conditions that should apply there. However, we deviate from the standard approach in an important way.
Crucially, the reservation of (2.25) at \( z = 1 \) for use as an objective function reduces the number of available ghost points at \( z = 1 \) from three to two, precluding the use of the typical second-order central finite difference for a fifth derivative, whose stencil involves three points on each side. Consequently, we use an off-center (but still second-order) difference scheme for the fifth derivative at \( z = 1 \), with weights \( \frac{1}{2}, -4, \frac{25}{2}, -20, \frac{35}{2}, -8, \frac{3}{2} \) at grid points \( z_{i-4}, z_{i-3}, z_{i-2}, z_{i-1}, z_{i}, z_{i+1}, z_{i+2} \) respectively. With our custom stencil, we require only two ghost points to the right of the interval, which are uniquely determined by the two boundary conditions at hand.

2.2.3. Verification and convergence

To verify the accuracy of our approach, we compare the analytical solution in the small \( A \) limit (reference) with the numerical solution for a small value of \( A = .001 \). In Figure 2.2(a),

\[
\begin{align*}
\text{analytical} & \quad \text{numerical, } A = .001 \\
\end{align*}
\]

Figure 2.2. (a) Comparison between analytical and numerical results for small \( A \) and normal incidence. (b) Convergence analysis of our finite difference solver for \( A = .2, Q = .7, \theta = 0 \), using \( h = \frac{1}{128}, \frac{1}{64}, \frac{1}{32}, \frac{1}{16} \) and taking \( h = \frac{1}{256} \) to be the true solution. We obtain a slope of 1.9943 between values associated with \( h = \frac{1}{256} \) and \( h = \frac{1}{128} \) on the log-log plot, consistent with second order convergence.
we have reproduced the small-A dispersion relation from our numerical results, confirming that our combination of root-finding and finite difference methods is viable. In Figure 2.2(b), we have conducted a convergence analysis for $Q = .7$ at $A=.2$ and normal incidence, taking mesh spacing $\Delta z = \frac{1}{256}$ to be the “true” solution, then coarsening the mesh and recording the $L^2$ norm of the error between “true” and obtained. We see a slope of approximately 2, reflecting roughly second order convergence, as expected. Other parameter choices produce similar convergence plots, giving further assurance that our method works correctly.

2.3. Stability results

We now use our numerical method to obtain two main sets of results. First, we obtain the full dispersion relation and relative flow fields within the film at normal incidence, and conduct a careful study of their scaling in the swelling rate $A$. Second, we consider the $(Q, \theta)$ dependence of both pattern growth and translation for several relevant values of $A$. We will demonstrate that the key results for vanishingly-small swelling rate $A$ are reproduced for arbitrary $A$, while also investigating the complexities of the non-small rate.

2.3.1. Dispersion relation computed numerically: normal incidence

We begin with a simple depiction of the growth rates $\text{Re}[\Sigma]$ in the normal incidence case. We note that, because swelling is an isotropic effect, the incidence angle appears in the governing equations only indirectly, through the relationship between the top and bottom boundaries. For normal incidence, we expect the bottom boundary to be a simple downward translation of the top boundary by a distance $h_0$. This means the perturbations to the bottom boundary are in phase with the perturbations to the top boundary – i.e.

$$h_{11} = h_{12}$$

We also note that under normal incidence irradiation, the growth rate is purely real due to symmetry considerations.

In Figure 2.3(a) we have numerically computed the growth rate for several values of swelling rate $A$ ranging from $A=.01$ (1 percent volume increase before sputtering) to $A=1$.
(100 percent volume increase before sputtering). It is clear that the characteristic swelling-induced pocket of stable wave numbers below $Q = 2$ persists for all swelling rates depicted. We observe that the magnitude of the growth rate $\text{Re}[[\Sigma]]$ increases significantly as the swelling rate $A$ increases. This is expected, but complicates a comparison of the qualitative features of $\text{Re}[[\Sigma]]$ for different values of $A$. Therefore, in Figure 2.3(b), we scale each growth rate curve by its limiting value $C_\infty := \lim_{Q \to \infty} \Sigma(Q, A)$. The scaled curves are remarkably similar at all values of $A$, nearly collapsing onto the curve associated with the small-$A$ limit. This observation, alone, significantly answers in the affirmative some of our motivating questions concerning the applicability of the small-$A$ result in experimental settings.

Closer examination of Figure 2.3b shows that the neutrally stable wavenumber $Q_{\text{neutral}}$ is pushed slightly to the right for increasing $A$. Compared to its value $Q_{\text{neutral}} \approx 1.508$ in the small-$A$ limit, we observe that for $A=.1$, $Q_{\text{neutral}} \approx 1.525$, and for $A=1$, $Q_{\text{neutral}} \approx 1.653$. Thus, an increased swelling rate $A$ is seen to slightly increase the range of stable wavenumbers for normal-incidence irradiation. Finally, we observe that the roots at $Q=0$ and $Q \approx 1.508$ do not change very much for different values of $A$, while the curvature of the dispersion relation between these two roots appears to somehow scale by $A$. This prompts an exploration of the long-wave dispersion relation, and we find that, indeed, scaling by the second-derivative in $Q$ of the arbitrary-$A$ long-wave dispersion relation provides a good approximation (see Appendix) or, equivalently, by twice the coefficient. In addition, the results of the long-wave analysis analytically confirm the angle-independent stabilization of small wavenumbers.

As a visual aid to understanding the effect of the swelling mechanism on the relative flow field within the amorphous thin film, we consider in (Figure 2.4) a few wavenumbers for normal-incidence ion beam irradiation at both $A=.1$ (10 percent volume increase before sputtering) and $A=1$ (100 percent volume increase before sputtering). The wavenumbers chosen are $Q \approx 0.7$, which is approximately the most stable wave number, a neutrally stable wave number depending on $A$, and $Q \approx 2$, a wave number which is unstable for all $A$ that we have considered here.

It is clear from the relative flow field that for $Q=0.7$, material tends to flow from hilltops
into valleys, which has the net effect of reducing disparities between highs and lows over time. Near each respective neutrally stable wave number, vortices have begun to form which exactly balance the loss of material from hilltops with a cycling of material forced below the surface in the valleys back into the hilltops. This has the net effect of preserving the distance between hilltop and valley over time, hence neutral stability. Finally, the relative flow field for unstable wave number Q=2 depicts the case where the redistribution is dominated by shear forces, with material from valleys being driven beneath the surface and then forced uphill by the sub-surface vortical flow faster than lateral expansion can compensate. This leads to growth in distance between hill-top and valley-bottom, hence instability and pattern formation.

Fundamentally, these observations are attributable to a competition between two phenomena. First, the hilltops are driven to expand laterally into valleys by the swelling mechanism. Second, shear forces tend to redistribute material vertically due to the different rate of sputtering in the hilltops and valleys, because the time until sputtering (the “dwell time”) is proportional to the depth of film relative to a given parcel of matter. For long waves, the former dominates, whereas for short waves, the latter dominates. This basic mechanism is fundamentally unchanged by different values of A. As can be seen, the proposed swelling mechanism continues to exert a robust, qualitative effect on the dispersive behavior of the medium even when A is non-vanishing. This demonstrates that the earlier findings are not attributable to the smallness of the swelling rate that was originally considered, and are representative of a real effect that is manifest at all positive swelling rates.

2.3.2. Exploration of full (Q,A,θ) dependence

We now turn our attention to the behavior of Re[Σ] and Im[Σ] at incidence angles other than zero. To explore the effect of off-normal incidence, we follow [33, 48] and assume that the bottom boundary is still translated a distance \( h_0 \) from the top boundary, but in the direction of the ion beam. Hence, by simple geometry,

\[
h_1(x, t) = h_2(x - h_0 \sin(\theta), t) - h_0 \cos(\theta)
\]  

(2.31)
This reduces the steady film depth according to

\[ h_{0\theta} = h_0 \cos(\theta), \]  

(2.32)

which causes a corresponding re-definition of the dimensionless wave numbers:

\[ Q_\theta = Q \cos(\theta). \]  

(2.33)

Finally, it also induces a phase shift in the perturbations of the top and bottom boundaries, yielding:

\[ h_{11} = h_{21} e^{-iQ_\theta \tan(\theta)} \]  

(2.34)

We will consider the dependence of Re[\Sigma] and Im[\Sigma] on a range of angles between \( \theta = 0^\circ \) and \( \theta = 90^\circ \), but we will neglect the relative flow fields for these angles in favor of representing the data more broadly as two pairs of heat maps in \( A \) and \( \theta \). Recall that we are ultimately interested in the behavior of \( \epsilon \)-small perturbations to a steady state:

\[ f(x, z, t) = f_0(z) + \epsilon \tilde{f}(z)e^{\Sigma t + iQx}. \]  

Then we have

\[ f(x, z, t) = f_0(z) + \epsilon \tilde{f}(z)e^{\text{Re}[\Sigma]t + iQ(x + \frac{\text{Im}[\Sigma]}{Q})} \]  

(2.35)

such that Re[\Sigma] is associated with growth (\( >0 \)) or decay (\( <0 \)) in time, and Im[\Sigma] is associated with translation left (\( >0 \)) or right (\( <0 \)) on the x-axis.

2.3.2.1. Real part

Comparing with the results of [48], Figure 2.5 demonstrates that the angle dependence observed for the vanishingly-small swelling rate is reproduced at higher swelling rates with a strong analogy between each \( \Sigma(Q, \theta; A) \).

Comparing the first row of the heatmap (or, equivalently, Figure 2.3) to the associated result for the \( A \to 0 \) limit [48], we see near perfect agreement. As \( A \) increases the general form of the real part of sigma remains unchanged, while some interesting behavior is seen in the imaginary part, which we will discuss below. This again confirms that the qualitative behavior of the swelling mechanism is not strongly dependent on the swelling rate \( A \), further confirming the motivating hypothesis of this work.
Focusing on the real part (left-hand column), some minor dependence on \( A \) may be noted. In particular, increased \( A \) is associated with an expansion of the unstable regions in general, and a contraction of the stable regions. At the same time, the small-angle stability frontier is pushed into higher wave numbers, and the large-angle stability frontier is reduced in its extent. This is most evident at \( \theta = 0 \) and \( \theta = 45 \). For \( \theta = 0 \), \( A = .1 \) yields \( Q \approx 1.525 \) as first neutrally stable wave number greater than 0, whereas \( A = 1 \) yields \( Q \approx 1.653 \) as the first neutrally stable wave number greater than 0, suggesting that increased swelling rate is associated with slightly larger wave numbers becoming stable: a stabilization of larger wave numbers at small angle of incidence. On the other hand, for \( \theta = 45 \), with \( A = .1 \), the first non-zero neutrally stable wave number is \( Q \approx 6.5 \), and for \( A = 1 \), it is \( Q \approx 6 \). We also note that previous results for vanishingly-small \( A \) yield \( Q \approx 7 \) ([48]). This effect is perhaps most evident around \( \theta = 35 \), where the expansion of the unstable region for smaller angles has “cut off” a previously stable region, now destabilizing it. This illustrates that although the existence of a swelling mechanism always induces stability for small wave numbers, it may stabilize or destabilize wave numbers in the general phase space depending on \( A \) and \( \theta \).

2.3.2.2. Imaginary part

Turning our attention to the imaginary part of \( \Sigma \), we again see marked similarity between images for different values of \( A \), with one notable exception: the apparent replacement of some of the neutral stability regions by sudden changes in the direction of translation, which is unexpected. Indeed, investigating a “slice” of the heat map at \( \theta = 45^\circ \) for different values of \( A \) (Figure 2.6), we observe that, for sufficiently high \( A \), discontinuities appear to form!

The key to understanding these apparent discontinuities is twofold. First, after further investigation, we notice that they occur when the upper and lower interfaces \( h_{11} \) and \( h_{21} \) are exactly out of phase, which is to say that \( \frac{h_{11}}{h_{21}} = -1 \). Equation (2.34) implies that this occurs when

\[
e^{-iQ\sin(\theta)} = -1 = e^{-i(2n+1)\pi}
\]  

(2.36)
or explicitly, when

$$\theta = \arcsin \left( \frac{(2n + 1)\pi}{Q} \right)$$

(2.37)

These curves are plotted as dashed black lines in Figure 2.5, and it can be seen in Figure 2.6 that the apparent discontinuities lie directly upon them. Second, we must realize that the out-of-phase interface configuration exhibits left-right symmetry, such that any solution with non-zero translation rate must have a partner solution moving in the opposite direction with the same speed. These observations suggest that the solution for $\Sigma$ contains more than one branch, and that what appear to be discontinuities in $\text{Im}[\Sigma]$ may in fact simply represent an exchange of stability between branches.

With such consideration in mind, we now treat the regions around the apparent discontinuities more carefully. By adjusting the initial guesses that our numerical solver makes in minimizing the error (2.29), we indeed are able to obtain more than one solution branch for $\Sigma$, which are shown in Figure 2.7.

We now see clearly that at the point of the “discontinuity” in $\text{Im}[\Sigma]$, the two branches of $\text{Re}[\Sigma]$ cross. This complicates visualizations of the dispersion relation, which reflect a traditional focus of stability analyses only on the branch with the most positive value of $\text{Re}[\Sigma]$. Our more careful computations reveal that this maximal value changes branches at a critical value of $Q$, resulting in the apparent cusps and discontinuities seen in Figure 2.6.

It turns out that the separation of the two intersecting branches of $\text{Im}[\Sigma]$ begins around $A^* \approx 0.55$ for $\theta = 45$. Although we have not shown it here, analogous transitions occur near $Q \approx \frac{6\pi}{\sqrt{2}}$ for $\theta = 45$, and, more broadly, near the entire level curve defined in (2.37) at various critical thresholds $A^*(Q, \theta)$. Thus all of the rapid transitions in Figures 2.5 and 2.6 can be explained.

2.4. Discussion

In this section, we consider the implications of our results for the interpretation of certain experimental observations. We begin with some general observations on how the swelling mechanism should affect the pattern-forming properties of a system. It is essential to note
that, as discussed in [48], swelling operates simultaneously with other, previously-studied mechanisms such as sputter erosion [7, 8, 9, 10], mass redistribution [11, 12, 13, 14], and anisotropic stress-driven flows [31, 58, 32, 58, 33]. A schematic exploration of the addition of the swelling mechanism to these existing models is illustrated in Figure 2.8. There, we see that a modest amount of swelling will be expected to increase the transition angle separating flat surfaces from rippled surfaces, whereas a large amount of swelling might be expected to suppress ripple formation entirely. A detailed attempt to estimate all relevant parameters is beyond the scope of this work, but in what follows we provide some qualitative discussion in the context of relevant experiments.

2.4.1. Comparison with selected experiments

\( \text{Ar}^+ \rightarrow \text{Si below 1000 eV.} \) We first review observations in a “reference” system of \( \text{Ar}^+ \rightarrow \text{Si} \) at energies below 1000 eV, as studied by Madi et al. [43, 68, 45]. At near-normal incidence, patterns are not observed, but above a transition angle of about 45°, ripples emerge with wavevector parallel to the ion beam direction. Finally (although this is not our focus), above a second transition angle of around 80°, these “parallel-mode” ripples are replaced by “perpendicular-mode” ripples with wavevector perpendicular to the ion beam. In this energy range, the transition angles are not observed to depend strongly on the ion energy. We would conclude that any swelling in this system is either present in low amounts, or does not depend strongly on the ion energy within this energy range.

\( \text{Ne}^+, \text{Ar}^+ \rightarrow \text{Ge below 2000 eV.} \) We now turn to the irradiation of Ge by various ions at similar energies, as reported by Teichmann et al. [57]. Surprisingly, for \( \text{Ne}^+ \) and \( \text{Ar}^+ \) ions, no pattern formation is observed at any angle of incidence in this energy range (at least, no angle less than 75°, which was the maximum angle studied). This observation is consistent with our model if we assume that Ge experiences much more swelling than Si under similar irradiation conditions. And in fact, this is precisely the case – Ge can be shown to readily double in volume [63, 64] due to the stabilization of vacancies and their accumulation into
voids [66, 75].

**Kr⁺,Xe⁺→ Ge below 2000 eV.** Teichmann et al. also studied irradiation of Ge by the heavier ions Kr⁺ and Xe⁺ [57], where “standard” patterning behavior is observed, with two important differences. First, the transition angles are much higher than for the Ar⁺→Si system (in the vicinity of 65°). Second, in a series of experiments at 65°, strong ripples were observed at 400 eV, weaker ripples at 800 and 1200 eV, and no ripples at 2000 eV. Repeating these experiments at 75°, the authors observed ripples at all energies, and therefore concluded that the transition angle had increased with increasing ion energy. These behaviors are also consistent with our findings, if we assume that the relevant strength of swelling increases with ion energy; indeed, this is suggested by the experiments of Böttger et al [76].

**Ar⁺ → Si above 2000 eV.** Finally, we consider some observations for the Ar⁺→Si system at higher energies between 650 eV and 10 keV, as reported by Hofsäss et al. [22]. These authors performed a series of experiments at 65°, observing strong patterns at 650 eV, weaker patterns at 1 keV and 1.3 keV, and essentially no patterns between 3 keV and 10 keV. Being essentially identical to the results in [57] for Kr⁺ and Xe⁺ on Ge, these observations are also consistent with our findings, under the assumptions described above.

2.4.2. Directions for future work

**Transition angles: vanished or moved?** The observations above naturally suggest the question of whether a transition from flat to rippled surfaces was, in fact, entirely suppressed in any of the above systems, or whether it simply moved to a higher angle of incidence. For the Kr⁺,Xe⁺→ Ge system, when increasing energies caused ripples to vanish at 65°, Teichmann et al. repeated their experiments at 75°, confirming that ripples persist at all energies for this higher incidence angle. However, for the Ne⁺,Ar⁺→ Ge system, when ripples were not observed at any angle below 75°, those authors did not report further experiments at even higher angles. Similarly, for the Ar⁺→ Si system, when increasing energies caused
ripples to vanish at 65°, Hofsäss et al. did not report further experiments at higher angles [22]. We therefore consider these regions of parameter space to be of high interest for continued study.

We note that in Ref. [22], Hofsäss et al. argued that ripples could not form for any angle at these energies, based on simulations using a modified version of the Crater Function Framework proposed in [20, 21]. However, this reasoning assumes prompt erosive and redistributive effects alone determine patterning behavior, which is inconsistent with recent studies of ion-induced stresses [31, 58, 32, 69, 33, 28]. Moreover, as we have noted elsewhere [6], several of the proposed modifications do not appear to be mathematically justified.

**Model refinements: swelling saturation.** The simple model of swelling represented by Equation (2.3) has the advantage of mathematical simplicity, and generalization across different atomic-level volumization mechanisms. One disadvantage is that it does not contain a saturation mechanism. This was not a problem in Ref. [48] when the rate of swelling was assumed to be small. For larger swelling rates, this may or may not need refinement. We note that material parcels undergo swelling only until they are sputtered away at the top surface. Therefore, if the “dwell time” \( \tau_{\text{dwell}} = \frac{h_0}{V} \) needed to sputter away one film depth’s worth of material is less than the time \( \tau_{\text{sat}} \) at which the stress would saturate without sputtering, then the linear approximation in Equation (2.3) remains reasonable. This seems to be the case at least for the “reference” system of Ar\(^+\)→Si below 1 keV [44]. If \( \tau_{\text{FD}} > \tau_{\text{SS}} \), a refinement would be needed. We defer such refinement to future work.

**Energy dependence: a complex space.** The energy dependence of patterning behavior is highly complex. For instance, in the Ar\(^+\)→Si system, [22] identify three distinct energy regimes. In “Region 1” (below 1 keV), patterns are readily formed at angles above 45 degrees [43]. In “Region 2” (from 1 keV - 20 keV), patterns are less easily formed, with a higher transition angle that can exceed 65 degrees, and may potentially vanish entirely [22]. Finally, in “Region 3” (above 20 keV), patterns again readily formed, and the transition angle decreases again, potentially as low as 30 degrees at an energy of 40 keV [77]. So,
although we have suggested that an energy-dependent swelling may explain the transition from “Region 1” to “Region 2” type behavior in some relevant systems [57, 22], a simple extrapolation of this trend to all energies is obviously insufficient.

There are several reasons to expect that a more nuanced approach should be needed. First, as shown in [48], the “dwell time” \( \tau_{\text{dwell}} \) introduced above has a minimum value at around 1-2 keV. Above this energy, it increases steadily, doubling by 10 keV, and doubling again by 30 keV. As material spends increasing amounts of time in the amorphous film prior to sputtering, we may expect the importance of bulk physics (such as stress-driven viscous flow [31, 58, 32, 58, 33]) to increase relative to surface physics (such as surface sputtering and redistribution [7, 8, 9, 11, 12, 13, 18, 19]). An increasing dwell time may also necessitate the inclusion of a saturation mechanism as just described, which in turn, could limit the suppressive effect of swelling as energies continue to increase. Second, the fundamental physics of ion/solid interactions are changing at these energy levels. Ions moving through solids undergo both nuclear and electronic stopping [78], and above a certain energy, electronic stopping becomes dominant. Indeed, for the Ar→Si system, nuclear stopping reaches a peak at around 20 keV [79] – exactly the boundary between “Region 2” and “Region 3” as described by [22]. In short, because the underlying physics of the system are changing, it is not unreasonable that trends in the relative magnitude of parameters might change direction in this region. Indeed, ion-solid interactions at higher energies have previously been described using entirely different modeling approaches (see, e.g. [34, 60, 35, 61, 36, 37, 38, 39, 40, 41]. Much more work is needed, but we hope this contribution motivates future studies.

2.5. Conclusion

As described above, past work from our group [48] has shown that a simple mathematical model of radiation-induced swelling, a mechanism historically neglected in theoretical treatments of ion-induced nanopatterning, is unconditionally stabilizing. This may, in principle, provide a means of explaining the disappearance of ion-induced nanopatterns observed for
certain ion/target/energy combinations [57, 22]. However, an important limitation to the previous work was its restriction to the limit of small swelling rates. While the analysis showing unconditional stability of the swelling mechanism used the assumption of a small swelling rate \( A \) in order to render the governing equations analytically solvable, the actual suppression of pattern formation observed experimentally would only be expected to appear for relatively large swelling rates. Hence, some means of “bridging the gap” between these two regimes was needed. This work has provided such a bridge by employing a numerical study of the linear stability equations, an approach not commonly seen but broadly applicable to other systems.

The present work overcomes the previous limitations on parameter \( A \), successfully bridging the gap: the stabilization of observed wavelengths is not a transient artifact of the small swelling rate assumption made originally, but is, rather, due to the nature of the swelling mechanism itself. Indeed, there are very strong similarities between the behavior associated with small swelling rates, and that associated with even very large swelling rates. In fact, at normal incidence, the behavior over different wavenumbers and swelling rates is very nearly separable, and we have derived a simple prefactor function that scales the small-swelling result into the arbitrary-swelling regime. The persistence of these stabilization effects for all swelling rates strengthens the argument for adding a swelling-type mechanism to models of nanopattern formation, and the great similarities in the dispersion relation across all swelling rates suggests that the small-swelling limit obtained in [48] is an adequate approximation to the full mechanism.

Having verified the behavior of the swelling mechanism for arbitrary swelling rates, we also made some testable predictions of its effect on experimental systems. The most important of these is that swelling should increase the transition angle between flat and patterned surfaces, potentially eliminating patterns entirely if it is strong enough. Combined with the knowledge that irradiated Ge readily swells to almost double its initial volume [64], this prediction would explain the observations of Teichmann et al. [57], who observed no ripples in the \( \text{Ne}^+ \rightarrow \text{Ge} \) and \( \text{Ar}^+ \rightarrow \text{Ge} \) systems at any incidence angle, and ripples only at high incidence angles in
the Kr$^+\rightarrow$Ge and Xe$^+\rightarrow$Ge systems. If we further predict that swelling increases with ion energy in this energy range, we can additionally explain the increasing transition angles observed by Teichmann et al. in Kr$^+\rightarrow$Ge between 400 eV and 2000 eV, and by Hofsaß et al. in Ar$^+\rightarrow$Si between 650 eV and 3000 eV [57, 22].

These theoretical advancements, and associated predictions, highlight a paucity of experimental measurements needed to compare the relative magnitudes of active mechanisms in ion-irradiated solids. For instance, to quantify the discussion presented in Section 2.4.1, we would need simultaneous measurements of both the (isotropic) swelling rate (isotropic stress) and the (deviatoric) stress-driven viscous flow rate, across a wide range of energies. Although both swelling and stress can be measured independently, we are aware of only one limited study that attempts to distinguish between these two quantities, by collecting angle-dependent measurements of cantilever curvature [47]. That study does appear to suggest the presence of both effects even at low energies (250eV Ar$^+$ on Si), further indicating that the inclusion of swelling effects may be necessary to obtain the quantitatively-accurate “predictive model” so long sought in this field [6]. More experimental work in this area will be crucial component of this ongoing pursuit.
Figure 2.3. Dispersion relations computed numerically. (a) Unscaled, showing behavior in absolute terms. We note an apparent preservation of shape. (b) Scaled by the large-$Q$ asymptote for each $A$, demonstrating that the $Q \to \infty$ limit provides a natural scaling factor. (c) Scaled by the second derivative of the long-wave coefficient.
Figure 2.4. Comparison of relative flow fields for $A=0.1$ versus $A=1$. We plot the velocity fields $\vec{v}_{11}(x, z; Q) - \vec{v}_{11}(x, z; 0)$ for several representative wave numbers. Subtracting off $\vec{v}_{11}(x, z; 0)$ isolates the components relevant to linear stability by removing a strong, constant, downward velocity associated with the traveling frame, as in [48]. First row: lateral expansion drives the flow of material from hilltops into valleys, tending to stabilize. Second row: lateral expansion and shear forces attempting to pull material uphill are evenly matched, leading to neutral stability. Third row: shear forces have overcome the lateral expansion, pulling more flow of material uphill than can be offset by lateral expansion, leading to instability.
Figure 2.5. Comparison of heat maps for select swelling rates A. Black dashed lines show the level curves $Q \sin(\theta) = \pi, 3\pi, 5\pi$ for which the upper and lower interfaces are exactly out of phase.
Figure 2.6. A slice of the heat map at $\theta = 45$, illustrating the increasingly sensitive dependence on $Q$ as $A$ increases.

Figure 2.7. Transition diagram of the multi-branch structure near $Q \approx \frac{2\pi}{\sqrt{2}}$ on the out-of-phase level curve as $A$ is toggled.
Figure 2.8. A schematic illustrating the expected effect of swelling on the stability coefficient $C_{11}$, in a longwave linearization of the form $h_t \approx C_{11}(\theta) h_{xx}$. (a) Compared to existing models, which all exhibit a band of unstable angles above some transition angle, the swelling mechanism is stable for all angles of incidence. (b) As swelling is added at increasing ratios to the existing stability profile, the critical angle shifts to higher angles, and eventually may vanish entirely. (Here, “ratio” indicates the relative magnitude of the swelling mechanism compared to the existing profile, and the resulting sums are scaled by the value of $C_{11}(0)$).
3.1. Introduction

In the present work, we conclude an ongoing investigation [48, 50] into the potential role of ion-induced swelling in explaining the variety of observed phenomena. We develop a model in the so-called “hydrodynamic” class, which has recently shown promise in reaching agreement between experiment and theory [31, 58, 32]. This model combines the effects of both anisotropic plastic flow (hereafter referred to as APF) and isotropic ion-induced swelling (hereafter referred to as IIS), along with a proposed experimental method for estimating the relative strength of these two effects. The model also allows for an arbitrary depth-dependence in the intensity of each effect, (following other recent explorations [33, 49]), as well as an arbitrary positioning of the bottom boundary of the amorphous irradiated film. Our primary finding is to confirm that swelling can significantly increase the critical angle, and indeed suppress patterns entirely if present in sufficient amounts. We also argue heuristically that a “sufficient amount” of swelling to completely suppress patterns is likely well-aligned with parameter values expected of Ar$^+$-irradiated Ge. In the course of our analysis, we also find two important additional results: (a) that contrary to previous studies [33], and subject to certain assumptions which will be discussed, our model predicts that the critical angle will not depend on the uniaxial variation of the strength of each of the mechanisms (see Discussion and Appendix), and (b) that the choice of bottom boundary geometry has a surprisingly large effect on predicted behavior. This latter finding is consistent with a passing comment made elsewhere [33] and motivates more careful study of interfacial geometries as a means of reconciling experiment and theory.
3.2. Model

Figure 3.1. Schematic depicting ion bombardment at an incidence angle of $\theta$ and stress induced in the thin film by ion implantation. Note that for off-normal incidence, the bottom boundary $z = g$ may not vertically align with the top boundary $z = h$. The above shows the simplifying assumption that all spatial variation occurs along the “down-beam” direction, which is coupled to the interfacial geometry.

Our model for the ion-irradiated amorphous layer of an irradiated semiconductor is illustrated in Figure 3.1, and begins with the typical statements of conservation of mass

$$\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \vec{v}) = 0 \quad (3.1)$$

where $\rho$ denotes density, $\vec{v}$ denotes the bulk velocity field $[u,v,w]$; and conservation of momentum in the limit of low Reynolds number (high viscosity)

$$\nabla \cdot \mathbf{T} = 0 \quad (3.2)$$

where $\mathbf{T}$ denotes the stress tensor. We note that we have not made the common simplification to $\nabla \cdot \vec{v} = 0$, as ion-induced swelling causes a decrease in the film density over time.

**Anisotropic plastic flow (APF).** By *anisotropic plastic flow* (APF), we mean flow that has directionality (anisotropy) and which does not exhibit a stress response to the strain rate (plasticity). Typically, fluids behave according to the viscous stress tensor, which relates the amount of stress in the film to the rate at which the film deforms (“strains”). In contrast with other models which have suggested that the ion beam acts upon the film by exerting an
Figure 3.2. (a) Schematic depicting the mechanism of *anisotropic plastic flow* (APF), in which the beam causes material parcels to compress in the beam direction, and swell in the orthogonal direction(s) so as to preserve volume. (b) Schematic depicting the mechanism of *ion-induced swelling* (IIS), in which the beam causes material parcels to swell isotropically in all directions.

"effective body force" or adding to the stress tensor [31, 33, 49], the APF model posits that the action of repeated ion impacts is to modify the stress tensor so as to induce a greater strain rate for a given level of stress within the film than would otherwise occur. The nature of this extra, "stress-free" strain is to produce a volume-preserving shear, with compression in the downbeam direction and elongation in the cross-beam direction, as illustrated in Figure 3.2a. For this reason, the effect of anisotropic plastic flow is sometimes characterized as "pancake strain" [47]. Although the notion of anisotropic plastic flow was originally used in the context of high energy irradiation in the MeV range [61, 60, 36], with localized melting and re-freezing along ion-tracks as the mechanism underlying the effect, it was later observed in the keV range on a phenomenological basis, with the underlying mechanism currently unknown [37, 38, 39].

Despite the phenomenological nature of its use in the present (and other) work, the APF mechanism has produced surprisingly good agreement between theory and experiment in a variety of contexts. In the context of nanopatterning, it reconciles experimental and theoretical wavelength data [32] when coupled with experimental stress measurements for 250eV Ar\(^+\) → Si, and predicts the sign-change in the in-plane stress measured as a function
of angle [47]. In other contexts, it has led to good agreement in computational models of pore closure [59], the well-known “ion-hammering” effect [60, 61] in colloidal silica ellipsoids [37, 38, 80] in the MeV range, and other phenomena in the keV range [39]. Hence, its use appears to be well-supported by the literature, even if the exact physics from which the effect emerges is unknown.

To incorporate the APF mechanism into our model, we add a term to the typical definition of the viscous stress tensor, producing

$$\mathbf{T} = -p\mathbf{I} + 2\eta\{\dot{\mathbf{E}} - \dot{\mathbf{E}}_b\}. \quad (3.3)$$

We note that, in the above, as before, we have excluded the bulk viscosity. This is in line with other continuum models of ion-induced nanopattern formation [31, 32, 33, 48], but has a consequence of explicitly decoupling the anisotropic plastic flow and ion-induced isotropic swelling mechanisms. In reality, it is possible that the bulk viscosity must be included; however, not enough is known about the underlying mechanisms, and the present work should be considered as a plausible, exploratory model, rather than a proposed “final model”.

Here $p$ is the pressure, $\eta$ is the viscosity, and $\dot{\mathbf{E}}$ is the deviatoric component of strain, defined as usual via

$$\dot{\mathbf{E}} = \frac{1}{2} (\nabla \ddot{v} + \nabla \ddot{v}^T) \quad (3.4)$$

In addition, the effect of the incoming ions is modeled via the “beam-induced stress” $\dot{\mathbf{E}}_b$, defined by

$$\dot{\mathbf{E}}_b = f A_D \mathbf{D}(\theta) \tau(z; g, h, ... \right)$$

where

$$\mathbf{D}(\theta) = \begin{bmatrix}
\frac{3}{2} \cos(2\theta) & 0 & \frac{3}{2} \sin(2\theta) \\
0 & 1 & 0 \\
\frac{3}{2} \sin(2\theta) & 0 & -\frac{3}{2} \cos(2\theta) - \frac{1}{2}
\end{bmatrix}. \quad (3.5)$$

In the above, $f$ denotes the nominal flux of incoming ions in units $\text{ions} / \text{nm}^2 \cdot \text{s}$, $A_D$ is a mechanism-specific deformation rate, where the subscript $D$ denotes that it is the source of deviatoric strains. $\mathbf{D}(\theta)$ is a volume-preserving tensor describing compression in the beam direction and
expansion in the two orthogonal directions. Collectively, \( f_A D(\theta) \) characterizes the stress-free strain rate, anisotropically permitting more strain than would be possible in a typical Newtonian fluid. Finally, the “intensity field” or “spatial variation” function \( \tau(z; g, h, ...) \) allows for the possibility of a general treatment of the inhomogeneous strength of APF at a given point \((x,z)\) in the film. As an aside, we point out that if \( A_D \to 0 \), the above simply describes viscous relaxation of surface energy, as in [27].

**Ion-induced swelling (IIS).** The phenomenon of ion-induced swelling (IIS), in which radiation causes the volume of a parcel of material to increase over time (see Figure 3.2b), has been observed for a variety of materials (see, for instance, [70, 63, 72, 73, 65, 67, 64]). However, this mechanism has not typically been included in studies of ion-induced pattern formation. It was first suggested as a stabilizing mechanism in [48], and shown to be stabilizing at all angles of incidence in the limit of small swelling rates. That analysis was extended to arbitrary swelling rates in [50], where it was then suggested that swelling could potentially modify \( \theta_c \) in an ion-, target-, and energy- specific manner. This has served as the motivation for the present work.

To include the IIS mechanism in our model, we introduce an equation of state, which we take to have the simplified “quasi-incompressible” form

\[
\rho(\Delta) = \frac{\rho^*}{1 + \Delta}.
\] (3.6)

This is a generalization of the “quasi-incompressibility” equation of state used in Chapter 2. Here, the density \( \rho \) throughout the film is postulated to begin with the value \( \rho^* \) of freshly-amorphized material, and remain independent of pressure. However, it varies over time according to a quantity \( \Delta \) characterizing the amount of “volumization” sustained by a parcel of matter as it experiences ion-induced swelling. To track this quantity, we use a simple forced advection equation of the form

\[
\frac{\partial \Delta}{\partial t} + \vec{v} \cdot \nabla \Delta = \alpha(z; g, h, ...),
\] (3.7)

in which moving material parcels acquire additional volume at the rate \( \alpha(z; g, h, ...) \), which
has units $\frac{1}{s}$. Like the spatial variation function $\tau(z; g, h, ...)$, we allow for the possibility that the swelling rate is non-uniform spatially, and $\alpha(z; g, h, ...)$ simply assigns an intensity of swelling to a given parcel of matter at the point $(x,z)$, where $x$ dependence is imparted only implicitly via $(g, h, ...)$. Elsewhere [48, 50], a special case of this model (with $\alpha \equiv 1$) has been shown to increase the linear stability of thin-film surfaces against topographical perturbations, increasing the critical angle $\theta_c$.

A note on flux-dependence for stress. In the present work, we have used $fA_D$ as a coefficient of the APF-term, borrowing from the notation of [39, 41, 44, 32]. As a coefficient of the IIS-term, we have used $fA_I$, notation similar to that of [48, 50]. Both quantities have natural units of $\frac{1}{s}$, being, fundamentally, rates. For simplicity and following common practice, we have here assumed both rates to be linear in the ion flux. However, we note that a nonlinear flux dependence has been useful in explaining certain stress-related phenomena [46].

Boundary conditions. At the free upper interface, $z = h$, we have

\[
\begin{align*}
v_{I,h} &= \vec{v} \cdot \hat{n} - V \frac{\rho^*}{\rho} \\
\mathbf{T} \cdot \hat{n} &= -\gamma \kappa \hat{n}
\end{align*}
\]  

(3.8)

where $[\ ]$ denotes the jump across the material interface. The first is a modified kinematic condition due to mass conservation at the upper interface, which takes into account the removal of material due to erosion. For details of the derivation, see [48]. The second equation is simply a statement of stress balance across the free interface (or, equivalently, Newton’s second law across an interface for a vanishingly-small volume). At the lower, amorphous-crystalline interface, $z = g$, we have

\[
\begin{align*}
\Delta &= 0 \\
\vec{v} \cdot \hat{t} &= 0, \\
\vec{v} \cdot \hat{n} &= 0
\end{align*}
\]  

(3.9)
and these latter two equations are simply the imposition of no-slip and no-penetration conditions at the amorphous-crystalline boundary, which reduce identically to

\[ u = v = w = 0 \] (3.10)

at \( z = g \). It is intuitive that \( \Delta = 0 \) should be the case because as the film erodes downward, the material at the crystalline-amorphous interface should always be the newest, hence having sustained no instantaneous damage (yet) at any moment.

### 3.3. Analysis

Here, we very briefly review the main calculations of the present work before presenting the primary theoretical result; the calculations are lengthy and do not, in themselves, contribute to the discussion. For full details of the asymptotic expansions and the systems of ordinary differential equations solved, we refer the reader to the Appendix.

**Steady state in a traveling frame.** We begin by converting all equations to a moving frame traveling downward with the eroding surface at steady-state erosion velocity \( V \); mathematically,

\[
\begin{align*}
    z &\rightarrow z - Vt \\
    h &\rightarrow h - Vt \\
    g &\rightarrow g - Vt \\
    v &\rightarrow \hat{v} - \hat{V} \hat{k} \\
    v_{I,h} &\rightarrow v_{I,h} - V(\hat{k} \cdot \hat{n})
\end{align*}
\] (3.11)

This results in equations which are equivalent to the above, except for \( w = V \) instead of \( w = 0 \) at \( z = g \). We then look for steady-state solutions \( \vec{\Phi}_0(z) \) for each of the bulk fields \( \vec{\Phi} = [\rho, u, w, p, \Delta] \) under the assumptions that \( g = 0, \ h = h_0, \ \frac{\partial}{\partial t} \rightarrow 0, \) and \( \frac{\partial}{\partial x} \rightarrow 0 \). This steady-state describes an eroding surface that has not been subject to any perturbations in its surface geometry.
Linear stability in normal modes. To explore the stability of the steady state just obtained to small perturbations in the film geometry, we next construct a linearization of the governing equations about these steady-state solutions in each of the bulk fields $\vec{\Phi} = [\rho, u, w, p, \Delta]$ and the two interfaces $g$ and $h$ as

$$
\begin{align*}
h(x,t) &\to h_0 + \epsilon h_1(x,t) \\
g(x,t) &\to g_0 + \epsilon g_1(x,t) \\
\vec{\Phi}(x,z,t) &\to \vec{\Phi}_0(z) + \epsilon \vec{\Phi}_1(x,z,t)
\end{align*}
$$

(3.12)

where we employ the common notation of subscript 0 denoting the steady-state part and subscript 1 denoting perturbative part. We have stripped out $y$-dependence (hence the bulk field $v$) in order to focus on the equations describing the evolution of the system in the projected downbeam direction. After collecting powers of $\epsilon$, we obtain the linearized, $O(\epsilon)$ equations. To these equations, we look for solutions $\vec{\Phi}_1 = [\rho_1, u_1, v_1, w_1, p_1, \Delta_1]$ in normal modes, using the ansatzes

$$
\begin{align*}
\vec{\Phi}_1(x,z,t) &\to \vec{\Phi}_1(z) \exp(\sigma t + ikx) \\
g_1(x,t) &\to \tilde{g}_1 \exp(\sigma t + ik(x-x_0(\theta))) \\
h_1(x,t) &\to \tilde{h}_1 \exp(\sigma t + ikx),
\end{align*}
$$

(3.13)

where $x_0(\theta)$ is a horizontal shift; we will comment on this below. The application of these ansatzes causes $\frac{\partial}{\partial t} \to \sigma$ and $\frac{\partial}{\partial x} \to ik$ everywhere in the governing equations, which greatly simplifies the analysis and converts the system in the leading-order correction terms ($O(\epsilon)$) into a system of boundary-value problems in independent variable $z$, the film depth in laboratory coordinates (the familiar Cartesian $x$-$z$). Solving this system yields $\sigma(k)$, the dispersion relation, as a compatibility requirement for the linearized kinematic condition at the upper interface, from which we may obtain theoretical predictions on patterning.

Simplifying assumptions. The resulting system, though linear, remains too difficult to solve analytically in closed form. Consequently, we make several additional simplifying assumptions.
Because we are interested primarily in the critical angle $\theta_c$ from stability to instability, and because that transition is observed to occur at small wavenumbers $k$ (i.e. in the long-wave limit [6]), we will make the formal assumption that $kh_0 \ll 1$, and perform Taylor expansions in this quantity.

Because we have previously shown that the qualitative behavior of isotropic swelling does not depend strongly on the speed at which it occurs [50], we will formally assume that the dimensionless parameter $\frac{f_A h_0}{v}$, which measures total volumization of a parcel by the time it is sputtered, is much less than 1. This assumption, also used previously in this series [48, 50], seems consistent with the behavior of Silicon, which is not known to exhibit meaningful amounts of swelling during low-energy irradiation. (See further discussion in the Appendix.)

We will assume that the related dimensionless parameter $\frac{f_A p h_0}{V}$ associated with anisotropic plastic flow is also small, in the sense that products of the form $\frac{f_A p h_0 f_A h_0}{V}$ can be neglected. This is based on parameter estimates of $A_D$ due to [59, 44, 28].

These simplifications allow us to obtain otherwise highly-general solutions with all fields $\rho, \vec{v}, p, \Delta$ expressed in terms of the as-yet unassigned spatial variation functions, interface relation, and the arbitrary components of the stress tensor $D_{ij}$. This one-time calculation then provides a highly general stability result in the form of a functional, or a “function of functions”, which accepts as its arguments functions rather than numbers and returns a scalar quantity. Hence we report $\sigma = \sigma(\tau, \alpha, D_{ij}, \frac{\rho}{h_1}, k)$.

**Dispersion relation.** In the limits just described, we obtain

$$\sigma(k) \approx 0 + [f_A D \sigma_{1D}(k) + f_A I \sigma_{1I}(k)] ik + [f_A D \sigma_{2D}(k) + f_A I \sigma_{2I}(k)] k^2 + \ldots, \quad (3.14)$$

where

$$\sigma_{1D} = -\frac{2D_{13}}{h_1} \left[ \int_0^{h_0} \int_0^{z_1} \tau_\epsilon(z_2) dz_2 dz_1 - \tau_0(0) \tilde{g}_1 h_0 + \tilde{h}_1 \int_0^{h_0} \tau_0(z) dz \right]; \quad (3.15)$$

46
\[ \sigma_{1I} = 0; \quad (3.16) \]

\[ \sigma_{2D} = \frac{2(D_{11} - D_{33})}{h_1} \int_{0}^{h_0} \left[ \int_{0}^{z_1} \int_{0}^{z_2} \tau_\epsilon(z_2) dz_2 dz_1 - z \left[ \tau_0(h_0) \tilde{h}_1 + \int_{0}^{h_0} \tau_\epsilon(z) dz \right] \right] dz \quad (3.17) \]

and

\[ \sigma_{2I} = \frac{1}{h_1} \int_{0}^{h_0} \left[ \int_{0}^{z_1} \int_{0}^{z_2} \tau_\epsilon_1(z_2) dz_2 dz_1 + z \left[ \alpha_0(h_0) \tilde{g}_1 - 2 \alpha_0(h_0) \tilde{h}_1 - 2 \int_{0}^{h_0} \alpha_1(z) dz \right] \right], \quad (3.18) \]

which we will use throughout the rest of the present work. In the above, \( \alpha_\epsilon, \tau_\epsilon \) are the perturbative, \( O(\epsilon) \) parts of whatever spatial distribution that IIS and APF (respectively) are hypothesized to have. Likewise, \( \alpha_0, \tau_0 \) are the leading order, \( O(1) \) parts which occur near the steady state—that is, in the case of unpatterned, flat interfaces for both \( g \) and \( h \).

As we will see, the linearity in coefficients \( fA_I \) and \( fA_D \) that emerges as a result of this limit simplifies the analysis substantially and is sufficient for the present work.

Simplified, we can express the dispersion relation as

\[ \sigma(k) \approx 0 + \left[ fA_D \sigma_{1D} + fA_I \sigma_{1I} \right] i k h_0 + \left[ fA_D \sigma_{2D} + fA_I \sigma_{2I} \right] (k h_0)^2 + \ldots, \quad (3.19) \]

where

\[ \sigma_{1D} = -3 \sin(2\theta) \left\{ \left< \tau_0(z) + \int_{0}^{z} \frac{\tau_\epsilon(\zeta)}{h_1} d\zeta \right> - \tau_0(0) \frac{\tilde{g}_1}{h_1} \right\} \quad (3.20) \]

\[ \sigma_{1I} = 0 \quad (3.21) \]

\[ \sigma_{2D} = -6 \cos(2\theta) \left\{ \tau_0(h_0) + h_0 \left< \frac{\tau_\epsilon(z)}{h_1} - \frac{1}{2} \int_{0}^{z} \frac{\tau_\epsilon(\zeta)}{h_1} \frac{d\zeta}{h_0} \right> \right\} \quad (3.22) \]

\[ \sigma_{2I} = \left\{ \alpha_0(h_0) + h_0 \left< \frac{\alpha_\epsilon(z)}{h_1} - \frac{1}{2} \int_{0}^{z} \frac{\alpha_\epsilon(\zeta)}{h_1} \frac{d\zeta}{h_0} \right> \right\} + \frac{1}{2} \alpha_0(0) \frac{\tilde{g}_1}{h_1} \quad (3.23) \]

where \( \langle \cdot \rangle \) denote averages over the domain \( z \in [0, h_0] \).
Figure 3.3. (a) Schematic depicting a bottom surface that - for mathematical simplicity - is displaced vertically from the top interface regardless of ion incidence angle. (b) Schematic depicting a bottom surface that is displaced from the top surface in a direction that rotates with the ion beam.

The interface relation. Using the expressions above, we may study the value \( \theta_c \) for which \( \sigma(k; \theta) \) transitions from negative for all \( k \) (i.e., the surface is stable to perturbations) to positive for at least some \( k \) (i.e., the surface is unstable to perturbations) for given spatial variation functions \( \tau(z; g, h, ...) \), \( \alpha(z; g, h, ...) \) and interface relation \( \frac{\partial z}{\partial h} \), which together can characterize a mathematically-general treatment of an ion-irradiated amorphous thin film. We take a moment to characterize this last quantity, which has seen little explicit treatment to date [48, 50], while being implicitly treated (by necessity) in all stress-based formulations of irradiated thin films [31, 58, 32, 48, 50].

As a thin-film hydrodynamic stability problem, any analysis requires some notion of the lower interface. Elsewhere, various assumptions about the relationship between the upper interface and lower interface have been explored, including a flat lower interface [31], a lower interface that is a vertical translation of the upper interface [32, 69] (as depicted in Figure 3.3a), and a lower interface that is translated diagonally in the ion beam direction [33, 48, 50] (as depicted in Figure 3.3b). Physically, these interface relations are rooted in the understanding that the ion-beam permits a certain amorphization thickness and, as material is sputtered away, this exposes more material to amorphization along the downbeam direction.
This dependence naturally appears in the linearization of the interfaces and is a fundamental quantity in the physical description of the system. We have therefore left the expressions for the perturbations to the interfaces, \( \tilde{g}_1 \) and \( \tilde{h}_1 \), arbitrary in the calculations in the Appendix. Notice that these terms only ever occur in the ratio \( \frac{\tilde{g}_1}{\tilde{h}_1} \) within the dispersion relation. This quantity, then, imparts upon the system the complex dependence between the free interface and the amorphous-crystalline interface: at any given time, the free interface determines the amorphous-crystalline interface by directly modifying power deposition in the bulk, while the amorphous-crystalline interface will be shown in the present work to substantially alter the growth rate of that free interface. Supposing that we had the expressions

\[
\tilde{h}_1 = \exp(\sigma t + ikx) \\
\tilde{g}_1 = \exp(\sigma t + ik(x - x_0(\theta)))
\]

in the original linearization, so that the lower interface is a copy of the upper interface shifted laterally by \( x_0 \) and displaced vertically by \( h_0 \) (due to Equation (4.8)), we then have

\[
\frac{\tilde{g}_1}{\tilde{h}_1} = \exp(-ikx_0),
\]

which appears as a parameter in the \( \mathcal{O}(\epsilon) \) equations. After evaluating Equation (3.19) with the ratio replaced by the right-hand side of the above, we may expand the above in \( k \approx 0 \), collect terms, and finally obtain a dispersion relation associated with the desired interface relation. Since \( x_0 \) is the horizontal shift, taking \( x_0 = 0, h_0 = \text{constant} \) imparts the vertical-translation interface relation upon the system. Likewise, \( x_0(\theta) = h_0(0) \sin(\theta), h_0(\theta) = h_0(0) \cos(\theta) \) imparts the diagonal-translation interface relation, so-called because of the presence of a vertical component and a horizontal component, by which hills on the upper and amorphous-crystalline interface are permanently fixed about a diagonal. Finally, \( \tilde{g}_1 = 0 \), hence the elimination of all \( \frac{\tilde{g}_1}{\tilde{h}_1} \) terms, imparts the flat lower interface assumption, which will not be studied in the present work.
3.4. Results

3.4.1. Theoretical results

After having computed the leading-order real term of the long-wave linear dispersion relation, we determine the beam-angle $\theta_c$ for which the real part changes sign from negative to positive, hence topographical perturbations destabilize the free interface and we may expect pattern formation. We consider the specialization of the dispersion relations stated in Equations (3.15)-(3.18) to each of two cases that have been previously studied.

**Result 1: Vertically-displaced interfaces; stress modification vertical.** One idealized treatment of the lower interface, explored in [32], models the lower amorphous-crystalline interface as a vertically-translated copy of the upper, free interface, regardless of the angle of incidence (see Figure 3.3a) — that is, $g(x,t) = h(x,t) - h_0$. Under such a simplification, we obtain

$$h_0(\theta) = h_0, \quad x_0(\theta) = 0, \quad \frac{\bar{g}_1}{h_1} = 1$$

(3.26)

To incorporate depth-dependent stress fields in a manner consistent with this geometry, we suppose that variation in the stress field also occurs along the vertical axis. As shown in the Appendix, we can describe such variation in a general way using Legendre polynomials:

$$\tau(z; g, h) = 1 + \sum_{k=1}^N \beta_{k,D} P_k \left( \frac{2z - (g + h)}{(g - h)} \right)$$

$$\alpha_1(z; g, h) = 1 + \sum_{k=1}^N \beta_{k,I} P_k \left( \frac{2z - (g + h)}{(g - h)} \right)$$

(3.27)

where the argument of $P_k$ serves to map the domain $z \in [g, h]$ to the Legendre polynomial’s “native” domain [-1,1]. The resulting dispersion relation takes the form

$$\text{Re}(\sigma) = - \left( (3 + \beta_{1,D}) f A_D \cos(2\theta) + (3 + \beta_{1,I}) \frac{f A_I}{6} \right) (kh_0)^2 + O((kh_0)^4)$$

(3.28)

Surprisingly, it is clear that the effect of spatial variation factors out completely, with $\theta_c$ determined fully by the ratio $(3 + \beta_{1,I}) A_I / (3 + \beta_{1,D}) A_D$. We note that even though we have used a second-order Legendre polynomial about the Cartesian $z$ axis, the dependence on $\beta_2$, the coefficient
of the quadratic term, has completely disappeared. Seeking the incidence angle $\theta_c$ at which this expression changes sign, we find a closed-form solution

$$\theta_c = \frac{1}{2} \arccos \left( \frac{-(3 + \beta_{1,I}) A_I}{6(3 + \beta_{1,D}) A_D} \right),$$

(3.29)

and, again, the only factor seemingly capable of determining $\theta_c$ is the ratio of mean strengths of the bulk stress-modifying mechanisms.

**Result 2: Diagonally-displaced interfaces; stress modification downbeam.** An alternative treatment of the lower interface, considered in [33, 48, 50] supposes that it is a diagonally-translated copy of the free interface in the direction of the incoming ions (see Figure 3.3b) — that is, $g(x, t) = h(x - h_0' \sin(\theta), t) - h_0' \cos(\theta)$. Under this approximation one obtains

$$h_0(\theta) = h_0' \cos(\theta), \quad x_0(\theta) = h_0' \sin(\theta), \quad \to \frac{\bar{g}_1}{h_1} = \exp(-ikx_0(\theta)),$$

(3.30)

To describe depth-dependent stress fields under this geometry, we suppose that variations occur along the direction of incoming ions. As shown in the Appendix, we can explore these generically using Legendre polynomials defined along a new coordinate $\tilde{z}$ oriented along a single axis, via

$$\tau(z; g, h, h_x) = 1 + \sum_{k=1}^{N} \beta_{k,D} P_k \left( \frac{2\tilde{z} - h_0'}{h_0'} \right),$$

$$\alpha_1(z; g, h, h_x) = 1 + \sum_{k=1}^{N} \beta_{k,I} P_k \left( \frac{2\tilde{z} - h_0'}{h_0'} \right),$$

(3.31)

where

$$\tilde{z} \approx \frac{(h(x) - z)}{\cos(\theta) + h_x \sin(\theta)}. $$

(3.32)

To clarify, the $h_x$ in the above is the x-derivative of the free interface $h$. Under this treatment of the amorphous film, we obtain the dispersion relation

$$\sigma = - \left( f A_D (3 + \beta_{1,D})(-1 + 2 \cos(2\theta)) + \frac{f A_I (3 + \beta_{1,I})}{6} \right) (kh_0' \cos(\theta))^2 + O((kh_0' \cos(\theta))^4)$$

(3.33)
Again, we see that depth-dependence factors out cleanly, and, in the case that IIS and APF have different spatial distributions, the only quantity capable of shifting the selected bifurcation angle is the ratio of $A_I$ to $A_D$. We compute the critical angle $\theta_c$,

$$\theta_c = \frac{1}{2} \arccos \left( \frac{1}{2} \left[ 1 - \frac{(3 + \beta_{1,I})A_I}{6(3 + \beta_{1,D})A_D} \right] \right).$$

(3.34)

Figure 3.4. Critical angle selection as a function of the ratio of IIS strength to APF strength and the description of the interfacial geometry. “Vertical displacements” correspond to the interface assumption in Equation (3.26) and “diagonal displacements” correspond to Equation (3.30).

Initial observations. The results for both vertically- and diagonally-displaced bottom boundaries are shown in Figure 3.4. In both cases, we find that the stabilizing effect of ion-induced swelling can significantly change the bifurcation angle, and if present in sufficient amounts, suppress pattern formation entirely. This finding seems particularly relevant to understanding experimental results on germanium, which exhibits significant swelling during irradiation [63, 64], and also exhibits significantly higher critical angles than silicon [57]. Interestingly, we find that depth-dependence of APF and IIS (appearing by way of the
Legendre polynomial coefficients $\beta_k$ merely modifies the dependence of the critical angle on the ratio $A_I/A_D$. Moreover, and in contrast to a previous investigation [33], the depth dependence cleanly factors out of the expression for $\sigma(k)$ - and therefore does not appear in the expression for $\theta_c$ - if either $A_I = 0$ (as was the case in that work), or if $\beta_{1,I} = \beta_{1,D}$. (We note that the coefficients $\beta_k$ may be relevant to e.g. the selection of wavelength $\lambda$, but we have not considered $\lambda$-selection here, partially due to the restriction of the present work to the long-wave limit wherein the estimation of $\lambda$ may give significantly different predictions than the full-spectrum prediction [32].)

Figure 3.4 plots the critical angle as a function of the expression $(3+\beta_{1,I})A_I/(3+\beta_{1,D})A_D$; hence, the effect of any depth-dependence of APF and IIS can be explored by examining this expression in more detail. We see that it increases with increasing $\beta_{I,1}$, and decreases with increasing $\beta_{D,1}$. Because an increase in this expression corresponds to higher critical angle, and hence greater stability, we conclude that

- the (destabilizing) effect of APF is enhanced when it occurs deeper in the film
- the (stabilizing) effect of IIS is enhanced when it occurs deeper in the film

Because we expect the coefficient $\beta_{k,I} \in [-1,1]$ to preserve a positive magnitude of these mechanisms throughout the film, we see that spatial variation of one or the other mechanism could change the effective relative strength of swelling by 33-50%.

Finally, and remarkably, we see that the choice of interface relation is, itself, a major contributor to $\theta_c$ selection. This is unexpected and alarming, as all existing analyses of hydrodynamic stability for ion-irradiated thin films have considered only one interface relation each, selecting the treatment of the interfaces primarily on a basis of mathematical simplicity, and drawing conclusions from the analysis that follows. We therefore consider the amorphous-crystalline interface a prime candidate for modeling refinement, as both the “vertical” and “diagonal” cases considered here are idealizations of the true interface relation, where there is likely to be some lateral displacement (hence not “vertical”, see Equation (3.26)), while the film thickness with respect to the $z$ coordinate does not vanish at grazing
incidence (hence not “diagonal”, see Equation (3.30)), as has been noted in [20, 28].

3.4.2. An approach for parameter estimation

The preceding analysis has shown the dependence of $\theta_c$, in our idealized case, on the ratio $\frac{A_I}{A_D}$. Because these parameters are so important in predicting the critical angle, and because the critical angle can vary so widely in their ratio, it is worth considering how they might be estimated from experimental data. We will here consider one approach by which this might be accomplished, using a data set available for the Ar$^+ \rightarrow$ Si system. We note that we do not necessarily anticipate swelling to have a strong effect in that system, but it is the only one for which the needed data exists [47], so it must serve as way to illustrate the approach we propose.

Idea: angular dependence of steady stress. We begin by noting that one step of the analysis we have performed is the identification of steady-state values of all bulk fields $\Phi_0(z) = [\vec{v}, \rho, p, \Delta]$. From these steady-state fields, we can infer a steady-state stress $T_0$ using Equations (3.3)-(3.5); as shown in the Appendix, the in-plane component of this tensor has the form

$$T_{01} = -2fA_D\eta\tau_0(z; 0, h_0)(D_{11} - D_{33}) - 2fA_I\eta\alpha_{01}(z; 0, h_0)$$

$$= -6fA_D\eta\cos(2\theta)\tau_0(z; 0, h_0) - 2fA_I\eta\alpha_{01}(z; 0, h_0).$$

We note that there are two components to this value, one associated with APF, and one associated with IIS. Importantly, the former has a dependence on the incidence angle of the form $\cos(2\theta)$, while the latter is constant for all incidence angles. If this expression were compared to measurements of the in-plane stress over multiple angles of incidence, then best-fit values of both $fA_D\eta$ and $fA_I\eta$ could be obtained.

An example data set: 250eV Ar$^+$ $\rightarrow$ Si. A data set of precisely this nature exists for the 250eV Ar$^+$ $\rightarrow$ Si system at a flux of $12 \text{ ions/mm}^2 \cdot \text{s}$, obtained by Perkinson [47]. This study performed wafer-curvature measurements of Si samples irradiated at varying angles of incidence $\theta \in [0, 20, 40, 60, 70, 80]$. For all angles, the time-series data appeared to exhibit two
peaks in the in-plane stress over time, referred to as “Peak A” and “Peak B” (see Figure 6.9 of [47]). The study’s authors suggested that the first peak at early times is due to transient processes such as amorphization and/or the removal of oxides, whereas the second peak at later times is representative of continuous processes such as APF and IIS. In contrast to some earlier studies [44], the inferred stresses did not appear to saturate, instead gradually becoming more tensile over time. Consequently, identifying a true “steady” value from these data is difficult. Nevertheless, for the sake of illustrating the approach, we will treat this study’s values for “Peak B” as steady-state values to be compared with Equation (3.35).

**Adjustment of angle-dependent film thickness.** In [47], the measured values of wafer curvature are converted into thin-film stresses via the Stoney equation

\[
\sigma_{\text{avg}} = \frac{\Delta K M_s h_s^2}{6 h_0},
\]  

(3.36)

where \(\sigma_{\text{avg}}\) is the average in-plane stress, \(\Delta K\) is the change in wafer curvature, \(M_s\) is the biaxial modulus of the substrate, \(h_s\) is the thickness of the substrate, and \(h_0\) is the thickness of the amorphous layer (in the notation of the present work). This naturally requires an estimate for the thickness of the stressed, amorphous film \((h_0)\). As a simplifying assumption, the authors used a value of \(h_0(\theta) = 3\text{nm}\) for all angles of incidence, consistent with the approximation of a “vertically-translated” bottom boundary described above. Alternatively, the approximation of a “diagonally-translated” lower interface would yield an alternate definition of the form \(h_0(\theta) = 3\cos(\theta)\text{nm}\). Instead of either of these, and motivated by discussion in [28], we instead directly compute \(h_0\) at each incidence angle from SRIM by taking the mean ion depth plus twice longitudinal straggle, as has been discussed elsewhere as an approximation [14]. This requires converting the reported peak stresses from [47] back to curvatures using \(h_0 = 3\text{nm}\), and then re-converting to stresses using the experimental values \(h_0(0^\circ) \approx 3.2\text{ nm}, h_0(20^\circ) \approx 3.12\text{ nm}, h_0(40^\circ) \approx 2.86\text{ nm}, h_0(60^\circ) \approx 2.45\text{ nm}, h_0(70^\circ) = 2.20\text{ nm}, h_0(80^\circ) = 1.91\text{ nm}\). The resulting stresses are shown in Figure 3.5.
Model fitting and parameter estimation. We now use the theoretical steady-state mean in-plane stress

\[ < T_{01}^{11} > = \frac{1}{h_0} \int_{h_0}^{h_0} T_{01}^{11} d\bar{z} \]  

(3.37)

for comparison with experimental data [47] of wafer curvature measurements. Because the spatial variation functions \( \alpha(z) \) and \( \tau(z) \) consist of Legendre polynomials with leading coefficient of 1, we have

\[ < \alpha(z) >= < \tau(z) >= 1, \]  

(3.38)

for all \( \beta_1, \beta_2 \); therefore the steady stress is simply

\[ < T_{01}^{11} > = -2 f_A D \eta (D_{11} - D_{33}) - 2 f_A I \eta = -6 f_A D \eta \cos(2\theta) - 2 f_A I \eta. \]  

(3.39)

We then perform a least-squares fit of this equation to the stress data using the SciPy package optimize.curve_fit and uncertainties. A comparison of the data to the fitted model is shown in Figure 3.5, and produces fitted values of

\[ f_A D \eta \approx 0.0786 \pm 0.008 \text{ GPa}, \]  

(3.40)

\[ f_A I \eta \approx 0.0286 \pm 0.0178 \text{ GPa}. \]

Implications for \( \theta_c \) in Si and Ge. The fitted values for the 250eV Ar\(^+\) → Si system suggest a ratio of \( \frac{A_I}{A_D} \approx .364 \). Comparing this value with Figure 3.4, and consistent with our expectations, we expect that swelling will not strongly modify \( \theta_c \) for this system – again it was chosen merely to illustrate a methodology rather than to explain a phenomenon. Much more interesting is the case of irradiated germanium, which is known to experience significant swelling upon irradiation, and also seems to exhibit significantly higher values of \( \theta_c \) (see, e.g. [57] and [33]).

Although we have no wafer-curvature data from which to infer the ratio \( \frac{A_I}{A_D} \) for a low-energy irradiated Ge system, the parameter estimates for 250eV Ar\(^+\) → Si may be informative. Ge is known to be capable of losing half of its density or more when irradiated by by various projectiles and energies [63, 70, 71, 64, 76]. This suggests \( \left( \frac{f_A I h_0}{V} \right)_{\text{Ge}} \approx 1 \), whereas
the corresponding value \((\frac{f_A \Delta h}{V})_{Si}\) for silicon is most likely only a few percent [72, 65, 67]. At the same time, we do not expect that \((\frac{f_A \Delta h}{V})_{Ge}\) and \((\frac{f_A \Delta h}{V})_{Si}\) should differ radically for the same projectile and energy, as the “ion-hammering” effect associated with \(fA_D\) is believed to scale either with momentum transfer [81, 82] or electronic stopping [39, 62]; if either of these are approximately true, the proposition that \((\frac{fA \Delta h}{V})_{Ge}\) \(\approx\) \((\frac{fA \Delta h}{V})_{Si}\) for the same projectile and energy is defensible.

For \((\frac{fA \Delta h}{V})_{Ge}\) \(\approx\) \((\frac{fA \Delta h}{V})_{Si}\), and supposing plausible values \((\frac{fA \Delta h}{V})_{Si}\) \(\approx\) .02 and \((\frac{fA \Delta h}{V})_{Gi}\) \(\approx\) 1.0, this would lead to \(\frac{A_I}{A_D}\) \(\approx\) 17, which, from Figure 3.4, is of approximately the right magnitude to completely suppress pattern formation as has been seen in [57] for Ne\(^+\),Ar\(^+\) \(\rightarrow\) Ge — but not so overwhelmingly that it could not be overcome by an increase in \(A_D\) by a factor of two, such as one might see as a result of using Xe\(^+\) instead of Ar\(^+\) if \(A_D\) turns out to scale approximately with momentum. Intriguingly, to this would lead to \(\frac{A_I}{A_D}\) \(\approx\) 8, which

Figure 3.5. Fitting the form of Equation (3.39) to the Peak B data of [47] after adjusting angle-dependent film-thickness \(h_0\). **Blue:** experimental data set. **Red:** the theoretical magnitude of \(T_{011}\), the steady-state in-plane component of stress, when fit to the data. Because \(< T_{011} >\) has only two free parameters while the data set consists of three observations at each of six different incidence angles, we may perform parameter-fitting from the data.
would evidently return the system to a state where patterns could form, consistent with [57]. Although speculative (for lack of experimental data), we present this line of reasoning as a plausibility argument: for a system which is known to form patterns and not exhibit much swelling, the present analysis and parameter estimation correctly suggest that patterns should not be strongly suppressed, while for a system known to not form patterns, the parameter values suggested above imply that patterns must be strongly suppressed. Then, a parameter adjustment such as one might expect under a plausible scaling of $A_D$ with momentum correctly suggests that we might observe patterns again, albeit only at high angles. Hence the proposed IIS mechanism appears capable of capturing at least some of the trends identified elsewhere [57, 50].

**Formal expansion validated.** In the Analysis section, we described a formal expansion in small parameter $\frac{f_{A_l} h_0}{V}$, i.e., the limit where a parcel of matter experiences little swelling before being sputtered away from the upper interface, and a corresponding assumption of small product $\frac{f_{A_l} h_0}{V} \frac{f_{A_D} h_0}{V}$. For our estimated ratio $\frac{A_l}{A_D} \approx 0.364$, and a supposed value $\frac{f_{A_l} h_0}{V} \approx 0.02$ for Si, we would obtain $\frac{f_{A_l} h_0}{V} \frac{f_{A_D} h_0}{V} \approx 0.001$, which indeed justifies the neglect of cross terms.

### 3.5. Discussion and conclusions

3.5.1. Primary results

**1: On swelling as a contributor to non-universality of $\theta_c$.** Previous work by our group has shown that ion-induced swelling stabilizes an irradiated thin film against a wide range of perturbative wavenumbers [48, 50], in particular the long-wave perturbations responsible for the Type II bifurcation leading to pattern formation in Group IV semiconductors [6]. The present work has shown that this effect is, furthermore, capable of completely stabilizing the surface in materials that exhibit large amounts of swelling, such as germanium. That this occurs not only for enough (or arbitrarily large) swelling but for plausible amounts of swelling lends weight to the hypothesis that ion-induced swelling may be an important
mechanism in the stabilization of irradiated Ge. We note that these parameter values, while not yet measured, may be inferred on an order-of-magnitude basis from the work of [63, 70, 64] and others, and are nonetheless likely to be experimentally accessible using the technique that we have described in the main text. Further experimental inquiry in this direction would be informative.

It is noteworthy that IIS is capable of introducing ion-, target- and energy dependence, all of which are seen to modify $\theta_c$ experimentally. As we have discussed in “Implications for $\theta_c$ in Si and Ge” above, there can be significant differences in the tendency of different substrates to swell, e.g., between Si and Ge. However, even within the same target species, the atomic mass of the projectile species and the beam energy may directly modify the formation of Frenkel pairs in the amorphous bulk. We have modeled IIS in a “mechanism-agnostic” manner, without any particular underlying mechanism in mind. At the same time, it is striking that such a mechanism, if ultimately driven by the reaction, advection, and diffusion of defects across all materials as we suspect to be the case for irradiated Ge, would immediately introduce ion-, target- and energy dependence. The nature and extent of these dependencies will be the subject of future work, and it is possible that the functional dependencies of IIS and APF on ion, target and energy are extremely nonlinear, leading to the “islands of stability” seen for $\text{Ar}^+ \rightarrow \text{Si}$ for certain energies [22].

2: Experimental parameter estimation. As a basis for a tentative comparison between theory and experiment, we have attempted to estimate the values of two parameters from a small wafer-curvature data set from experiments involving 250eV irradiation of Si by Ar$^+$ by fitting the angle-dependent data to the component of our steady-state stress tensor associated with in-plane stress. Reasonable, qualitative agreement was found between the data and the theoretical in-plane stress model, and parameter estimates for the 250eV Ar$^+ \rightarrow \text{Si}$ system appear to be quantitatively similar to those from elsewhere in the literature. In particular, we estimate $f_{AD} \approx 5.24 \times 10^{-4} \frac{1}{s}$ for this system as compared with $f_{AD} \approx 3 \times 10^{-4} \frac{1}{s}$ for the 1keV Ar$^+ \rightarrow \text{Si}$ system estimated elsewhere [28]. Our estimate for the value of $\frac{f_{AB}h_0}{V}$,
the percentage volumization of a parcel of matter by the time it is sputtered from the upper interface, is consistent with the expectation that irradiated Si should not exhibit large amounts of swelling.

Nonetheless, approximation by the steady-state stress is imperfect, and the data set was small. Further angle-dependent wafer curvature measurements leading to separation of isotropic and anisotropic stress components for various materials would be of great benefit in connecting theoretical and experimental results, especially across a variety of energies, projectiles and targets. Of particular interest would be angle-dependent stress data for the irradiation of Si and Ge by Ar$^+$, Kr$^+$, and Xe$^+$ in the 250eV to 2keV range, with data from irradiated Ge systems seemingly supplying the first known experimental measurement for our $fA_I$, the ion-induced swelling rate. Evidence of large, angle-independent compressive stresses in irradiated Ge would be consistent with our hypothesis that the swelling mechanism may be a dominant stabilizing mechanism in that system.

While we have suggested one way that $A_I$ and $A_D$ may be estimated, we do not mean to imply that this is the only way to obtain the relevant parameter values. In particular, $A_D$ has been estimated elsewhere by fitting a full-spectrum composite model to GISAXS data [28], and by normal-incidence wafer curvature measurements [44]. A somewhat more macroscopic approach has also been taken by [37, 38, 39], where $A_D$ is directly inferred by ion-irradiation of colloidal ellipsoids. However, since the swelling mechanism, as pertains to the linear stability of irradiated thin films, was only suggested in 2018 [48], there has been only little time for attention by experimental groups.

3.5.2. Secondary results

1: On the importance of the interface relation. Most analyses prior to [48, 50] have simply assumed a lower interface shape amenable to calculation, typically flat [31], vertically-translated from the top interface [32, 69], or diagonally-translated from the top interface [33], without considering the interface relation arbitrarily and carrying it throughout the entire
calculation in unevaluated form. In doing so, and in considering two idealizations of this quantity in our analysis, we find that the interface relation can strongly influence stress modification and, therefore, critical angle selection. For the diagonal case, we have $\theta_c \approx 30^\circ$ from the theory, while for the vertical case, we have $\theta_c \approx 45^\circ$ from the theory. This is a very significant disparity in theoretical predictions caused by a seemingly innocuous modeling choice, and immediately motivates a more refined treatment in future work. We also note that because of the appearance of a lateral shift $x_0(\theta)$ in our analysis, fitted values of $h_0(\theta)$ to a form $D_0 + D_1 \cos(\alpha \theta)$ obtained via SRIM [78] or other simulation software may not be sufficient for our purposes. Finally, our analysis predicts that an increase in the ratio $\frac{x_0(\theta)}{h_0(\theta)}$ should generally produce a decrease in $\theta_c$ in a target-dependent manner. It is interesting to note that this prediction is qualitatively similar to that of [33] (see their Figure 6), although the physical motivation and mathematical underpinnings are substantially different.

**2: On the inhomogeneity of stress.** We have shown that when IIS is not included in the model, or when the spatial variation of APF and IIS have the same functional form, there is a cancellation effect and the variation does not affect $\theta_c$ selection. In the main text, we have shown this up to the N=2 Legendre polynomial (Equations (3.27) and (3.31)), but we have verified this result up to N=11 and it appears to hold generally. On the other hand, if the dependencies do not have the same functional form, then whichever of APF or IIS is concentrated deeper within the film is strengthened relative to the other.

This observation has suggested a hypothesis. If we suppose that IIS is microscopically connected to the collision cascade dynamics, then we would expect its depth-dependence to reflect that of collision cascade statistics obtainable through simulation software such as SRIM [78]. Given statistics on the mean ion penetration depth $a$ and longitudinal straggle $\alpha$, we may approximate the film depth as $h_0 = a + 2\alpha$, and then the ratio $\frac{a}{a+2\alpha}$ describes a relative cascade depth. Based on our findings above, we would tentatively expect systems with larger simulated values of $\frac{a}{a+2\alpha}$ to have larger critical angles $\theta_c$, and interestingly, such a trend is observed across a wide range of experimental systems, as shown in Figure 3.6.
This will be explored more fully in future work.

Figure 3.6. We compare experimentally-observed values of $\theta_c$ to the mean depth of ion-implantation $a$ against an approximation of normal-incidence film depth, $a + 2\alpha$, where $\alpha$ is the standard deviation of the ion distribution. Experimental $\theta_c$ data are for 250eV-2keV $\text{Ar}^+, \text{Kr}^+, \text{Xe}^+ \rightarrow \text{Si}$ and $\text{Kr}^+, \text{Xe}^+ \rightarrow \text{Ge}$. **From left to right, we have for Si targets:** 1keV $\text{Ar}^+$ [58, 33], 500eV $\text{Ar}^+$ [33], 250eV $\text{Ar}^+$ [43], 2keV $\text{Kr}^+$ [83], 1.2keV $\text{Kr}^+$ [3], 1keV $\text{Kr}^+$ [47], 1keV $\text{Xe}^+$ [33], 500eV $\text{Xe}^+$ [33]. **From left to right, we have for Ge targets:** 1.2keV $\text{Kr}^+$ [57], 1keV $\text{Kr}^+$ [84], 500eV $\text{Kr}^+$ [55], 250eV $\text{Kr}^+$ [55], 2keV $\text{Xe}^+$ [57], 1.2keV $\text{Xe}^+$ [57], 600eV $\text{Xe}^+$ [57]. Orange and blue lines running through each grouping serve as a guide to the eye, and have the same slope.

3.5.3. Open questions and future work

1: **Refinement of interface modeling.** Throughout the literature, three main assumptions have been made about the relationship between the upper and lower interfaces: (1) a flat lower interface as well as (2) vertical and (3) diagonal displacements of the lower from the upper have been considered and incorporated into existing analyses. As discussed elsewhere [6], a major outstanding problem in the field is not merely achieving good agreement between theory and experiment (where fitting data to, e.g., anisotropic Kuramoto-Sivashinsky
can lead to impressive results even in the nonlinear regime without suggesting underlying mechanisms), but identifying the underlying mechanisms leading to good agreement: that is, a “bottom-up” approach to modeling the appropriate physics, and developing models which are precisely as simple or as complicated as needed (but no simpler nor more complicated). The present results suggest that a significant and hitherto-unappreciated component of any analysis is the correct analytical treatment of the interface relation, without which any apparent agreement between theory and experiment must be regarded as possibly coincidental. Indeed, a number of phenomena originally attributed to surface diffusion were later found to be due to viscous relaxation [26, 6], which naturally leads one to question whether similar misattributions could remain to be found, or whether important results could be obscured by the unexpected importance of seemingly minor details.

2: Fully resolved spatial variation. In the present work, we have studied idealized cases where all spatial variation occurs along a path of vanishing crossbeam thickness. In a more realistic treatment, we would not expect that all of the stress modification would act directly along a single axis, particularly in the nuclear stopping regime (i.e., the energetic regime wherein the majority of energy from incoming ions is lost due to interactions between ion and target nuclei, rather than electron cloud interactions; for more detail, we refer the reader to [79, 62]). Indeed, this was precisely the motivation for the Sigmund model of nuclear power deposition [7] and its subsequent, well-known analyses [9, 25]. We consider that in general, we should expect some stress-modification to occur “off-axis”, hence the inhomogeneity of depth-dependence may turn out to influence critical angle in all but certain idealized cases as those we have considered here. The unexpected, significant importance of the interfacial geometry is cautionary. This naturally requires an integral formulation of spatially-varying mechanisms, which will be the subject of future work.

3: Underlying physics in regime of dominant nuclear stopping. In the present work, we have found that variations in the strength of the two theorized bulk mechanisms lead to no changes in \( \theta_c \). We note that such variation may still be important for wavelength
selection, which is best studied outside of the context of a long-wave analysis as we have conducted here. This finding is entirely contingent upon having supposed that anisotropic plastic flow and isotropic swelling occur simultaneously and vary identically throughout the film depth. This is a fundamental assumption about the physical origins of each mechanism—and a rather simplistic one. The physical origin of anisotropic plastic flow, and its apparent good agreement as a theoretical model with experimental results, is a curiosity currently lacking a comprehensive explanation. It is likely that isotropic swelling is driven by either in-film defect dynamics or ion-implantation disrupting the energetically-preferred lattice in Si. Such explanations have led to good agreement between theory and experiment elsewhere [46],[42], while fewer good explanations exist for anisotropic plastic flow in low-energy irradiation. If it is shown that the anisotropic and isotropic components of ion-induced stress do not, in fact, vary together, our finding that depth-dependence cannot affect θc is immediately reversed, although it is possible that the dependence may still be weak. Nonetheless, because we have derived our results for fully arbitrary spatial variation, the present work will be easily adapted to any future findings on this matter.

4: Other systems of interest and predictions. As we have discussed above, further wafer curvature studies similar to those of [47] would be extremely useful, especially for Ge, where we anticipate that \( \frac{A_I}{A_D} \) would take on values substantially larger than those obtained for the system that we have considered, 250eV Ar\(^+\) \(\rightarrow\) Si. It would be interesting to consider how \( \frac{A_I}{A_D} \) changes with energy for the same ion-target combination; superlinearity or non-monotonic behavior of this quantity with respect to energy would seem to be consistent with the observations described in [22]. Additionally, we anticipate that the trendlines shown in Figure 3.6 are likely to continue for larger values of the ratio \( \frac{a}{a+2\alpha} \): we would expect that a system like 250eV Rn\(^+\) \(\rightarrow\) Ge, for which \( \frac{a}{a+2\alpha} \approx .71 \) should have \( \theta_c \approx 75^\circ \). A significant deviation from this trend as \( \frac{a}{a+2\alpha} \) increases would be extremely interesting, as it would appear to invalidate the (albeit tentative) hypothesis that IIS is coupled to ion distribution.

As a final note, angle-dependent wafer curvature data for Ne\(^+\), Ar\(^+\) \(\rightarrow\) Ge would be
perhaps the most interesting, as these systems do not exhibit pattern formation [57], while \( \frac{a}{a+2a} \approx .5 \) for these systems from 250eV-2keV. This observation and comparison with Figure 3.6 suggests that it is, perhaps, not an excess of IIS but, rather, a deficiency of APF that causes these systems to fail to develop ripple patterns. This is broadly consistent with discussion in [57]. Angle-dependent stress measurements would therefore be elucidating, as we would expect \( \frac{A_I}{A_D} \gg 1 \) by virtue of \( \frac{I_{A_D} h_A}{V} \ll 1 \)— that is, a very weak angle-dependence of stress, with little or no compressive-to-tensile transition beyond 45°, in contrast to what we observe in Figure 3.5 and [47].
Chapter 4
Interfacial geometry and phase-change

4.1. Introduction

Previous work by another group has suggested that a model of uniaxial spatially-varying stress may account for at least some of the complex dependence of $\theta_c$ on system parameters [33]. Later, work by our group suggested that a slightly different treatment of this uniaxial stress variation would lead to no effect in $\theta_c$ selection, while the relationship between the free interface and the amorphous-crystalline interface can easily account for up to $15^\circ$ of variation in theoretical predictions of $\theta_c$ (Chapter 3). Because this interface relation has been both the subject of widely-varied modeling assumptions and has proven to be of high significance in aligning theory with experiment, we will derive a precise, physically-realistic treatment of this relation compatible with linear stability analysis. This eliminates one degree of freedom from our, and all subsequent, analyses of this system.

Our earlier work has also shown that at least in the 250eV Ar$^+$ → Si system, for which we have parameter estimates suitable for obtaining theoretical predictions of $\theta_c$ and $\lambda(\theta)$, the effect of ion-induced isotropic swelling is essentially negligible to $\theta_c$-selection (Chapter 3), at least in the case of constant swelling rates throughout the film. This has seemingly eliminated one stabilizing mechanism previously theorized to explain the “pocket of stability” for Ar$^+$ → Si from about 1.5keV-20keV [22]. However, in following with the present theme of exercising significant caution in our treatment of the amorphous-crystalline interface, we will study the effect of the instantaneous loss of density of a parcel of matter as it enters the amorphous bulk from below. Similar effects are known to be important for linear stability problems in solidification and phase-change theory (Chapter 9 of [85]), but have seen little
attention in the theory of ion-induced nanopatterning, despite the amorphous-crystalline interface being an obvious locus of phase-change in the system of our concern, as has been noted [22]. This “instantaneous amorphization” at the amorphous-crystalline interface is shown to exert a strong stabilizing effect, possibly displacing post-amorphization swelling as a major, missing stabilization mechanism in low-energy irradiation of Si. As an aside, we also briefly consider the possibility of slip-flow along the lower interface.

In the present work, we consider the linear stability of ion-irradiated thin films, where the typical no-penetration boundary condition has been relaxed to a phase-change or mass-conservation boundary condition. This results in the modification of the bulk velocity field by the density jump across the amorphous-crystalline interface as new material enters the film and (near-)instantaneously changes volume. Although the amorphization process itself is somewhat ill-defined [33] and various approaches exist to defining and studying amorphization [86, 87, 88, 89, 90, 91, 92], it appears that the response of the amorphous-crystalline interface to changes in the free interface is quite rapid. We also note that the amorphization threshold suggested by [86, 88, 89, 90, 91, 92] in terms of deposited power should be easily achieved except possibly at very low fluxes, high temperatures where thermal annealing of defects may be rapid, or other “unusual” situations.

Such an effect is known to alter linear stability in the context of solidification theory but has not yet been considered in ion-induced nanopatterning. We also determine simple closed-form expressions for the amorphous-crystalline interface in terms of the free interface, appealing directly to the physics of the collision cascade, which was recently shown to strongly modify the critical angle at which pattern formation is predicted to begin on an irradiated target.

Speculatively, we then consider the effect of slip-flow. Due to the small number of atoms involved in a thin film of only a few nanometers thickness [78], and the possibility of relatively large displacements due to ion impacts [7, 62], it is possible that the treatment of irradiated, amorphous thin films as proper continua may be challenged. If so, our system may exhibit a non-vanishing Knudsen number, and it is possible for slip-flow to occur. Toward the
overarching goal of cataloging all relevant physical effects that might affect \( \theta_c \) and \( \lambda(\theta) \) selection, we briefly consider a simplified case of slip boundary conditions.

We find that while slip-flow is a weak contributor, the effect of phase-change at the amorphous-crystalline boundary may impart a strong ion-, target- and energy- dependence and, alongside the interface relation, may contribute to a unified model of critical angle selection across a wide range of experimental systems.

4.2. Model

**Bulk description.** We begin with the differential form of mass conservation in the bulk,

\[
\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \vec{v}) = 0, \tag{4.1}
\]

where \( \rho = \rho(x, z) \), prohibiting the common simplification \( \nabla \cdot \vec{v} = 0 \). In the limit of low Reynolds number, we have the differential form of linear momentum conservation in the bulk,

\[
\nabla \cdot \mathbf{T} = 0, \tag{4.2}
\]

where we define our stress tensor

\[
\mathbf{T} = -p \mathbf{I} + 2\eta \{ \dot{\mathbf{E}} - \dot{\mathbf{E}}_b \}
\]

\[
\dot{\mathbf{E}} = \frac{1}{2} (\nabla \vec{v} + \nabla \vec{v}^T)
\]

\[
\dot{\mathbf{E}}_b = f A_D \tau(z; g, h) \mathbf{D}(\theta)
\]

\[
\mathbf{D}(\theta) = \begin{bmatrix}
\frac{3}{2} \cos(2\theta) - \frac{1}{2} & 0 & \frac{3}{2} \sin(2\theta) \\
0 & 1 & 0 \\
\frac{3}{2} \sin(2\theta) & 0 & -\frac{3}{2} \cos(2\theta) - \frac{1}{2}
\end{bmatrix}.
\tag{4.3}
\]

This stress tensor imparts upon the film a description of *anisotropic plastic flow*, our proposed source of morphological instability in the stress-dominated regime. As discussed elsewhere [6, 32, 41, 37, 38, 39, 80], this stress tensor is in common use for low-energy irradiation of semiconductors on a phenomenological basis, having been borrowed from successful modeling
of higher-energy systems. We also consider ion-induced swelling (IIS) [48, 50], which appears through the equation of state,

$$\rho(\Delta) = \frac{\rho_a}{1 + \Delta}, \quad (4.4)$$

where $\Delta$ represents the accumulation of ion-induced damage leading to post-amorphization loss of density. The accumulation is damage is described by the advection-type equation,

$$\frac{\partial \Delta}{\partial t} + \vec{v} \cdot \nabla \Delta = \alpha(z; g, h), \quad (4.5)$$

which allows the possibility that a parcel of matter accumulates damage depending on its depth within the amorphous bulk. The presence of ion-induced swelling within the film has previously been shown to induce an additional stabilization of the free interface against perturbations [48, 50].

**Boundary conditions.** At the free upper interface, $z = h$, we have

$$v_{I,h} = \vec{v} \cdot \hat{n} - V \frac{\rho_c}{\rho} \quad \Rightarrow \quad [T] \cdot \hat{n} = -\gamma \kappa \hat{n}, \quad (4.6)$$

where the first equation simply describes the typical kinematic condition modified by ion-induced sputtering [48]. The second is a typical statement of stress balance at the free interface, from which the effect of viscous relaxation enters into the governing equations. At the amorphous-crystalline interface, $z = g$, we have

$$\Delta = 0$$

$$\vec{v} \cdot \hat{t} = \beta_s \hat{n} \cdot \nabla(\vec{v} \cdot \hat{t}) \quad \Rightarrow \quad [\rho \vec{v}] \cdot \hat{n} = [\rho] v_I. \quad (4.7)$$

The first equation implies via the equation of state that the density immediately on the amorphous side of the amorphous-crystalline boundary should be $\rho_a$, as newly-amorphized material has had no time to be acted upon by IIS.

The second two boundary conditions, and their interaction with the amorphous-crystalline interface shape itself, are the focus of the present work: the first is a Navier slip flow condition with slip length $\beta_s$. The second is a general statement of mass conservation at the
interface, which will permit the phenomenon of *volume convection* [85]: apparent flow at the boundary due to an instantaneous drop in density across the boundary. We have included the derivation of this condition in the Appendix. The inclusion of such an instantaneous density-drop is appropriate in the case of c-Si (crystalline silicon) transitioning to a-Si (amorphous silicon), as a-Si is well-known to have a lower density ($\rho_a$) than c-Si ($\rho_c$), with estimates ranging from about 3-6% [67] for the a-c transition in Si or about 2-10% [87], seemingly dependent on the preparation of the substrate. We note that in the case where $\rho_a = \rho_c$, the tacit assumption throughout the irradiated thin film linear stability literature so far, this condition simply reduces to $\vec{v} \cdot \hat{n} = 0$. When slip-length $\beta_s = 0$, the Navier condition reduces to $\vec{v} \cdot \hat{t} = 0$. Together, these two reductions reproduce the familiar $u = w = 0$ at the lower interface.

![Figure 4.1. Schematic depicting the effect of the phase-change boundary condition for a planar amorphous-crystalline interface.](image)

**Remarks.** We note that there is a great deal of uncertainty in the exact value of $\rho_a$ for a given system, with the value seemingly depending on fluence, ion implantation, and possibly energy and projectile species [72, 65, 87, 67]. As we will show, quantitatively resolving this uncertainty may lead to improvements in the understanding of critical angle and wavelength selection in ion-irradiated surfaces. In the present work, we will consider Ar$^+$ → Si at a
flux of $12 \frac{\text{ions}}{\text{nm}^2 \cdot \text{s}}$, a system for which we have a small data set of angle-dependent in-plane stress measurements as well as time series data for the in-plane stress ([47], Chapter 3), as an illustration. As discussed in Chapter 3, we regard the so-called Peak B from the experimental work of [47] as occurring near steady-state; if this is correct, the fluence at that time is expected to be greater than that required in [67] to induce a $\sim 5.5\%$ reduction in $\rho_a$ compared to $\rho_c$, consistent with the values measured by [22] of about a 4-5% density reduction—although perhaps not instantaneous density reduction. We also note that density reductions of up to 10% in the amorphous-to-crystalline transition for Si are apparently known in the experimental literature—again, seemingly due to the means of preparation [87]. Finally, it is worth observing that $\frac{\rho_a}{\rho_c}$ in the keV range is apparently larger than that in the MeV range [67], suggesting the possibility that $\frac{\rho_a}{\rho_c}$ may vary weakly and inversely with energy, which would again contribute to the non-negligibility of the interfacial phase-change phenomena in the present system.

4.3. Analysis

Our analysis comprises three main areas. First, we will derive a highly-general expression for the growth rate of a perturbation (i.e., the linear dispersion relation) from the governing equations via a standard linear stability analysis. This is a further generalization of Chapter 3, where we now take Navier slip and generalized mass-conservation (or “phase-change”) boundary conditions at the amorphous-crystalline interface so that we may consider the effect of instantaneous phase-change as new material enters the irradiated bulk. As will be seen, the present work differs substantially from our previous work in that estimating parameters $f A_D \eta$ and $f A_I \eta$ as groups is insufficient due to the extra terms added into the growth rate by phase-change at the lower interface which do not scale with $\eta$; see Equation (4.17). We therefore require an explicit estimate of $\eta$ if we wish to compare with the data due to [47], or in general if we wish to study angle-dependent wavelength $\lambda(\theta)$.

Within the expression of this growth rate, there are three immediate obstacles to the prediction of $\theta_c$ and $\lambda(\theta)$. The first is determining the geometric relationship between the
upper and lower interface, some assumption about which is naturally required by any thin-film linear stability analysis of the present type. Remarkably, this simple relationship is capable of introducing up to 15° of uncertainty in the value of \( \theta_c \) (Chapter 3), even in the absence of swelling, and therefore demands attention. The second is obtaining reliable parameter estimates of the deviatoric and isotropic parts of ion-induced stress modification, \( f_{AD} \) and \( f_{AI} \). From our previous work (Chapter 3), we have estimates for the 250eV Ar\(^+\) → Si system, and our estimate of \( f_{AD} \) appears to agree roughly with other existing estimates throughout the literature. Third is estimating viscosity \( \eta(\theta) \), which we will only briefly touch on here; we will choose one plausible value of \( \eta(0) \) based on a comparison of data and two scaling arguments, rather than a range, in order to avoid a parameter space that is larger than necessary for our present purposes.

4.3.1. Linear stability

We expand the governing equations described above in the neighborhood of steady-state solutions, which are determined by solving Equations (4.11)-(4.13). We will group all scalar fields as \( \Phi = [\rho, u, w, p, \Delta] \), denote the two interfaces \( g \) and \( h \), and expand formally as

\[
\Phi(x, z, t) \rightarrow \Phi_0(z) + \epsilon \Phi_1(x, z, t)
\]

\[
h(x, t) \rightarrow h_0 + \epsilon h_1(x, t)
\]

\[
g(x, t) \rightarrow g_0 + \epsilon g_1(x, t),
\]

representing small perturbations to the steady state fields of the form \( \Phi_1 = [\rho_1, u_1, v_1, w_1, p_1, \Delta_1] \).

We then apply the well-known linear stability ansatz as

\[
\Phi_1(x, z, t) \rightarrow \Phi_1(z) \exp(\sigma t + ikx),
\]

and we will seek the linear dispersion relation \( \sigma \), whose real part represents the growth rate of a given mode. Implicitly, we have \( \sigma = \sigma(k; \theta) \), which allows us to determine experimentally observed wavelengths from the theory by computing the fastest-growing mode \( k \) at a given irradiation angle \( \theta \). We are also able to compute \( \theta_c \) from the theory, being the minimal \( \theta > 0 \) such that \( \text{Re}[\sigma(k; \theta)] > 0 \); this implies the onset of instability (since perturbations
would then grow rather than decay), hence pattern formation. Following the conversion to a moving frame,

\[
\begin{align*}
  h &\to h - Vt \\
  g &\to g - Vt \\
v_{I,h} &\to v_{I,h} - V(\hat{k} \cdot \hat{n}) \\
z &\to z - Vt \\
v &\to v - V\hat{k},
\end{align*}
\]

we obtain our linearized equations. In the steady state, we have

\[
\begin{align*}
  \frac{\partial}{\partial z}(\rho_0 w_0) &= 0 \\
  \eta u_{0zz} &= 2\eta f A_D D_{13} \tau_{0,z} \\
  -p_{0z} + 2\eta w_{0zz} &= 2\eta f A_D D_{33} \frac{\partial \tau_0}{\partial z} \\
  \rho_0 &= \frac{\rho_a}{1 + \Delta_0} \\
  w_0 \Delta_{0z} &= \alpha_0(z; 0, h_0).
\end{align*}
\]

At \(z=0\), we have

\[
\begin{align*}
  \Delta_0 &= 0 \\
  u_0 &= \beta_s u_{0z} \\
  w_0 &= V \left( \frac{\rho_c}{\rho_a} \right),
\end{align*}
\]

At \(z=h_0\), we have

\[
\begin{align*}
  0 &= w_0 - V \frac{\rho_c}{\rho_0} \\
  u_{0,z} &= 2f A_D \tau_0(z; 0, h_0) D_{13} \\
p_0 &= 2\eta \{ w_{0z} - f A_D \tau_0(z; 0, h_0) D_{33} \}.
\end{align*}
\]
In the bulk at $O(\epsilon)$,

\[ \rho_{1t} + \rho_0 u_{1x} + \rho_0 w_1 + \rho_0 w_{1z} + \rho_1 w_0 + \rho_1 w_{0z} = 0 \]

\[-p_{1x} + \eta \{2u_{1xx} + u_{1zz} + w_{1xz}\} = 2fA_D\eta \{D_{11}\tau_{\epsilon,x} + D_{13}\tau_{\epsilon,z}\} \]

\[-p_{1z} + \eta \{w_{1xx} + 2w_{1zz} + u_{1xz}\} = 2fA_D\eta \{D_{31}\tau_{\epsilon,x} + D_{33}\tau_{\epsilon,z}\} \]

\[ \rho_1 = \frac{-\rho^*\Delta_1}{(1 + \Delta_0)^2} \]

\[ \Delta_{1t} + u_0\Delta_{1x} + w_0\Delta_{1z} + w_1\Delta_{0z} = \alpha_\epsilon \]

At $z = 0$,

\[ \Delta_{0,z}(z; 0, h_0)g_1 + \Delta_1(z; 0, h_0) = 0 \]

\[ u_{0z}g_1 + u_1 + g_{1x}(w_0 - V) = \beta_s(g_{1u_{0zz}} + u_{1z} + g_{1x}(w_{0z} - u_{0z})) \]

\[ -g_{1x}u_0 + w_{0z}g_1 + w_1 = \left( \frac{\rho_a - \rho_c}{\rho_a} \right) g_{1t} \]

At $z = h_0$,

\[ h_{1t} = w_1 - u_0h_{1x} + h_1w_{0z} + \frac{V\rho^*}{\rho_0^*} \{\rho_{0z}h_1 + \rho_1\} \]

\[ 2\eta\left\{\frac{1}{2}(u_{1z} + w_{1x}) - fA_D\tau_0 D_{13}\right\} - h_{1x}T_{011} = 0 \]

\[ -p_1 + 2\eta\{w_{1z} - fA_D\tau_0 D_{33}\} + T_{033}^{33} = 0, \]

The application of the linear stability ansatz and the further expansion in small wavenumber are described in the Appendix. In the above, $T_0$ refers to the steady-state stress tensor, and $T_0^{ij}$ refers to its $(i, j)$ component, where we have used superscripts to avoid confusion with subscripts, which we have used elsewhere to keep track of expansions performed. The subscript $z$ refers to the derivative with respect to spatial variable $z$.

For the sake of generality, and as a staging point for future work, we have performed the linear stability analysis with spatially-varying stresses included, although we will not make use of them here (following from Chapter 3 that some other explanation may be needed). Following the solution of the equations described in the Appendix, we obtain a very general expression for the linear dispersion relation. Under the simplification that APF and IIS are of constant strength throughout the film (hence $\tau = 1, \alpha_{01} = 1, \alpha_{\epsilon 1} = 0$), we obtain the linear dispersion relation up to the leading terms at $O((kh_0)^2)$. We then simply append the
well-known long-wave viscous relaxation term due to Orchard [27]. Of interest to us is the 
real part,

$$
\text{Re}(\sigma) = \left( -\frac{V}{\hbar_0(\theta)} \left( 1 - \frac{\rho_a}{\rho_c} \right) - \frac{fA_I}{2} \left( \frac{\rho_a}{\rho_c} \right)^2 \right)
\begin{align*}
& -3fA_D \left( \frac{\rho_a}{\rho_c} \right) \left( 1 + \frac{2\beta_s}{h_0(\theta)} \right) \cos(2\theta) - \left[ \frac{x_0(\theta)}{h_0(\theta)} \right] \left( 1 + \frac{\beta_s}{h_0(\theta)} \right) \sin(2\theta) \right) (kh_0(\theta))^2 \tag{4.17}
\end{align*}
$$

which will determine both $\theta_c$ and the long-wave approximation of $\lambda(\theta)$. We note that this 
result expresses the growth rate $\text{Re}(\sigma)$ for arbitrary $x_0(\theta)$ and $h_0(\theta)$, which emerge via the 
assumption

$$
g(x, t) = h(x - x_0(\theta), t) - h_0(\theta), \tag{4.18}
$$

itself a generalization of treatments of the amorphous-crystalline interface used elsewhere [31, 
32, 33, 48, 50]. In the present work, we will offer a heuristic explanation for the existence 
of this simple form in a linear stability context, and in Chapter 5, we will derive it from 
first principles. At this point, it becomes apparent that we require a description of the 
amorphous-crystalline interface $g_1$ so that these two quantities are specified. In our previous 
work (Chapter 3), we have shown that the relationship of this interface to the free interface 
can strongly influence $\theta_c$ selection, and we anticipate that it will affect $\lambda(\theta)$ selection as 
well. We therefore seek a description of $g_1$ directly accessible from BCA simulations [78].

“BCA” is the binary collision approximation, a computational technique that allows much 
faster and still quite detailed simulations of the collision cascade by making simplifying 
assumptions about the number of recoils that need be tracked. We refer the reader to [78] 
and its documentation for more details.

4.3.2. Interface relation from BCA

Physically, the amorphous-crystalline interface is characterized by the furthest region 
amorphized by incoming ions due to, e.g., power deposition, damage to the crystalline lattice, 
or bond elongation [62]. While essentially flux-dependent, as noted elsewhere, the film
thickness is not expected to exhibit a simple \( \cos(\theta) \)-dependence on the beam angle \([20, 28]\), and \([20]\) has suggested that the film thickness may be an important quantity modifying \( \theta_c \)-selection; for further commentary, see \([6]\). However, in neither of these previous studies was the lateral shift \( x_0(\theta) \) explicitly considered or incorporated into the analysis, while the form of the linear dispersion relation, Equation (4.17), reveals its importance in modifying \( \theta_c \) within the present framework. This motivates study of both \( h_0(\theta) \) and \( x_0(\theta) \), as even a fitted \( h_0(\theta) \approx D_0 + D_1 \cos(c\theta) \) \([20]\) is insufficient for our purposes. Indeed, it is primarily the \( \frac{x_0(\theta)}{h_0(\theta)} \) term that responsible for the change in \( \theta_c \) observed in Chapter 3, at least for APF and IIS constant in space.

Figure 4.2. Schematic depicting the derivation of the amorphous-crystalline interface location as described in Equations (4.19)-(4.23).

Following the usual Sigmund model \([7, 8]\) which expresses the recoil distribution (or deposited power) as a multivariate Gaussian in downbeam-crossbeam coordinates. Similarly, we take downbeam mean \( a \), downbeam standard deviation \( \alpha \) and crossbeam standard deviation \( \beta \), with \((\hat{x}, \hat{z})\) as crossbeam-downbeam coordinates respectively. Then we have

\[
E_D(\tilde{x}, \tilde{z}) = \frac{1}{2\pi\alpha\beta} \exp \left( -\frac{(\tilde{z} - a)^2}{2\alpha^2} - \frac{\tilde{x}^2}{2\beta^2} \right). \tag{4.19}
\]
and we may change from crossbeam-downbeam coordinates to typical Cartesian, while simultaneoulsy choosing \((0, 0)\) as the location at which the incoming ion enters the film, via the transformation
\[
\tilde{x} \rightarrow x \cos(\theta) + z \sin(\theta) \\
\tilde{z} \rightarrow x \sin(\theta) - z \cos(\theta).
\] (4.20)

We then have, in \((x,z)\),
\[
E_D(x, z) = \frac{1}{2\pi \alpha \beta} \exp \left( -\frac{[x \sin(\theta) - z \cos(\theta) - a]^2}{2\alpha^2} - \frac{[x \cos(\theta) + z \sin(\theta)]^2}{2\beta^2} \right),
\] (4.21)

Now we consider the simple level curve of such an ellipse, absorbing the coefficient into \(E_A\), a parameter which defines the amorphization threshold.
\[
f(x, z) = \frac{1}{2} \left( \frac{(a + z \cos(\theta) - x \sin(\theta))^2}{\alpha^2} + \frac{(x \cos(\theta) + z \sin(\theta))^2}{\beta^2} \right) - \ln \left( \frac{E_0}{E_A} \right),
\] (4.22)

and we seek the locations on this level curve such that
\[
\nabla f \cdot <1, h_X> = 0,
\] (4.23)

where we have not yet performed any expansion in \(h_X << 1\) or in \(\epsilon << 1\). That is, we suppose that a reasonable approximation of the amorphous-crystalline interface is obtained by determining the region amorphized by a single Gaussian ellipsoid resulting from time-averaged power deposition due to ions originating from a single location, finding the point in this region that represents a maximal distance from the unperturbed (but possibly inclined) interface, and then performing this process for all points on the upper interface—essentially “dragging” that ellipsoid across the irradiated surface, such that each point on the upper interface generates exactly one point on the lower interface. The relationship between the upper-interfacial point and the lower-interfacial point may then be expressed simply in terms of a vertical shift \(h_0(\theta)\) and a lateral shift \(x_0(\theta)\), leading to our desired interface relation.

For simplicity, we suppose that the point on the lower interface generated by a given point on the upper interface is determined by the location for which the normal vector to the level curves of the Gaussian ellipsoid is orthogonal to the tangent vector to the patch of surface from which that ellipsoid was generated. The above produces two equations which
may be solved for $x$ and $z$ in terms of $a, \alpha, \beta, E_0, E_A, h_X$. The solutions for $x$ and $z$ prior to expansion in $h_x \ll 1$ are too lengthy to record here (and are not especially interesting), so we will not show them here, although they can be recovered easily. In the $h_x \ll 1$ expansion, we find

$$h_0(\theta) = a \cos(\theta) + 2 \left( \sqrt{\alpha^2 \cos^2(\theta) + \beta^2 \sin^2(\theta)} \sqrt{\frac{\ln\left(\frac{E_0}{E_A}\right)}{2}} \right) + O(h_x^2), \quad (4.24)$$

and

$$x_0(\theta) = a \sin(\theta) + 2 \left( \frac{(\alpha^2 - \beta^2) \sin(\theta) \cos(\theta)}{\sqrt{\alpha^2 \cos^2(\theta) + \beta^2 \sin^2(\theta)}} \sqrt{\frac{\ln\left(\frac{E_0}{E_A}\right)}{2}} \right) - h_x \left( a \cos(\theta) + \frac{(\alpha^2 - \beta^2) (4(\alpha^2 + \beta^2) \cos(2\theta) + (\alpha^2 - \beta^2)(3 + \cos(4\theta))))}{(\alpha^2 \cos^2(\theta) + \beta^2 \sin^2(\theta))^{3/2}} \right) + O(h_x^2) \quad (4.25)$$

by identifying $z = h_0$ and $x = x_0$. There are two important consequences of these forms in the context of linear stability analysis. First, in any linearization $h = h_0 + \epsilon h_1$, the correction for $h_0$ will be $O(\epsilon^2)$, hence negligible in the present work. Second, if we first observe that $x_0(\theta)$ appears in the dispersion relation (see Appendix) via

$$\frac{\tilde{g}_1}{h_1} = \exp(-i k x_0(\theta)); \quad (4.26)$$

it is clear that the $O(h_x)$ correction also becomes negligible within the linearization: expansion of the above in small $kh_0(\theta)$ would also lead to a term $\propto \epsilon^2$. Hence for our purposes, we may truncate these correction terms and use the leading-order expressions only, which offers a convenient simplification without sacrificing accuracy. Truncating and assigning

$$E_A = E_0 \exp(-2) \to \ln\left(\frac{E_0}{E_A}\right) = 2, \quad (4.27)$$

i.e., supposing that the threshold for amorphization is the angle-dependent mean plus two angle-dependent standard deviations in the projected $z$ axis, the above further reduce to

$$x_0 = a \sin(\theta) + 2 \left( \frac{(\alpha^2 - \beta^2) \sin(\theta) \cos(\theta)}{\sqrt{\alpha^2 \cos^2(\theta) + \beta^2 \sin^2(\theta)}} \right); \quad (4.28)$$

$$h_0 = a \cos(\theta) + 2 \left( \sqrt{\alpha^2 \cos^2(\theta) + \beta^2 \sin^2(\theta)} \right),$$
which are surprisingly concise forms, and all parameters $a, \alpha, \beta$ may be obtained directly from simulation software, such as SRIM [78]. In principle, we only require that $a, \alpha, \beta$ describe a bivariate Gaussian that is associated with “damage” to the substrate; we do not strictly require that it be the recoil distribution, power deposition, or any other quantity. In the present work, we will choose the implanted ion distribution, as it is most easily accessible from SRIM, broadly tracks recoils and deposited power, and is a source of lattice-damaging interstitials, while still being well-approximated as a bivariate Gaussian.

In principle, a much more complicated description of the amorphous layer is possible, but would introduce significant complexity beyond the scope of our present interest; we refer the curious reader to the work of [86, 88, 89, 90, 91, 92] for detailed discussion of the crystalline-to-amorphous transition of planar surfaces (rather than the nonplanar, corrugated surfaces under consideration here). In context of the aforementioned work, the derivation of the amorphous-crystalline interface shape described above is essentially a “rapid-amorphization” limit. Nonetheless, we will see that it is sufficient, and, in Chapter 5, we revisit it in greater rigor.

**Some interesting limits.** We note that the above expressions are easily specialized to previously-studied interface relations by taking appropriate limits, which is consistent with our claim of their generality. In particular, the “vanishing cross-beam width” limit,

$$\lim_{\beta \to 0} h_0(\theta) = (a + 2\alpha) \cos(\theta)$$

$$\lim_{\beta \to 0} x_0(\theta) = (a + 2\alpha) \sin(\theta),$$

is the “pencil-like” relation used in [33, 48, 50]. A particular “no angle-dependence” limit,

$$\lim_{\theta \to 0} h_0(\theta) = (a + 2\alpha)$$

$$\lim_{\theta \to 0} x_0(\theta) = 0,$$

(4.29)
is the “vertical-translation” case used in [32, 69]. The simple case of perfectly spherical collision cascades, $\alpha = \beta$, is

$$\lim_{\beta \to \alpha} h_0(\theta) = a \cos(\theta) + 2\alpha$$

$$\lim_{\beta \to \alpha} x_0(\theta) = a \sin(\theta) \qquad (4.31)$$

The spherical approximation has been previously considered at least by [29, 11] as a simplifying assumption broadly appropriate for low-energy noble gas ion irradiation of Si.

4.3.3. Two scaling arguments for $\eta$

**The linear scaling of Vauth and Mayr.** In this Chapter, we would like to continue our comparison between theory and the experimentally-inferred values for $f_A I$ and $f_A D$ due to [47] and the discussion in Chapter 3. However, the results of Chapter 3 return values for $f_A D \eta$ and $f_A I \eta$ rather than $f_A I$ and $f_A D$ themselves. Unlike the work of [32], the dispersion relation Equation 4.17 cannot be multiplied through by $\eta$ in order to “bypass” the need to know an exact value of $\eta$. This introduces a new difficulty, because $\eta$ is notoriously difficult to measure for systems of present interest [6], and few measurements exist. However, [93] have attempted to infer values of $\eta$ for 1keV Ar$^+$ $\rightarrow$ Si at room temperature from MD simulations. Their calculations have been leveraged as a scaling argument throughout the literature [14, 18, 20] in order to estimate $\eta$ for systems at different energies. Briefly, they suggest

$$\eta(0) = \frac{1}{H \times N_{DPAPS}}, \qquad (4.32)$$

where

$$N_{DPAPS} = \left( \frac{\Omega f}{d} \right) N_{recoils} \qquad (4.33)$$

are *defects per atom per second*, $\Omega$ is atomic volume, $f$ is flux, $d$ is film depth, and $N_{recoils}$ is the number of recoils per atom. Via the well-known Kinchin-Pease model [62],

$$N_{recoils} = \frac{0.8E}{2E_D}, \qquad (4.34)$$
Vauth and Mayr provide a value of $H$ for 1keV Ar$^+$ → Si at 300K based on MD simulations [93]. $E_D$ is the displacement threshold for the irradiated target, and $E$ is nominal beam energy. Using $H$ as a constant of proportionality, we obtain

$$\eta_2 = \left( \frac{f_1}{f_2} \right) \left( \frac{d_2}{d_1} \right) \left( \frac{E_1}{E_2} \right) \eta_1,$$

which may be used to compare the normal-incidence viscosities of two different experimental systems—at least if we believe that $H$ truly is a constant, or at least approximately so, across a sufficiently wide range of experimental systems. Using the experimentally-inferred value for 1keV Ar$^+$ → Si at $2 \times 10^{-2}$ ions nm$^{-2}$, $\eta(0) \approx 1.5 \times 10^2$ GPa·s, the above scaling suggests $\eta(0) \approx .5$ GPa·s for 250eV Ar$^+$ → Si at $1.2 \times 10^1$ ions nm$^{-2}$ s. However, there is some uncertainty about the value used in [28], as it is reported to disagree with a value determined in [14] for the same experimental system by about an order of magnitude. Additionally, some discussion in [47] and [44] suggests that the argument of Vauth and Mayr [93] may underpredict $\eta$ at higher fluxes, as stress relaxation may suggest $\eta$ higher than predicted by Vauth and Mayr.

**Scaling motivated by bimolecular annihilation.** On the other hand, recent work by [94, 46] has suggested that viscosity in ion-irradiated targets may be substantially more complex than suggested by the scaling argument described above. In their work, the amorphous bulk is treated as the setting for ongoing creation of defects, annihilation of defects by the recombination of vacancy-interstitial pairs, and sputter-removal of defects. This leads to a scaling argument that predicts a significantly higher value of $\eta$ for the present system.

It is hypothesized that viscosity scales inversely with the concentration of flow defects in the bulk; following the notation of [46], we have

$$\eta(0) = \frac{1}{\alpha C},$$

where $\alpha$ is fluidity per flow defect, and $C$ is concentration of flow defects. We can seek a steady-state $\eta$ via

$$\eta(0) = \frac{1}{\alpha C_{ss}},$$

$$C_{ss} = \left( \frac{fC_0}{D_2d} \right)^{1/2},$$
where \( f \) is flux, \( C_0 \) is flow defects per ion, \( D_2 \) is the reaction rate for bimolecular annihilation of defects, and \( d \) is film thickness. The solution for \( C_{ss} \) is given in [46]. Rewriting the two equations above, we find

\[
\eta = \sqrt{\frac{D_2}{\alpha^2}} \sqrt{\frac{d}{fC_0}}. \tag{4.38}
\]

From [94], we expect \( D_2 \), the bimolecular annihilation rate, to exhibit temperature-dependence, according to

\[
D_2(T) = D_{2,0} \exp\left(\frac{-Q_r}{K_BT}\right), \tag{4.39}
\]

where \( D_{2,0} \) is a constant, \( Q_r = Q - \dot{\eta} \), with \( Q \) the isoconfigurational activation enthalpy and \( \dot{\eta} \) the average activation enthalpy. Witvrouw and Spaepen have determined \( Q_r \approx 1.7 \), based on \( Q \approx 1.8 \) and \( \dot{\eta} \approx .1 \) in \( \text{eV} \) at high temperatures [94]. \( K_B \) is Boltzmann’s constant, \( 8.617333 \times 10^{-5} \text{eV K} \). Now we hypothesize that \( D_{2,0} \) and \( \alpha \) (fluidity per defect) are essentially constants across all energies, fluxes, and temperatures of interest to us. Then we group \( \sqrt{\frac{D_{2,0}}{\alpha^2}} \) as a proportionality constant and we find

\[
\eta_2 = \eta_1 \sqrt{\frac{f_1 C_{0,1}}{f_2 C_{0,2}}} \sqrt{\frac{d_2}{d_1}} \exp\left(\frac{-Q_r}{2K_B \left[\frac{1}{T_2} - \frac{1}{T_1}\right]}\right) \tag{4.40}
\]

as an alternative scaling across fluxes, temperatures, and materials. This naturally leads to questions of the temperature of the substrate during ongoing irradiation, and we have ignored other effects (like thermal relaxation of the lattice [94]) entirely, which would be expected to counteract the predicted increase in viscosity due to faster bimolecular annihilation. We propose a full exploration of these effects as future work.

Here, we will suppose that the experiment of [47] is effectively isothermal, or that the structural relaxation effects approximately cancel out the apparent increase in viscosity due to faster (thermally-activated) bimolecular annihilation:

\[
\eta_2 \approx \eta_1 \sqrt{\frac{f_1 C_{0,1}}{f_2 C_{0,2}}} \sqrt{\frac{d_2}{d_1}}. \tag{4.41}
\]

This simplification is made also, in part, to avoid the need to consider \( Q_r \) in the present work, which requires experimental measurement clearly beyond the scope of this work. If \( Q_r \)
is fairly small at lower temperatures, it may turn out that the thermal dependence is weak at the temperatures reached by [47] (room temperature up to 100° C). We also note that \( D_2 \) values reported in [46] appear to exhibit an inverse-scaling with \( E \), and it is possible that this scaling with \( E \) dominates aggregate thermal effects.

Regardless, the above equation exhibits a square-root scaling with flux, rather than a linear scaling; for large differences in flux, this may lead to very significant differences in the results of the scaling. The above argument, when compared to the value of \( \eta(0) \) obtained in [28], leads to \( \eta(0) \approx 8.5 \text{ GPa} \cdot \text{s} \). Such a value is roughly an order of magnitude higher than that anticipated by scaling [28] with the argument of [93], and it is seemingly consistent with the value obtained by scaling [14] with the argument of [93]. Because this value is close to other estimates while seeming to predict somewhat longer relaxation times, consistent with the experimental results described in [44] and [47], we will adopt it on a tentative basis.

The above considerations of \( \eta \) possibly scaling with system temperature due to thermally-activated bimolecular annihilation raise numerous questions beyond the scope of the present work. We do not assert that \( \eta(0) \approx 8.5 \text{ GPa} \cdot \text{s} \) must be the correct value for the 250eV \( \text{Ar}^+ \rightarrow \text{Si} \) system—only a plausible value, motivated by comparison with experimental and theoretical results, so that we can study our proposed treatment of the amorphous-crustalline interface in context of a known experimental system. The theoretical treatment of this interface and qualitative results that come out of it, rather than theoretical-experimental agreement \textit{per se}, remain the focus of this work.

4.4. Results

Finally, we would like to obtain theoretical estimates of \( \lambda(\theta) \) and \( \theta_c \) from Equation (4.17) in order to qualitatively study the effects of our treatment of the amorphous-crustalline interface. Here, we will compare the implications of different forms for \( x_0(\theta) \) and \( h_0(\theta) \). When we refer to the “diagonal interfaces” case, we use Equations (4.29); likewise, Equations (4.30) characterize “vertical interfaces”. For the collision-cascade based interface relation, Equations (4.28), we have made the assumption that the Gaussian distribution of the final
resting places of implanted ions (via SRIM [78]) is a sufficient proxy for power deposition or “accumulated damage” capable of inducing amorphization, while noting that it is more common to use the recoil statistics [20, 43]. However, the final ion distribution is frequently deeper into the film than the recoil distribution; as we are primarily concerned with “damage” in general, we follow this somewhat simpler assumption. Accordingly, we use $a = 1.8$ nm, $\alpha = .7$ nm, $\beta = .8$ nm for 250eV Ar$^+$ → Si and $a = 3.5$ nm, $\alpha = 1.6$ nm, $\beta = 1.5$ nm for 1keV Ar$^+$ → Si, which were obtained from SRIM [78].

When an estimate of $\eta(0)$ is required, we have used $\eta = 8.5$ GPa · s based on the above discussion of work due to [93] and [46]. For the values of $f_{AD}$ and $f_{AI}$, we have used $f_{AD} \approx 9.247 \times 10^{-3} \frac{1}{s}$, $f_{AI} \approx 3.365 \times 10^{-3} \frac{1}{s}$ based on this value of $\eta(0)$ and the estimates obtained in Chapter 3. The surface energy $\gamma$ has been estimated as 1.36 $\frac{J}{m^2}$ based on the work of [93, 95]. We note that $f_{AI}$, $f_{AD}$ and $V$ all scale with flux; hence the theoretical prediction for $\theta_c$ is flux-independent, relying only on the sign-changing coefficient of the real part of the dispersion relation. Finally, we consider that in most prevailing analyses, viscosity $\eta(\theta)$ has been treated as a constant. As discussed elsewhere [28], this quantity is likely to scale inversely with $\cos(\theta)$, as viscosity in the present system is specifically ion-induced, rather than an innate material property. We simply consider

$$\eta^{-1}(\theta) = \cos(\theta)\eta^{-1}(0),$$

expressing the flux-diluted, ion-enhanced fluidity. From the discussion in [28, 46], this appears to be a reasonable approximation, reinforced by both fits to GISAXS data and an underlying theory of ion-enhanced fluidity based on defect production and annihilation. When computing $\lambda(\theta)$ or $\theta_c$, it is appropriate to take $f \rightarrow f \cos(\theta)$, leading to each term being $\propto \cos(\theta)$ due to flux dilution (to leading order). However, due to the form of Equation (4.17), this flux dilution factor is essentially irrelevant, as $\theta_c$ and $\lambda(\theta)$ both emerge from calculations where this factor of $\cos(\theta)$ can be safely canceled.
4.4.1. Theoretical results

Figure 4.3. Using the fitted value $\eta = 8.5$ GPa s, and values of $f_A D \approx 9.247 \times 10^{-3} \text{ s}$, $f_A I \approx 3.365 \times 10^{-3} \text{ s}$ for 250eV Ar$^+$ $\rightarrow$ Si at $1.2 \times 10^3 \text{ ions mm}^{-2} \text{ s}$. Increasing the slip-length to film-thickness ratio tends to drive $\theta_c$ to a fixed value, but for physically-expected values of this ratio $\frac{\beta_s}{h_0(\theta)} \ll 1$, the effect is quite weak. On the other hand, the effect of the density-drop across the amorphous-crystalline boundary is quite strong even within the physically-expected range of values.

Slip flow is likely negligible. We note that the dimensionless constant $\frac{\beta_s}{h_0(\theta)}$ naturally arises in the linear dispersion relation (Equation (4.17)). We consider $\theta_c$ selection as determined by this ratio, the choice of interface relation, and the density drop ratio $\frac{\rho_a}{\rho_c}$ in Figure 4.3. It is clear that the effect of a large $\frac{\beta_s}{h_0(\theta)}$ is simply to induce an asymptote in $\theta_c$ as $\frac{\beta_s}{h_0(\theta)} \rightarrow \infty$; however, we do not anticipate that $\frac{\beta_s}{h_0(\theta)}$ should be large. Already, it can be seen by comparison of the left and right panels of Figure 4.3 that for $\frac{\beta_s}{h_0(\theta)} \ll 1$, $\theta_c$ is most strongly influenced by the choice of interface relation and the value of $\frac{\rho_a}{\rho_c}$. Unlike $\frac{\beta_s}{h_0(\theta)}$, $\frac{\rho_a}{\rho_c}$ is already strongly influential on $\theta_c$ even for small values. Hence we will drop consideration of $\frac{\beta_s}{h_0(\theta)}$ beyond this point in order to focus on these other two, more influential, factors.

Interface relation and boundary amorphization. In Figure 4.4, we neglect slip-flow altogether in order to focus on the relationship between $\theta_c$, $\frac{\rho_a}{\rho_c}$, and the interface relation. This is motivated from the observation of Figure 4.3 that the influence of slip-flow on $\theta_c$
selection is quite weak at physically-expected values (and is even weaker for the collision cascade-informed interface relation), and we wish to minimize the dimensionality of our parameter space (for simplicity). This should not be taken as an assertion that slip-flow is not occurring, or that its influence is not necessarily relevant in other contexts; however, we are primarily interested in $\theta_c$-selection here.

We note that the experimental range of $\rho_a/\rho_c$ for Si is likely around 92-96% [87, 22]. Interestingly, sufficient $\rho_a/\rho_c$ is capable of strongly stabilizing the surface to perturbations, and for the 250eV Ar\(^+\) → Si system, this sufficient value appears to be (1) surprisingly small and (2) contingent upon modeling of the interfaces. The shapes of the resulting curves are also intriguing, which we will discuss below.

We will develop some intuition regarding the slope of $\theta_c(\rho_a/\rho_c)$ in the vicinity of $\rho_a/\rho_c = 1$. Like the post-amorphization swelling mechanism (IIS), the primary means by which the phase-change at the amorphous-crystalline interface stabilizes the free interface is through the modification of the bulk velocity field. In the case of IIS, the isotropic expansion of all parcels of matter simultaneously leads to material from the sides of “hills” being pushed into.

Figure 4.4. Using $\eta = 8.5$ GPa \(\cdot\) s, and values of $f_A D \approx 9.247 \times 10^{-3} \frac{1}{s}$, $f_A I \approx 3.365 \times 10^{-3} \frac{1}{s}$ for 250eV Ar\(^+\) → Si at $1.2 \times 10^1 \frac{\text{ions}}{\text{nm}^2 \cdot \text{s}}$. 
the bottoms of “valleys”; see Chapter 3, [96], or [50]. On the other hand, the phase-change boundary condition induces an inward-normal flow originating from the lower interface; when lower-interfacial “valleys” are vertically aligned with upper-interfacial “valleys”, this addition to the flow field will also tend to drive material into the bottom of the upper interfacial valley, stabilizing it against perturbations in a manner similar to that of IIS. Hence alignment of upper- and lower-interfacial valleys at all angles of incidence (i.e., the “vertical interfaces” relation) will lead to a somewhat stronger stabilization, while interfaces that misalign rapidly as a function of irradiation angle should see a weaker stabilization effect, such as the “diagonal interfaces” case. Indeed, we find

\[ \frac{\partial \theta_c}{\partial \left( \frac{\rho_a}{\rho_c} \right)} \bigg|_{\left( \frac{\rho_a}{\rho_c} \right) = 1, \text{vert}} \approx -40, \]  
\[ \left( \frac{\partial \theta_c}{\partial \left( \frac{\rho_a}{\rho_c} \right)} \right) \bigg|_{\left( \frac{\rho_a}{\rho_c} \right) = 1, \text{diag}} \approx -14.6, \]  
\[ \left( \frac{\partial \theta_c}{\partial \left( \frac{\rho_a}{\rho_c} \right)} \right) \bigg|_{\left( \frac{\rho_a}{\rho_c} \right) = 1, \text{CC}} \approx -24. \]

This is consistent with the additional stabilization effect, for small density drops, being due to the modification of the bulk flow field by the alignment of the interfaces. We also note that in Figure 4.4, the “diagonal interfaces” case predicts a very rapid approach to \( \theta_c \approx 70^\circ \) as \( \frac{\rho_a}{\rho_c} \) decreases. This is because the contribution of the phase-change boundary condition to stabilization strengthens with thinner films, all else being equal, and the diagonal case predicts zero thickness as \( \theta \to 90^\circ \), and is a further indication of the need to refine the treatment of the interfaces.

4.4.2. Comparison of theory and experiment

**Film thickness: 1keV Ar\(^+\) → Si.** Angle-dependent film thickness is perhaps the simplest point of comparison between the present analytical work and experimental measurements. Using our expression for \( h_0(\theta) \), Equation (4.28), we compare with the angle-dependent thickness inferred from fits to GISAXS data in [28]. It is immediately evident (and unsurprising) that the agreement is far superior to that given by either the assumption of permanently “vertical” interfaces or the “diagonal” interfaces whose film thickness is \( \propto \cos(\theta) \).
Figure 4.5. Film thickness prediction using theoretical expression $h_0(\theta)$ from Equation (4.28) compared to film thickness inferred from [28] for 1keV Ar$^+$ → Si and other interface relations.

**Wavelength selection: 250eV Ar$^+$ → Si.** Finally, we compare theoretical predictions to experimental wavelength measurements [43] for 250eV Ar$^+$ → Si at the same flux using the long-wave approximation of the linear dispersion relation, Equation (4.17). As has been seen in [32], the long-wave approximation is imperfect, and can disagree with the full-spectrum wavelength predictions even by $\sim 10$ nm (or roughly 50% in the case of [32]), with the approximation becoming worse near grazing incidence. The reason for this is intuitive: near grazing incidence, wavelengths are no longer necessarily large, hence the long-wave approximation breaks down. Nonetheless, we will make such a comparison in the absence of a full-spectrum result on a tentative basis, primarily as a means of studying qualitatively the effect of the phase-change boundary condition and the BCA-informed interface relation on $\lambda(\theta)$. Strikingly, the BCA-informed interface relation appears to provide superior agreement with $\lambda(\theta)$ when compared with either of the two simplified relations, especially far from grazing incidence (where the long-wave approximation is expected to be more accurate).

**Possible stabilization by amorphous-crystalline phase change: Ar$^+$ → Si and Xe$^+$ → Ge.** It is interesting to notice the form introduced into the linear dispersion relation
by the phase-change condition at the amorphous-crystalline boundary,

$$-rac{V}{h_0(\theta)} \left( 1 - \frac{\rho_a}{\rho_c} \right).$$

(4.43)

Since for the Si target of interest in the present work $\frac{\rho_a}{\rho_c} < 1$, this term is stabilizing for all $\theta$. Crucially, for erosion held constant, it strengthens with decreasing film thickness. At the same time, erosion, and the corresponding amorphization of fresh c-Si, should be faster with collision cascades nearer to the surface, as more power is deposited there [7, 8, 9]. In Figure 4.7, we consider the energy dependence of this term at normal incidence for two systems of interest across a range of energies: Ar$^+$ → Si and Xe$^+$ → Ge. In Ar$^+$ → Si, a strong suppression of ripple growth in about the 1.3keV-20keV range has been experimentally observed [22].

A similar weakening of pattern formation occurs near 1.2keV-2keV Xe$^+$ → Ge [57]: below this energy range, $\theta = 65^\circ$ is sufficient to induce ripples. By 2keV, patterns are completely suppressed at 65$^\circ$, but return when the incidence angle is increased to 75$^\circ$, consistent with an
increase in some stabilization mechanism, such as IIS, or the decrease of some destabilization mechanism such as APF with energy. For lack of experimental work, it is unclear what happens for $\text{Xe}^+ \rightarrow \text{Ge}$ at even higher energies; however, based on Figure 4.7, we might anticipate that, like for $\text{Ar}^+ \rightarrow \text{Si}$, patterns return at higher energies.

Interestingly, Figure 4.7 shows that, for normal-incidence irradiation, $\frac{Y(\theta)}{h_0} = \frac{V(\theta)}{h_0(\theta)}$ is increasing in the vicinity of strong ripple suppression for both ion-target combinations and decreasing near the energy level for which patterns seemingly return [56] for $\text{Ar}^+ \rightarrow \text{Si}$. These observations are qualitatively consistent with the present findings of an additional stabilization mechanism that strengthens with erosion and weakens with increased film thickness: indeed, the sputter yield begins to decrease as $E$ increases into the electronic-stopping regime [79, 62], while the depth of damage continues to grow. This appears to bolster the argument that modification of the bulk flow field due to phase-change at the amorphous-crystalline boundary is an important and hitherto-unappreciated stabilization mechanism in irradiated thin films.

Somewhat more specifically, we consider the well-known Davis scaling of in-plane compressive stress with energy as $E^{-7/6}$ [97]. In short, this scaling is applicable in the case of films which accumulate thickness due to ion-implantation slowly, as is the case for our low-energy systems of interest; the full details are substantially more complex than this and we refer the reader to the extensive discussion in [97].

We also notice from the discussion of $\eta$, and the observation that depth $d \sim E^{-4} - E^{-6}$ [98], that compressive stress $\propto A_D \eta \sim E^{-17/30} - E^{-23/30}$. Accordingly, we take the midpoint of this range and consider the very simple case

$$A_D \propto E^{-2/3}, A_I = 0,$$

with constant of proportionality $3 \times 10^{-2}$, in order to make a very preliminary assessment of what sort of qualitative behavior might be induced by this scaling. We also take $A_I = 0$ because we are not yet certain of parameter values for $A_I$ or $A_D$, nor the scaling of $A_I$ with energy. Regardless of the precise value, we find that $A_D = E^{-2/3}$ combined with the dispersion relation (4.17) above and uncertainty in the value of $\frac{\rho_a}{\rho_c}$ can easily lead to a sudden
Figure 4.7. Here, we consider the energy-dependence of $\frac{V}{h_0 f}$, which informs the strength of the phase-change induced stabilization, (4.43). In the above, higher values are associated with stronger stabilization, hence an increased value of $\theta_c$. Notice the non-monotonicity, and that the value increases with higher energies on the interval of roughly 100eV-7keV for Ar$^+$ → Si. As we discuss in the main text, a term with this behavior could possibly account for the strong suppression of patterns for Ar$^+$ → Si between about 1.2 and 20keV, and the return of patterns at even higher energies. Curiously, the effect is quite weak for Ne$^+$ projectiles, suggesting that some other mechanism will be needed to account for the lack of pattern-formation observed by [57]. We will revisit this topic in Chapter 5.

increase in $\theta_c(E)$ in the vicinity of 1.2keV, as is experimentally observed. This is because the phase-change term $\propto \frac{V}{h_0(\theta)}$ is at its strongest for Si between about 1keV and 7keV (see Figure 4.7), and $A_D$ is actually a decaying function of energy— hence it is unsurprising (and, indeed, rather generic) that two non-monotonic functions should cross and exchange dominance. While the Davis scaling is seemingly reliable for energies below about 1-2keV, we cannot speculate as to the scaling of APF with energy much outside of this range. However, it is noteworthy that the lower limit of $\theta_c$ is evidently around 30°, as is experimentally observed [56] for 40keV Ar$^+$ → Si. It is not difficult to imagine that as energy increases, higher-energy anisotropic plastic flow, described by [60, 61, 39] and related to melt-cycles, becomes active, and drives $\theta_c$ lower due to increased destabilization. Discussion in [32, 38,
suggests that this may happen for some materials around 100keV, although it is expected that the threshold for melt-cycles to begin is material-dependent [79, 62], and appears to scale with electronic stopping power [39], and Ar$^+$ → Si was not explicitly studied in the referenced work.

Figure 4.8. Here, we assume the Davis scaling $A_D \propto E^{-2/3}$ and consider a flux $f = \frac{\text{ion}}{\text{nm}^2 \cdot \text{s}}$. For Ar$^+$ → Si with this scaling, the existence of a region where the stabilizing phase-change term in (4.17) becomes dominant over APF is generic.

**Remark on the Davis scaling.** We have considered the Davis scaling in the above, since it appears to be consistent with ion implantation at low energies [97, 33]. Of course, the question remains whether the present model can possibly explain the apparent return of pattern formation at higher energies [22]. In fact, the Davis scaling may also explain this as well: if the ratio $\frac{R}{j} \gg 1$, stress scales as $\sqrt{E}$ rather than $E^{-7/6}$, where $R$ is net implanting flux and $j$ is net bombarding flux (using the notation of [97]). Since $\frac{R}{j}$ is a measure of the growth-rate of the film thickness per unit ion flux, and the sputter rate per film thickness begins to decay for increasing energy for some materials (such as Si), it is possible that stress-based effects could strengthen as energy increases into the electronic stopping regime.
leading to a return of patterns. This would lead to a relatively fast accumulation of material per unit bombarding flux, which could lead to a transition from the $R_j \ll 1$ limit (so that in-plane stress scales like $E^{-7/6}$) to the $R_j \gg 1$ limit, where in-plane stress scales like $\sqrt{E}$. We consider this an intriguing area for future study, as it may provide a means for some system parameters (such as $A_I$ or $A_D$) to exhibit non-monotonicity with respect to energy.

4.5. Discussion

4.5.1. Primary results

1: On the amorphous-crystalline interface and its boundary conditions. It has been noted elsewhere that seemingly-innocuous modeling choices about the amorphous-crystalline interface can lead to large differences in the theoretical prediction of $\theta_c$ (Chapter 3). In the present work, we have sought to model this interface with greater attention. In particular, we have connected the expressions for the angle-dependent film thickness $h_0(\theta)$ and the lateral shift $x_0(\theta)$ directly to parameter values easily obtained from SRIM for the first time. Our theoretical $h_0(\theta)$ correctly captures the angle-dependence of film thickness as experimentally inferred from previous GISAXS work in the 1keV $\text{Ar}^+ \rightarrow \text{Si}$ system [28], as well as [20]. Then, we have considered a phase-change boundary condition at the amorphous-crystalline interface, appropriate for studying the effect of the instantaneous change in density $\rho_c$ to $\rho_a$ as the amorphous layer erodes into the crystalline substrate.

We have shown that with plausible values of $\frac{\rho_a}{\rho_c}$, the effect of phase-change at the amorphous-crystalline interface may account for as much as 5-10° in the 250eV $\text{Ar}^+ \rightarrow \text{Si}$ system. This suggests that the shape of the amorphous-crystalline interface, and the density-drop across it, may substantially alter $\theta_c$ selection. Hence, while post-amorphization swelling, when modeled as a constant across the film, is seemingly a weak contributor to $\theta_c$ selection in the 250eV $\text{Ar}^+ \rightarrow \text{Si}$ system, the instantaneous swelling associated with near-
boundary phase-change is expected to be important. These findings clearly prompt further study of the mathematical treatment of the amorphous-crystalline interface and additional experimental work on the value of \( \frac{\rho_a}{\rho_c} \) for materials of interest.

2: Parsimony in critical angle, wavelength, film thickness, stress coefficients, and viscosity. Having refined our theoretical treatment of the amorphous-crystalline interface, we then seek theoretical predictions for angle-dependent wavelength \( \lambda(\theta) \). Although currently restricted to the long-wave approximation of the linear dispersion relation, we find that agreement with \( \lambda(\theta) \) is improved for our treatment of the amorphous-crystalline interface when compared with two commonly used assumptions. The present work therefore obtains reasonable agreement with \( \lambda(\theta) \) and \( \theta_c \) for the 250eV Ar\(^+\) \( \rightarrow \) Si system using only two experimentally-informed parameter values, \( fA_D, fA_I \), and an estimated value of \( \eta(0) \), all of which could be obtained for any other projectile-target combination at any other energy and flux if desired. We note that the value of \( \eta(0) \) used here is within about an order of magnitude of other estimates for similar systems within the literature when scaled down to the present flux and energy by the argument of [93, 43]. The need for only three experimentally-accessible parameter values to substantially improve agreement in modeling of \( \lambda(\theta) \) and \( \theta_c \) suggests a highly parsimonious model that approaches a “full picture” of the relevant phenomena. That the selection of \( \lambda(\theta) \) and \( \theta_c \) from the present work is rather sensitive to parameter values also suggests that our parameter values are expected to be quite close to “true” values, hence all aspects of the present work are “mutually-assuring”. Hence our model appears to represent a potential candidate for the “unifying model” capable of connecting a small number of experimentally-accessible parameter values directly to experimental outcomes.

4.5.2. Secondary results

1: The importance of \( \eta \) for linear stability analysis. In previous work on APF and IIS, both individually [32] and together (Chapter 3), it was sufficient to estimate \( fA_D\eta \) and
\[ fA_I \eta \] in order to extract estimates of \( \theta_c \). This was because the long-wave dispersion relations for APF and IIS both scaled with \( fA_D \) and \( fA_I \) and were the only terms involved in the dispersion relation; hence multiplication by \( \eta \) would allow the use of experimentally-accessible quantities \( fA_D \eta \) and \( fA_I \eta \) instead of the difficult-to-obtain coefficients \( fA_D, fA_I \) alone. In the present work, the appearance of a term in the dispersion relation unrelated to stress foils this approach, and we are forced to approximate \( \eta \) directly. However, approximation of \( \eta \) for a given irradiated system has proven difficult and costly \([93, 95, 18, 14]\), and the MD-oriented approach of \([93]\) apparently leads to significant disagreement with the relaxation times observed in the irradiation experiments of \([44, 47]\) at 250eV and fluxes of 12 and \( 32 \text{ ions nm}^2 \text{ s}^{-1} \).

2: Negligibility of slip-flow. We have also considered how deviation from the typical no-slip, no-penetration boundary conditions at the lower interface affect \( \theta_c \). While the normal-incidence flow field induced by boundary amorphization is evidently important, the inclusion of a hypothetical, non-zero slip length is apparently unimportant at realistic values of the slip length. In the case of unrealistically large values of the slip length, the effect of slip flow is simply to introduce an asymptote in \( \theta_c \left( \frac{\beta_s h_0}{h_0} \right) \). This might be explained intuitively by the observation that for very large values of \( \frac{\beta_s h_0}{h_0} \) in the long-wave limit, the horizontal component of the velocity field dominates all others, restoring its linear profile in the film-depth which was originally associated with APF alone \([32]\). Hence we might anticipate that an excessive slip-length would tend to produce \( \theta_c \) comparable to APF acting in isolation. Indeed, with vertically-aligned interfaces, \( \theta_c = 45^\circ \) is recovered for large \( \frac{\beta_s h_0}{h_0} \) in the case of no boundary amorphization (Figure 4.3); for diagonally-aligned interfaces, \( \theta_c = 35^\circ \). This is in line with our intuition, and characterizes slip flow primarily as a modifier of APF. At physically-reasonable values of \( \frac{\beta_s h_0}{h_0} \), it appears that slip flow exerts minimal effect on \( \theta_c \). We therefore anticipate that it may be safely disregarded in future efforts.

3: Relationship with a class of crater function models. The effect of a dynamically-adjusting interface has been considered heuristically elsewhere \([20, 22, 23]\) in the context of
a class of crater function models [17, 6]. In that work, as here, it is found that a coefficient in the evolution equation of the surface associated with the angle-dependent film thickness exerts a stabilizing effect on the surface. However, as we will discuss, the underlying physical causes are quite different: as with any crater function approach, the bulk flow field is essentially neglected, and certain aspects of the collision cascade are simply projected onto the evolution equation describing the free interface. In contrast, our results emerge from a linearization of the entire flow field that emerges through the interactions of APF, IIS, viscous relaxation and the interfacial phase-change. As is well-known, the flow field can elude simple characterization and may substantially modify linear stability ([31, 32, 33, 48, 50]; see also Supplementary Figures of [18]). It may also act alongside erosion and redistribution to inform $\theta_c$ and $\lambda(\theta)$ selection [28]. They must therefore be handled with care.

In [20, 22, 23], the effect of the interface shape is implemented by imposing the direct modification of the kinematic condition at the free interface, while standard no-slip and no-penetration conditions are imposed at the amorphous-crystalline interface. Within the modified kinematic condition, an average, lateral “drift velocity” is computed across the film depth, and a curvature-dependent coefficient is introduced based on this drift velocity; we refer to Equation (11) of [20]. This lateral drift velocity, being the lateral displacements of all atoms averaged throughout the film depth, also strengthens with thinner films, all else being equal, like our Equation (4.43), while being rooted in a different physical theory. At the same time, in Section 2.3 of [20], the inward-directed transfer of mass from the crystalline substrate is considered, and it is suggested that this effect is negligible except at grazing incidence, where it is responsible for inducing the “ripple-rotation” phenomenon.

This approach has been questioned elsewhere [6], as the kinematic condition is merely a statement of local velocity at an interface, which would not be ordinarily expected to contain any explicit dependence on the film thickness below that interface. By way of contrast, the present approach only indirectly modifies the kinematic condition at the upper interface by way of the fully-resolved velocity field in between the two interfaces, where the velocity field is modified simply by the imposition of a phase-change boundary condition at
the amorphous-crystalline interface, and the approach of directly modifying the kinematic condition implicitly makes assumptions about the flow field occurring within the amorphous bulk. Within this framework, stress-driven flow and potentially even defect dynamics (in the style of [46, 42]) can be taken into account in a straightforward way, and the resulting flow fields are naturally taken into account. As has been seen elsewhere, the flow fields resulting from ion-bombardment may be highly complex and localized [48, 18, 52], leading to difficulties in the top-down imposition of a single, simple form of averaged velocity for the purposes of computing the modified kinematic condition postulated in [20, 22, 23].

Finally, we note subtle differences between our model and that of [20]. In that work, it is claimed that modification of the lateral component of the velocity field is a strong contributor to stabilization due to its interaction with the free interface, while inward-directed mass transfer from the amorphous-crystalline interface is unimportant far from grazing incidence. In fact, this is quite contrary to the claims of the present work: here, we have found that modification of the lateral component of the velocity field via the incorporation of a slip boundary condition has virtually no effect on $\theta_c$ for physically-expected amounts of slip, while the inward-normal modification of the bulk flow field due to instantaneous phase-change— analogous to the inward-directed mass transport described in Section 2.3 of [20]— is of extreme importance at all angles. We have found additional stabilization due to this inward-directed mass transport, while [20] has found additional stabilization due to lateral mass transport. In both cases, each claimed effect is strongest for thinner films, as discussed in our Results section, leading to a high degree of apparent correlation, while the underlying mechanisms are fundamentally at odds. We anticipate that further experimental-theoretical studies will eventually discern which of the two approaches is correct, perhaps by contrasting predictions of ripple velocity, direction of propagation, and transition angle, for which there is some disagreement between purely erosive-redistributive theories and experimental work [99, 100]; or by GISAXS studies examining ripple growth rates [28], which appear to necessitate the inclusion of stress effects at least for low energies.
4.5.3. Future work and open questions

1: Spatial resolution of stress. We have considered the influence of the collision cascade on the shape of the amorphous-crystalline interface, and we have shown that the interfacial geometry interacts with the ongoing phase-change at that interface to induce a significant additional stabilization effect. The apparent necessity of exercising caution with the interface, previously regarded as an innocuous modeling choice, has now been shown to hold unexpected importance. This naturally engenders questions of what other aspects of the stress-driven approach to ion-irradiated thin films may benefit from a more physically-realistic treatment. In particular, while previous work ([33] and Chapter 3) has shown that a simple assumption about the spatial homogeneity of stress may or may not influence the selection of \( \theta_c \) and \( \lambda(\theta) \) in a manner that, again, seems to depend on modeling choices, both works considered only uniaxial inhomogeneity. A natural follow-up is to consider the full spatial resolution of stress, completing an analysis for the amorphous bulk analogous to that performed by [9, 25] for the surface. This is potentially of interest, as the Bradley-Harper instability only becomes apparent when power deposition is formulated non-locally as an integral: without the integral formulation, it would not appear that “hills” and “valleys” should sputter at different rates. Because no such non-local treatment of inhomogeneous stress has been considered within the family of stress-oriented models, we consider this, and any resulting non-local phenomena, of high research priority.

2: Application to other experimental systems. Although we have focused on 250eV \( \text{Ar}^+ \rightarrow \text{Si} \) here, with a brief consideration of film thickness for 1keV \( \text{Ar}^+ \rightarrow \text{Si} \), the general approach of the present work (and related work in Chapter 3) should be applicable to any irradiated, amorphous target. In particular, we consider the collection of angle-dependent stress data for 250eV \( \text{Kr}^+ \) and \( \text{Xe}^+ \rightarrow \text{Si} \) to be of significant interest, so that we may obtain further data on the role of ion mass and collision cascade details in the selection of \( \theta_c \) and \( \lambda(\theta) \). Angle-dependent stress data for \( \text{Ar}^+ \rightarrow \text{Si} \) across the 250eV-40keV range would also be greatly useful. We note that the hypothesized, stress-induced destabilizing mechanism in the
present work is that of anisotropic plastic flow, whose in-plane stress component is expected to change from compressive to tensile at around 45°. Experimental work to validate or reject the presence of such a mechanism is clearly of value, especially because not all stress-driven mechanisms are capable of inducing such a sign change; see [33] and the Appendix to Chapter 3. It would also be very interesting if it turned out that the component of angle-independent stress associated with ion-induced swelling were much stronger than expected, or exhibited a nonlinear scaling in strength with energy for the same projectile-target pair.
Chapter 5
Spatially-resolved isotropic swelling

5.1. Introduction

In this Chapter, we seek to make full use of the framework of Chapter 3, where formulas were derived for growth rates expressible in terms of arbitrarily distributions for each of IIS and APF. Until this point, we have made simplified assumptions about these distributions: that they are constant throughout the amorphous layer, or that they vary uniaxially in the downbeam direction.

However, we have no particular reason to expect that either of these simplifications should be physically accurate, and in Chapter 4, we showed that the details of the collision cascade can be used to directly inform details of the thin-film model that had been previously subject to various *ad hoc* (and not necessarily physical) assumptions. We then showed that these assumptions can significantly impact the ability to obtain agreement between theory and experiment. We take this as an invitation to include even more simulation-informed details into the modeling, seeking to push the limits of the stress-based approach. We are in part motivated by the observation that the well-known Bradley-Harper instability does not become apparent until the energy deposited is “spatially-resolved”: that is, taken to vary biaxially in downbeam and crossbeam coordinates, and then integrated over the irradiated surface. When this is done, it becomes apparent that the power deposited at valleys is greater than that at hilltops, leading to an instability. This naturally prompts the linearization of the same type of integral describing deposited energy (or other quantities) within the amorphous bulk.

In particular, we will seek to use BCA simulations [78] to inform both the amorphous-crystalline geometry (the interface relation) and the spatial distribution of stress, which
we will infer directly from the collision cascade details, no longer relying on simplifying assumptions. While this will introduce some difficulty, we find that, remarkably, the long-wave linear growth rate (linear dispersion relation) is still expressible in closed form. We also use the full collision cascade details to validate, in greater rigor, the approach of Chapter 4, where the amorphous-crystalline geometry was explored in the long-wave limit.

5.2. Preliminaries

We very briefly summarize the mathematical model at the center of the present work, whose development has been shown in Chapters 2-4. The validity of this model will be assumed, and the resulting linear growth rate (linear dispersion relation) is stated without further justification based on our earlier work (Chapter 4), and will constitute a staging point for the analysis of interest in the present work.

**Bulk description.** As with any viscous continuum model, we begin with statements of bulk mass and linear momentum conservation,

\[
\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \vec{v}) = 0, \quad (5.1)
\]

\[
\nabla \cdot \mathbf{T} = 0,
\]

where we define our stress tensor

\[
\mathbf{T} = -p \mathbf{I} + 2\eta \{ \dot{\mathbf{E}} - \dot{\mathbf{E}}_b \}
\]

\[
\dot{\mathbf{E}} = \frac{1}{2} (\nabla \vec{v} + \nabla \vec{v}^T)
\]

\[
\dot{\mathbf{E}}_b = f A_D \tau (z; g, h) \mathbf{D}(\theta)
\]

\[
\mathbf{D}(\theta) = \begin{bmatrix}
\frac{3}{2} \cos(2\theta) & -\frac{1}{2} & 0 & \frac{3}{2} \sin(2\theta) \\
0 & 1 & 0 & 0 \\
\frac{3}{2} \sin(2\theta) & 0 & -\frac{3}{2} \cos(2\theta) & -\frac{1}{2}
\end{bmatrix}.
\]

In the above, we have not exploited the common simplification to \( \nabla \cdot \vec{v} = 0 \), because we will allow for the possibility of localized changes in density. These are incorporated via the
equation of state

\[ \rho(\Delta) = \frac{\rho_a}{1 + \Delta}, \quad (5.4) \]

where \( \Delta \) is characterized by

\[ \frac{\partial \Delta}{\partial t} + \vec{v} \cdot \nabla \Delta = \alpha(z; g, h), \quad (5.5) \]

a forced advection equation which tracks the damage accumulated by a parcel of matter as it dwells within the irradiated film. Hence our model is “almost” Stokes flow, except that we permit localized, radiation-induced loss of density, and an anisotropic stress-free strain rate (caused by the ion beam), both of which act to modify the velocity field, evolving stress, and, ultimately, control a morphological instability.

**Boundary conditions.** At \( z=h \), we have

\[ v_{I,h} = \vec{v} \cdot \hat{n} - V \frac{\rho_c}{\rho} \]

\[ [T] \cdot \hat{n} = -\gamma \kappa \hat{n}, \quad (5.6) \]

and at \( z=g \), we have

\[ \Delta = 0 \]

\[ \vec{v} \cdot \hat{t} = \beta_s \hat{n} \cdot \nabla (\vec{v} \cdot \hat{t}) \]

\[ [\rho \vec{v}] \cdot \hat{n} = [\rho] v_I. \quad (5.7) \]

**Linear stability analysis.** As before, we perform a linear stability analysis by converting the above equations to a moving frame via

\[ z \rightarrow z - V t \]

\[ h \rightarrow h - V t \]

\[ g \rightarrow g - V t \]

\[ \vec{v} \rightarrow \vec{v} - V \hat{k} \]

\[ v_{I,h} \rightarrow v_{I,h} - V (\hat{k} \cdot \hat{n}), \quad (5.8) \]

which simply tracks the average downward velocity of the film due to erosion. Then, we seek *steady-state* solutions \( \Phi_0(z) \) for each of the bulk fields \( \Phi = [\rho, u, w, p, \Delta] \) under the
steady-state assumptions that $g = 0$, $h = h_0$, $\frac{\partial}{\partial t} \to 0$, and $\frac{\partial}{\partial x} \to 0$. We then expand about a small perturbation,

$$h(x, t) \to h_0 + \epsilon h_1(x, t)$$
$$g(x, t) \to g_0 + \epsilon g_1(x, t)$$,  \hspace{1cm} (5.9)
$$\Phi(x, z, t) \to \Phi_0(z) + \epsilon \Phi_1(x, z, t)$$

and then again in normal modes,

$$\Phi_1(x, z, t) \to \Phi_1(z) \exp(\sigma t + ik x).$$  \hspace{1cm} (5.10)

The expansion

$$g_1 \to \tilde{g}_1 \exp(\sigma t + ik x)$$
$$h_1 \to \tilde{h}_1 \exp(\sigma t + ik x)$$

leads to the substitution

$$\frac{\tilde{g}_1}{\tilde{h}_1} \to \exp(-ik x_0(\theta)),$$  \hspace{1cm} (5.12)

which will appear in the main linear stability result (see Chapters 3 and 4; [48, 50]).

**General linear stability result.** Solving the resulting system of ordinary differential equations, we finally obtain a general result, which was discussed previously in Chapter 4:

$$\sigma = (\sigma_{10} k + f A_I \sigma_{11}) k + (\sigma_{20} k + f A_I \sigma_{12}) k^2 + O(k^3),$$  \hspace{1cm} (5.13)

where

$$\sigma_{10} = \frac{-2 f A_D i D_{13}}{h_1} \int_0^{h_0} \left( \int_0^z \tau_c(z_1) dz_1 + \beta_s (\tilde{g}_1 \tau_{0, z}(0) + \tau_c(0)) - \tilde{g}_1 \tau_0(0) \right) dz$$
$$+ 2 f A_D i \beta_s D_{13} \tau_0(0) \frac{\tilde{g}_1}{\tilde{h}_1}$$
$$+ \sigma_{10} \left( 1 - \frac{\rho_c}{\rho_a} \right) \left( \frac{\tilde{g}_1}{\tilde{h}_1} \right)$$
$$- 2 f A_D i D_{13} \left( \int_0^{h_0} \tau_0(z) dz + \beta_s \tau_0(0) \right);$$  \hspace{1cm} (5.14)

$$\sigma_{11} = \left( 1 - \frac{\rho_c}{\rho_a} \right) \sigma_{11} \left( \frac{\tilde{g}_1}{\tilde{h}_1} \right);$$  \hspace{1cm} (5.15)
and

\[
\sigma_{20} = 2f A_D (D_{11} - D_{33}) \int_0^{h_0} \int_0^z \int_0^{z_1} \tau_\epsilon(z_2) d z_2 d z_1 d z
- \frac{i}{h_1} \left( \frac{c_9 h_0^2}{2} + c_{10} h_0 \right) + \left( 1 - \frac{\rho_c}{\rho_a} \right) \sigma_{20} \left( \frac{\rho_c}{\rho_a} \right),
\]  

(5.16)

where

\[
c_9 = -2i h_1 f A_D \tau_0(h_0) \left( D_{11} - D_{33} \right) - 2f A_D i \left( D_{11} - D_{33} \right) \int_0^{h_0} \tau_\epsilon(z) d z;
\]

\[
c_{10} = i \tilde{g}_1 V \left( 1 - \frac{\rho_c}{\rho_a} \right) + \beta_s c_9;
\]

and

\[
\sigma_{21} = \frac{1}{h_1} \int_0^{h_0} \left( \frac{\sigma_{20,0}}{V} \left( \frac{\rho_c}{\rho_a} \right) \right) \left[ \int_0^z \alpha_01(z_1) d z_1 - \tilde{g}_1 \alpha_{01}(0) \right]
+ \int_0^z \int_0^{z_1} \alpha_01(z_2) d z_2 d z_1 - i c_{13} z - i c_{14}
+ \tilde{w}_{120,0} \left( \frac{\rho_c}{\rho_a} \right) \alpha_{01}(z) \right) d z + \left( 1 - \frac{\rho_c}{\rho_a} \right) \sigma_{21} \left( \frac{\tilde{g}_1}{h_1} \right),
\]  

(5.18)

where

\[
c_{13} = -2i h_1 \alpha_{01}(h_0) - 2i \int_0^{h_0} \alpha_01(z) d z + i \tilde{g}_1 \alpha_{01}(0)
\]

\[
c_{14} = \beta_s \left[ c_{13} + i \tilde{g}_1 \alpha_{01}(0) \right];
\]

\[
\sigma_{20,0} = \frac{h_0 V \left( 1 - \frac{\rho_c}{\rho_a} \right) \left( \frac{\tilde{g}_1}{h_1} \right)}{\left( 1 - \left( 1 - \frac{\rho_c}{\rho_a} \right) \left( \frac{\tilde{g}_1}{h_1} \right) \right)}
\]

\[
\tilde{w}_{120,0} = \tilde{g}_1 V \left( 1 - \frac{\rho_c}{\rho_a} \right) z + \left( 1 - \frac{\rho_c}{\rho_a} \right) \sigma_{20,0} \tilde{g}_1.
\]

While the above is obtained by solving the system of equations outlined in the Appendix, we do not present a full solution as we did in Chapter 3, as the method of solution is essentially the same.

Remarks. Although the above is a summary of our own previous work (Chapter 4), we will here significantly extend that work by considering the connection of the above form directly to details of the collision cascade. This requires the determination of the functions \( \tau_0, \alpha_{01}, \tau_\epsilon \) and \( \alpha_{01} \) in terms of ion-implantation parameters which may be obtained via simulation software, such as SRIM [78]. We will also validate an earlier approach to computing \( h_0, \frac{\tilde{g}_1}{h_1} \) in terms of the collision cascade.
We have presented the above result in full generality; in what follows, we will neglect slip-flow (hence $\beta_s \to 0$), and the integrals will be evaluated, leading to a much simpler final expression.

5.3. Analysis

Our approach will be similar to that of [9, 25] insofar as we consider the linearization of an integral representing some quantity (as-yet unspecified) $q$ deposited at a point, below. Then, whereas [9, 25] were interested in the deposited power reaching the surface, leading to an erosion rate, we are interested in the deposition of $q$ at points in the bulk, leading to spatial inhomogeneity in the modification of the stress field by incoming ions. We will also consider the level curves of this deposition, which will suggest the shape of the amorphous-crystalline interface. We consider the density

$$P(x, z) = \int_{-\infty}^{\infty} \left( \cos(\theta) + h_X \sin(\theta) \right) E_D(x, z; X, h(X)) dX \quad [\text{=} \frac{q}{\text{ion} \cdot \text{nm}}]$$

(5.20)

which computes the volume-density of $q$ at a point $(x,z)$ due to all ions entering across the entire surface $h(X)$ with fluence $1 \text{ ion nm}^{-2}$. In the above and throughout this work, $[\text{=}]$ denotes “units of”; tracking units will inform some of the discussion. As before, $h_X$ denotes $\frac{\partial h}{\partial X}$. Note that this density’s units are, equivalently,

$$\frac{q/\text{nm}^3}{\text{ion/\text{nm}^2}},$$

(5.21)
a volume-density of $q$ per fluence.

In the above, $E_D$ denotes the function that describes the distribution of $q$ deposited at the point $(x, z)$ due to a single unit-fluence entering the film at the point $(X, h(X))$, where the slope is $h_X$. We will take the standard Sigmund [7, 8] model of Gaussian-distributed $q$ per ion impact, imagining that, whatever $q$ is, it should be distributed similarly to the energy deposition, recoils, or ions, all of which are at least approximately bivariate-Gaussian.
in the distribution that results from a single ion. Hence we consider

\[
E_D(x, z; X, h(X)) = \frac{1}{2\pi \alpha \beta} \exp \left( -\frac{[(x - X) \sin(\theta) - (z - h(X)) \cos(\theta) - a]^2}{2\alpha^2} 
-\frac{[(x - X) \cos(\theta) + (z - h(X)) \sin(\theta)]^2}{2\beta^2} \right) \left[=\right] \frac{q}{\text{ion} \cdot \text{nm}^2},
\]  

(5.22)

where \(\alpha\) and \(\beta\) are downbeam and crossbeam standard deviations respectively, and \(a\) is mean penetration depth in the downbeam direction. Despite borrowing the common notation for energy deposition, ion implantation, or the recoil distribution, we intend to consider the distribution itself for the moment, without yet assigning a specific physical quantity to be distributed.

The integral above, Equation (5.20), is clearly intractable analytically for arbitrary \(h\), so we instead consider the simplification \(h(X) = h_0 + \epsilon h_1(X)\), the “small perturbations” assumption, which is suitable for the present linear stability work. Then, we apply the usual expansion in normal modes via \(h_1(X) \rightarrow \tilde{h}_1 \exp(\sigma t + ikX)\). We provide full details of the calculation in the Appendix; they are straightforward but somewhat tedious. We point out that \(X\) denotes the surface coordinate, while \(x\) denotes the bulk coordinate, hence the expansion is performed in terms of \(X\). This notation will become important in the calculation described in the Appendix.

**Localization of intensity.** From the linearization of the density

\[
P(x, z) = P_0(z) + \epsilon \tilde{h}_1 \exp(\sigma t + ikx) P_1(z) + O(\epsilon^2),
\]

(5.23)

we have obtained the “small-amplitudes” volume-density of \(q\) in the bulk with the forms

\[
P_0 = \frac{\cos(\theta)}{2\pi \alpha \beta} \left[\sqrt{\pi} \exp \left( \frac{\tilde{B}^2}{4A} - C \right) \right],
\]

\[
P_1 = \frac{\cos(\theta)}{2\sqrt{\pi} A^3 \beta^3} \exp \left( \frac{B^2}{4A} - C \right) \left( c_1 - \frac{Bc_2}{2A} \right)
\]

(5.24)

where \(c_1 = c_1(z)\), \(B = B(z)\), \(C = C(z)\). Although these parameters are not of immediate importance (see the Appendix for their definitions), it is interesting to note that \(P_0\) is simply a Gaussian in \(z\),

\[
\exp \left( \frac{\tilde{B}^2}{4A} - C \right) = \exp \left( -\frac{(z - h_0 + a \cos(\theta))^2}{2(\alpha^2 \cos^2(\theta) + \beta^2 \sin^2(\theta))} \right),
\]

(5.25)
and $P_1$ is simply the product of a Gaussian and an affine-linear function, each in $z$. In the general linear stability result (Equations (5.14)-(5.19)), we apply the above by simply equating

$$A_I\alpha_{01}(z) = K_I P_0(z)$$
$$A_I\alpha_{\epsilon 1}(z) = K_I \hat{h}_1 P_1(z).$$

(5.26)

This assigns to the arbitrary spatial variation in our general linear stability result (Equation (5.37)) the spatial variation generated by the distribution $E_D$ per ion, and $K_I$ is a conversion factor that converts the volume-density of $q$ into local volumization. This is necessary because $\alpha_{01}$ and $\alpha_{\epsilon 1}$ are dimensionless, and $A_I$ has units $\text{nm}^2$ ion$^{-1}$, while $P$ has units of $\frac{q}{\text{ion-nm}}$. A natural interpretation of this is simply that $K_I$ converts $\frac{q}{\text{nm}^3}$ into $\frac{\text{nm}^2}{\text{ion}}$; that is,

$$K_I \; [\text{=} \; \frac{\text{nm}^3}{q}].$$

(5.27)

Then, multiplication by flux $f \; [\text{=} \; \frac{\text{ions}}{\text{nm}^2 \cdot s}]$ naturally restores the familiar parameter group $fA_I \; [\text{=} \; \frac{1}{s}]$.

Hence the parameter values $a, \alpha, \beta$ obtained from SRIM (or other means) fully characterize the stress distribution generated by the irradiation of the surface $h(X)$ via the integral (Equation (5.20)), and thereby the linear stability via the dispersion relation (Equations (5.14)-(5.19)). This immediately imparts a novel component of material specificity to our analysis which may be directly tied to aspects of the irradiation process, once we have correctly identified which components of that process result in APF and which result in IIS.

Of course, assigning strain rate $A_I$ this way naturally engenders the question of how $A_D$ may be assigned, since we have taken APF to be spatially-constant in the present analysis. The natural interpretation is simply that a single unit fluence induces a volume-density $\propto \frac{1}{\text{ion-nm}}$, so that aggregate (but homogeneous) APF throughout the film is a response to this uniform density. That is,

$$A_D\tau_0(z) = K_D \cdot 1$$
$$A_D\tau_1(z) = 0$$

(5.28)

in the present work. We have chosen to take APF spatially-homogeneous here for convenience, but also because, as will be shown, spatially-inhomogeneous APF will be unnecessary.
to make substantial progress. For details of spatially-inhomogeneous APF based on the same Gaussian ellipsoidal construction as described above for IIS, we refer the reader to the Appendix, where they have been recorded for posterity.

**Amorphous-crystalline interface.** We have previously considered the derivation of the amorphous-crystalline interface relation from an “ellipse-dragging” argument in Chapter 4. This argument made two assumptions: (1) that the interface relation could be localized to a single collision cascade due to a slowly-varying surface, and (2) that the amorphization threshold is simply where the deposited power falls below 5% of the peak (i.e., outside of two standard deviations of the distribution). Here, we will loosen the first assumption. Because our linear stability analysis involves computing the deposition of arbitrary (but Gaussian-distributed) quantity $q$ non-locally, we no longer require the long-wave limit to allow localization of interface selection to a single ellipse. We will therefore consider two standard deviations of the distribution generated *non-locally* in order to compute an interface relation valid for any wavenumber $k$. From the linearization of Equation (5.20) we seek level curves

$$P(x, z) = P_0(z) + \epsilon \tilde{h}_1 \exp(\sigma t + ikx) P_1(z) + O(\epsilon^2) = P_c, \quad (5.29)$$

where $P_c$ is some amorphization threshold, such that

$$z = g = g_0 + \epsilon \tilde{g}_1 \exp(\sigma t + ikx), \quad (5.30)$$

and we will solve for $g_0$ and $\tilde{g}_1$. For our purposes, we first establish $P_c$ by computing the central moments of the linearized density

$$E[z] = \int_{-\infty}^{\infty} z \left( P_0(z) + \epsilon \tilde{h}_1 \exp(\sigma t + ikx) P_1(z) \right) dz,$$

$$E[z^2] = \int_{-\infty}^{\infty} z^2 \left( P_0(z) + \epsilon \tilde{h}_1 \exp(\sigma t + ikx) P_1(z) \right) dz. \quad (5.31)$$

Then $\mu = E[z]$ and $\sigma = \sqrt{E[z^2] - E[z]^2}$, and, without loss of generality, we will specifically take $P_c = P(x, z = \mu - 2\sigma)$, so that the amorphization threshold is assigned as two standard deviations $\sigma$ away from the mean $\mu$. $P_c$ (regardless of its definition) is then linearized in $\epsilon$.
as \( P_{\alpha 0} \) at \( O(1) \) and \( P_{\alpha 1} \) at \( O(\epsilon) \), and the level curve equation (Equation (5.29)) becomes two equations which are easily solved. At \( O(1) \),

\[
g_0 = P_0^{-1}(P_{\alpha 0}).
\]  

(5.32)

We note that \( P_0 \) is non-invertible due to its symmetry about \( z = \mu \) (see Equation (5.25)); we therefore compute \( P_0^{-1} \) on the domain \( z < \mu \) in order to restore uniqueness. Then, at \( O(\epsilon) \),

\[
\tilde{g}_1 \tilde{h}_1 = \left( P_{c1} - P_1(g_0) \right) - \left. \frac{\partial P_0}{\partial z} \right|_{z=g_0} + i \left. \frac{\partial P_0}{\partial z} \right|_{z=g_0}.
\]  

(5.33)

However, the above expression is not immediately useful, nor does it lend itself to intuition. We will instead consider the equivalent form

\[
\tilde{g}_1 \tilde{h}_1 = r_0 \exp(-ikx_0)
\]  

(5.34)

in order to establish analogy with our previous results ([48, 50], Chapter 3, Chapter 4) where \( x_0 \) is a horizontal shift and \( h_0 \) is a vertical shift. From complex arithmetic, we determine

\[
x_0 = -\frac{1}{k} \arctan \left( \frac{\text{Im}(P_{c1} - P_1(g_0))}{\text{Re}(P_{c1} - P_1(g_0))} \right).
\]  

(5.35)

and

\[
r_0 = \sqrt{\left[ \text{Re} \left( \left. \frac{\partial P_0}{\partial z} \right|_{z=g_0} \right) \right]^2 + \left[ \text{Im} \left( \left. \frac{\partial P_0}{\partial z} \right|_{z=g_0} \right) \right]^2}.
\]  

(5.36)

Now the interpretation of the expression for \( \tilde{g}_1 \tilde{h}_1 \) is clear, and the lateral shift of the amorphous-crystalline interface is \( x_0 \). A factor \( r_0 \) emerges which was not present in the “ellipse-dragging” argument of Chapter 4. Consistent with this argument, we find that given our choice of \( P(x, z) \) based on Equation 5.20, \( r \to 1 \) as \( k \to 0 \). Interestingly, \( r_0 \to 0 \) as \( k \to \infty \), suggesting that for sufficiently large wavenumbers, the amorphous-crystalline interface may appear to “flatten” relative to the free interface (i.e., the relationship between the two is no longer simply characterized by a vertical shift and a horizontal shift, but an amplitude change also). Given the apparent sensitivity to the amorphous-crystalline geometry shown in Chapters 3 and 4, it is possible that this flattening effect may influence wavelength selection (from full-spectrum results rather than the long-wave approximation) and nonlinear behavior. We do
not anticipate that it will affect $\theta_c$-selection, as the Type II bifurcation that governs $\theta_c$ for a given system is fundamentally a long-wave bifurcation, and for long waves we have $r_0 \approx 1$.

5.4. Results

5.4.1. Theoretical

![Graph](image)

Figure 5.1. For 1keV Ar$^+$ → Si, we compare the integral calculations of the present work to the “single-ellipse” argument we described in Chapter 4. We find excellent agreement between the integral calculation and the simplified argument for long waves. Here, we have taken $k=0.01$ as a point of comparison (with $h_0(0) \approx 6.8$ nm).

**Single-ellipse approximation of interface relation.** In our previous work (Chapter 4) we have suggested that a concise expression for the location of the amorphous-crystalline interface might be obtained directly from BCA simulations such as SRIM [78]. This argument was based on superimposing the 95th-percentile level curves of ion implantation originating from each point on the upper interface, defining a criterion for which some point on each resulting level curve would be considered as belonging to the amorphous-crystalline interface, and then collecting all such points. However, this “ellipse-dragging” argument is fundamentally limited to the case of vanishing wavenumber, as it tacitly relies on the level curve being much narrower in its cross-beam axis than the radius of curvature of each length of sinusoidally-perturbed upper interface. In the present work, we have considered
the distribution of stress that results from power deposition, ion implantation, or other phenomena. We have derived more general expressions for the amorphous-crystalline interface than those which were obtained from the “ellipse-dragging” argument, being applicable to linear stability analysis for arbitrary wavenumbers. In Figure (5.1), we have verified that the derivation of \( x_0(\theta) \) and \( h_0(\theta) \) from a linearized surface integral agrees with that from the “ellipse dragging” argument of Chapter 4.

**Linear stability result.** By evaluating the expressions (5.14)-(5.19) using the linearization of (5.20) as described above, and considering the term \( \propto fK_I \) to leading order in \( \frac{\rho_a}{\rho_c} \) (which we will discuss), we obtain the linear dispersion relation with

\[
\text{Re}(\sigma) \approx \left( -\frac{V}{h_0(\theta)} \left( 1 - \frac{\rho_a}{\rho_c} \right) - fK_I \left( \frac{\Omega_0}{h_0^2(\theta)} \right) \right) - 3fK_D \left( \frac{\rho_a}{\rho_c} \right) \left( \cos(2\theta) - \left( \frac{x_0(\theta)}{h_0(\theta)} \right) \sin(2\theta) \right) (kh_0(\theta))^2
\]

\[
-\frac{\gamma}{3h_0(\theta)\eta(\theta)} (kh_0(\theta))^4,
\]

which will determine \( \theta_c \) and \( \lambda(\theta) \) selection. We will use

\[
x_0 = a \sin(\theta) + 2 \left( \frac{\alpha^2 - \beta^2 \sin(\theta) \cos(\theta)}{\sqrt{\alpha^2 \cos^2(\theta) + \beta^2 \sin^2(\theta)}} \right),
\]

\[
h_0 = a \cos(\theta) + 2 \left( \frac{\alpha^2 \cos^2(\theta) + \beta^2 \sin^2(\theta)}{\sqrt{\alpha^2 \cos^2(\theta) + \beta^2 \sin^2(\theta)}} \right)
\]

and we have the coefficient

\[
\Omega_0 = \frac{c}{\sqrt{8\pi(\alpha^2c^2 + \beta^2s^2)}} \left[ -2(\alpha^2c^2 + \beta^2s^2) \left[ \exp\left( \frac{-a^2c^2}{2(\alpha^2c^2 + \beta^2s^2)} \right) - \exp(-2) \right] 
\right.

\[
- \left. ac\sqrt{2\pi(\alpha^2c^2 + \beta^2s^2)} \left[ \text{erf}\left( \frac{ac}{\sqrt{2(\alpha^2c^2 + \beta^2s^2)}} \right) + \text{erf}(\sqrt{2}) \right] \right],
\]

where \( c = \cos(\theta), s = \sin(\theta) \), which occurs as the leading-order term in the expansion of the \( fA_I \) coefficient about \( \frac{\rho_a}{\rho_c} \approx 1 \). We find that while including the phase-change boundary condition leads to a substantially more complicated expression, there is little benefit in using this expression rather than the one obtained in the case of \( \frac{\rho_a}{\rho_c} = 1 \). The full expression for this coefficient, without expansion in \( \frac{\rho_a}{\rho_c} \), is available in the Appendix.

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An added benefit of this simplification is that some intuition may be gained about how IIS strengthens and weakens with different collision cascade parameters. It is also useful to notice that \( \left( \frac{\Omega_0}{h_0^2(\theta)} \right) \) is the result of passing the density \( P(x, z) \) into the integral expression for the linear growth rate and, accordingly, has units \( \frac{q}{\text{ion-nm}} \).

![Graphs showing trends in experimental data](image)

Figure 5.2. A comparison of trends in experimental \( \theta_c \) across 250eV-2keV Ar\(^+\), Kr\(^+\), Xe\(^+\) → Si, and Kr\(^+\), Xe\(^+\) → Ge. **Top left:** \( \theta_c \) plotted against mean depth of ion deposition as computed from SRIM [78]; the linear trend is consistent with the form of \( \frac{\Omega_0}{h_0^2(\theta)} \). **Top right:** \( \theta_c \) plotted against MVDPI = \( \frac{\text{vacancies}}{2aV} \), the vacancies averaged by the volume of the ion distribution. **Bottom left:** the product of mean depth of ion deposition \( a \) and MVDPI. **Bottom right:** the quadratic long-wave coefficient due to [25], showing that the increase in \( \theta_c \) due to deeper ion implantation cannot be attributed to a reduction of erosion-induced instability.

**Experimental trends; toward a scaling argument for IIS.** In the present work, our overarching goal is to improve quantitative agreement between theory and experiment across a range of experimental systems. However, the lack of relevant parameter estimates signifi-
cantly complicates this effort. As a simple alternative, we will attempt to deduce a scaling argument from which known parameter values might be used to infer unknown parameter values—or to reduce the degrees of freedom for the nonlinear fitting procedure that we will perform later in this work. We note that many irradiated thin film systems exhibit simple trends in energy [97] or momentum [101]; this motivates us to consider whether we might obtain some empirical results that would be useful in estimating unknown parameters.

We first consider the form of \( \frac{\Omega_0}{h_0(\theta)} \) qualitatively, which will be useful as a simple first test of our model in the absence of high-precision parameter values \( fA_D, fA_I, \eta \) and \( \gamma \) for systems of interest, while also informing a choice of possible scaling argument. Interestingly, although \( \frac{\Omega_0}{h_0(\theta)} \) is a somewhat complicated function, we notice that for fixed \( \theta \) it is nearly linear in \( a \) (i.e., becoming more stabilizing as \( a \) increases), and \( \alpha, \beta \) do not qualitatively change the shape. Hence the form \( \frac{\Omega_0}{h_0(\theta)} \) itself suggests that the depth of the deposition of \( q \) is a major determining factor in how strongly-stabilizing IIS should be; this is consistent with results discussed in Chapter 3. In Figure (5.2), we plot \( \theta_c \) for 15 different experimental systems against mean depth of ion-implantation \( a \) and we find that not only does the predicted trend exist, but, further, lines of best fit for each of Si or Ge targets exhibit very nearly the same slope. Although preliminary, this appears to strongly indicate that ion implantation itself is a driver of IIS, as was speculated may be the case in [48, 50]. We also note that IIS is the only component of our model that is capable of explaining this simple linear trend; neither the phase-change contribution nor APF appear to scale simply with \( a \) (or \( \frac{a}{h_0} \)).

Next, based on discussion [63, 64, 48, 50] that defect kinetics are likely to be involved in the process of volumization (i.e., density loss), we hypothesize that IIS scales with the production of Frenkel pairs (vacancy-interstitial pairs) within the damaged crystalline lattice. Beardmore et al [87] suggest that this may be the underlying, atomistic mechanism responsible for density loss. In that case, we would expect the mean vacancy density per ion (MVDPI) to be an important quantity such that \( fA_I \propto \text{MVDPI} \) for systems with comparable fluxes, bearing in mind that this quantity acts also as a measurement of mean interstitial...
density per ion (since vacancies and interstitials occur in pairs). We define

\[ \text{MVDPI} = \frac{\text{VPI}}{\frac{42}{3} \pi \alpha \beta^2}, \]  

(5.40)

reflecting the “averaging” of vacancies per ion (VPI) throughout the volume of the ion-implanted region associated with repeated collision events originating from the same patch of free interface. Again, the trend is quite strong (Figure 5.2(b)). We then compare the product \( a \times \text{MVDPI} \) measured for 250eV Ar\(^+ \) → Si against our other systems of interest; this suggests how much stronger IIS might be for a given system in the absence of any other factors (such as changes in \( f_A D \), erosion rate, or \( \frac{\rho_a}{\rho_c} \)). The trend is once more quite strong, suggesting that this may offer a fairly complete explanation of how IIS scales. We note that although the parametric dependence may be more complicated than this, we anticipate that the parameter \( \frac{f_A h_0(\theta)}{V(\theta)} \) should be quite small (see Chapter 3) for most of our systems of interest. We may therefore consider Figure 5.2 as depicting roots \( \theta_c \) of the growth rate in the regime of small dimensionless deformation rates.

The obvious outlier is Xe\(^+ \) → Si, and we have not considered any systems which do not form patterns at all, such as Ne\(^+ \), Ar\(^+ \) → Ge below 2keV, or Ar\(^+ \) → Si from about 2keV to about 10keV. These systems will be addressed in the Discussion, and we hypothesize that their different behavior is due not to IIS but to APF. Shortly, we will show that Xe\(^+ \) → Si is explainable by the present model.

**Dimensional analysis and a speculative scaling argument.** The qualitative trends discussed above suggest that the defect concentration (“MVDPI”) should appear in the expression for \( K_I \), and that scaling with \( a \) emerges naturally from the form of the function \( \Omega_0 \). We note that these trends are consistent with others in the literature. Since the defining feature of IIS is isotropic compressive stress due to volumization (or, volumization due to isotropic compressive stress), MD simulations showing compressive stresses [102, 33] associated with ion-implantation are indicative.

Of course, any comprehensive explanation of the general irradiated thin film system requires not only agreement with \( \theta_c \), but with observed stresses as well. It has been widely
observed that at low energies, ion-induced compressive stresses due to ion bombardment tend to scale with momentum \([81, 101, 103, 82]\). Since the mean in-plane stresses resulting from ion bombardment according to our model should be

\[
<T_{11}^0 > \approx -6 f A_D \eta \cos(2\theta) < \tau_0(z; 0, h_0, ...) > -2 f A_I \eta < \alpha_{01}(z; 0, h_0, ...) >, 
\]

where the mean values of \(\tau_0\) and \(\alpha_{01}\) are reported in the Appendix, we hypothesize that momentum should appear in the scaling of both \(A_D\) and \(A_I\). Hence we take

\[
K_I = \hat{K}_I \text{MVDPI} \times \sqrt{2mE}, 
\]

with \(\hat{K}_I\) a material-specific parameter independent of energy or target, so that all parametric dependence on energy, ion species, etc., is contained within the vacancy density and momentum. In this work, we use units \(m = \text{amu}, E = \text{eV}\).

For \(K_D\), we expect \(\sqrt{2mE}\) to appear so that stress scales according to experimental observations, but we do not anticipate that vacancies should appear. While vacancies are likely the source of tensile stresses in certain contexts \([42]\), there is no obvious means by which they would induce the anisotropy that characterizes APF, and previous work by our group \([96]\) has shown that tracking the displacement field does not appear to produce a stress tensor similar to APF.

We note that the above form of \(K_I\) anticipates \(K_I \propto E^0\), when considering that vacancies \(\propto E\) via Kinchin-Pease, and each of the length scales of the ion-implanted region \((a, \alpha, \beta)\) are approximately \(\propto E^{4} - E^{6}\) \([98]\); based on this, we will suppose that each of \((a, \alpha, \beta)\) are \(\propto E^{1/2}\). Then, agreeing with the well-known Davis scaling of in-plane stress \(\propto E^{-7/6}\) at low energies is incumbent upon \(K_D\) \([97]\).

Davis \([97]\) shows that under some conditions, the same “ion-peening” model that gives rise to the work of \([101]\) and others can lead to a scaling with energy of \(E^{-7/6}\). However, this work was done only for normal incidence, included at least one fit parameter, and does not explicitly reject a role for ion mass. Indeed, \([97]\) shows that an \(E^{1/2}\) scaling of stress occurs in the case of high net-deposition rates. Hypothesizing that the scaling coefficient includes \(\sqrt{m}\) immediately restores the result of \([101]\) (and others); we will therefore include this \(\sqrt{m}\).
in the present work, even though it is not explicitly included by [97]. This would appear to offer a full reconciliation of the work of [97] and [101], and provides a scaling argument amenable to the present work.

We therefore require

\[ T_{0}^{11} \propto K_D \eta \propto E^{-7/6}, \quad (5.43) \]

and from discussion in Chapter 4, we suspect that roughly \( \eta \sim E^{-1/2} \). Since we already expect momentum \( \sim E^{1/2} \) to appear in the scaling, we write

\[ K_D \eta \propto E^{1/2} E^{-x} E^{-y} = E^{-7/6}, \quad (5.44) \]

where \( x \) is the missing exponent attributable to \( K_D \) and \( y \) is the exponent attributable to \( \eta \), for which we have roughly \( y \approx \frac{1}{2} \). This leads to roughly

\[ x \approx 1 \quad (5.45) \]

so that we might anticipate a denominator involving about two length scales (each of which are \( \sim E^{4/6} \)), or about one scaling with vacancies \( \sim E^{1} \). (Other combinations of parameters are possible, but these appear to be the simplest.) However, we do not anticipate that APF should be inversely related to vacancies— if anything, increased vacancies should strengthen APF (based on previous discussion of tensile stresses, [42]). Furthermore, \( K_D \propto \text{vacs}^{-1} \) would appear to “double-count” the effect of vacancy creation and we would expect it to lead to a nonlinear trend in Figure (5.2) above. The simplest interpretation is then that \( x \approx 1 \) above requires inverse-and-joint proportionality with two length scales associated with ion implantation. Although this is a small leap of logic, this leads us to believe that an area should appear in the denominator. It appears natural that the area should be the surface area of the ion-implanted region whose volume already appears in our model; hence we consider

\[ K_D = \hat{K}_D \frac{\sqrt{2mE}}{4\pi \left( 2(\alpha\beta)^{1.6} + (\beta)^{3.2} / 3 \right)^{1/1.6}}; \quad (5.46) \]
i.e., that APF scales inversely to the *surface area* of the 95\textsuperscript{th} percentile of the final ion distribution (where we have used a common approximation). We will reserve comment on the implications of this form until the Discussion.

For the moment, we will note that the above scaling argument, *while admittedly speculative*, should be straightforward to test using angle-dependent wafer curvature experiments like those of [47], from which we obtained estimates of \( f_A I \) and \( f_A D \) in Chapter 3. Angle dependence is required because in-plane stresses attributable to IIS and APF are both compressive at normal incidence, hence inseparable.

We also note that, while the above forms are appealing and attempt to connect isotropic and anisotropic stresses directly to atomistic phenomena, they are fundamentally speculative—even if empirically-motivated. As an alternative, we could have simply taken, as approximations,

\[
K_D \propto \sqrt{m}, \quad K_I \propto \sqrt{\frac{m}{E}}. \]

We note that these scalings will not fully capture the dependencies of the forms above, as the scaling of lengths \((a, \alpha, \beta)\) with \(E^{-4} - E^{-6}\) [98] is only approximate.

5.4.2. Comparison of experiment and theory

5.4.2.1. Several pattern-forming systems: \( \theta_c \) for 250eV-2keV \( Ar^+ \), \( Kr^+ Xe^+ \to Si \) and \( Kr^+, Xe^+ \to Ge \)

Here, we are primarily interested in \( \theta_c \), as the ability for a model to correctly identify \( \theta_c \) across a variety of experimental systems is considered a minimal condition for a successful theory. We will consider several systems known to form patterns for some value of beam angle \( \theta \), and we will defer discussion of systems which do not form patterns to the end, as we attribute these cases to a different aspect of the model than that which was the focus of this work.

For each of these systems, we will consider the least-squares fitting of 15 experimental systems, 8 for Si and 7 for Ge, using two fit parameters for each target. We will use two fit parameters \( \hat{K}_I \) and \( \hat{K}_D \) per target, each under the same hypotheses about the spatial
distribution and scaling of IIS and APF, which are described above. The severely restricted degrees of freedom and the great dissimilarities between the systems under comparison will be informative, as a good fit to the data would suggest the correctness (and robustness) of the underlying physical model. Our data come from [58, 33, 33, 43, 83, 3, 47, 33, 57, 84, 55] and are representative of all data for 250eV-2keV Ar$^+$, Kr$^+$, Xe$^+$ → Si and Kr$^+$, Xe$^+$ → Ge of which we are aware.

The fitting is performed using the SciPy global optimization package opt.brute and “polished” using opt.fmin. As shown in Figure 5.3, we obtain good fits to all data. Below, in “On model reduction”, we will show that this the ability to fit the data is not generic, and depends rather sensitively on spatially-inhomogeneous IIS.

5.4.2.2. Implications for $\lambda(\theta)$ for 250eV, 500eV Ar$^+$ → Si

By fitting $\theta_c$ above, we have obtained parameters $\hat{K}_D$ and $\hat{K}_I$ for each of Si and Ge. These parameters are used to convert a scaled quantity into $A_D$ and $A_I$ for a given experimental
system (ion, target, and energy), and lead to quantitative agreement with $\theta_c$ for several ion-target systems from 250eV-2keV. A natural extension is to take the values of $A_D$ from these estimates, deduce a theoretical $\lambda(\theta)$, and compare with systems for which experimental $\lambda(\theta)$ is known.

For two low-energy systems, 250eV Ar$^+$ → Si [43] and 500eV Ar$^+$ → Si [33], we compare $\lambda(\theta)$ generated by our model to the experimental data for a few values of $\eta$ in the vicinity of $\eta$ expected due to the scaling argument of [93, 14, 18]. This is because there is some uncertainty in $\eta$, up to about an order of magnitude [43, 28]. For simplicity, we employ the typical scaling argument due to Vauth and Mayr [93] while noting that alternative scaling arguments appear to be implied by other work [46], and that at least some experimental work suggests the insufficiency of [93], at least for very high fluxes [44, 47]. The scaling argument of [93] leads to an estimate of $\eta \approx .6 \text{ Gpa} \cdot \text{s}$ for 250eV Ar$^+$ → Si at $35 \frac{\text{ions}}{\text{nm}^2 \cdot \text{s}}$, and about $\eta \approx 15 \text{ Gpa} \cdot \text{s}$ for 500eV Ar$^+$ → Si at $\sim 2.5 \frac{\text{ions}}{\text{nm}^2 \cdot \text{s}}$. We consider a few values near these, within about an order of magnitude. Interestingly, the best fit in Figure 5.4(a) uses a value of $\eta(0)$ nearly the same as that considered in Chapter 4.

Based on these values, we find surprisingly good agreement between experimental and
theoretical wavelengths, with the shape of the angle-dependence and the selected length-scales both well-reproduced by the model, with $\theta_c$ within a few degrees of experimental observations (although we remind the reader that this last point is, of course, because these parameter values originated from fitting to $\theta_c$).

That such a good fit to $\lambda(\theta)$ is achieved is remarkable. Since parameter values were fit to $\theta_c$, fundamentally by a root-finding procedure on the underlying growth rate, parameter fits to $\theta_c$ will not necessarily fit $\lambda(\theta)$, which is fundamentally related to the magnitude of the underlying growth rate. Hence good agreement with experimental $\lambda(\theta)$ using parameter fits to $\theta_c$ leads us to believe that we have successfully captured at least some of the underlying physics.

5.4.2.3. Implications for $\theta_c$ for 250eV-100keV Ar$^+$ → Si: a possible bifurcation

It is observed that for some energies, Ar$^+$ → Si produces either no ripples or extremely small growth rates [22], and any successful model should be able to explain—or at least accommodate—this phenomenon. Using the parameter values obtained from fitting $\theta_c$, we consider higher-energy irradiation of Si by Ar$^+$. We find that in the vicinity of our fitted values, and subject to the scaling argument described above, it is possible that stabilization due to boundary amorphization becomes competitive with, or even dominant over, the stress-based mechanisms, leading to a rapid increase in $\theta_c$. While this depends on the value of $\frac{\rho_a}{\rho_c}$, the values considered in Figure 5.5 are within physical uncertainty. Given that $\frac{\rho_a}{\rho_c}$ is known to vary with energy [87], it is plausible that some combination of stress-based mechanisms and the phase-transition boundary condition can explain the sudden, bifurcation-like suppression of pattern formation that begins around 1.2keV [22]. We also note that lower values of $\frac{\rho_a}{\rho_c}$ are capable of producing $\theta_c \approx 30^\circ$, which is evidently a lower bound for Ar$^+$ irradiation of Si [56]. For higher energies, electronic stopping also gains importance, and it is possible that stress effects become less important to pattern formation well-within the electronic stopping regime, although even within the present framework, a return to pattern formation could be explained by energy-dependent changes in $\frac{\rho_a}{\rho_c}$. As noted in [87], more study of this parameter
Figure 5.5. Here, we consider $\theta_c$ vs beam energy for several different values of $\rho_a/\rho_c$ where we consider $\hat{K}_D$ taken as the fitted value. For these parameter values, the region around 1.2keV beam energy coincides with a very sharp increase in $\theta_c$ as experimentally observed. These values of $\hat{K}_I$ and $\hat{K}_D$ coincide with the region in which the effect of the boundary amorphization term can dominate stress effects. Also note that these values of $\rho_a/\rho_c$ are well-within experimentally-determined values, and that the lowest value of $\theta_c$ is around $30^\circ$. For each of the values of $\rho_a/\rho_c$ considered, it is possible to obtain similar “jumps” in $\theta_c$ for a corrected value of $\hat{K}_D$.

is warranted.

5.4.2.4. Comparison with previous estimates of $A_D$: 250eV, 1keV $Ar^+ \rightarrow Si$

For 250eV $Ar^+ \rightarrow Si$, [43, 44] have estimated $A_D \approx 1.145 \times 10^{-2} \text{nm}^2/\text{ion}$. For the same system, our estimate leads to $A_D \approx 1.3 \times 10^{-4} \text{nm}^2/\text{ion}$, which is quite different. Also, for 1keV $Ar^+ \rightarrow Si$, [28] has estimated parameter group $fA_D \approx 3 \times 10^{-4} \text{s}$, as compared with our estimate of $fA_D \approx 6.7 \times 10^{-5} \text{s}$. This is smaller than the estimate of [28] by a factor of about 5. Due to the uncertainties involved, we consider this a fairly good level of agreement.

However, we note that neither [28] nor [43, 44] considered the isotropic stresses generated by IIS, which could contribute to the slight disagreement between our respective parameter estimates, especially since the estimates of [43, 44] were produced from normal-incidence
wafer curvature experiments, where IIS and APF would be indistinguishable in their influence on in-plane compressive stress. In particular, we have estimated $A_I \approx 1.5 \times 10^{-2} \text{nm}^2 / \text{ion}$ for 250eV Ar$^+ \rightarrow \text{Si}$ at normal incidence; while the conversion to a stress is not straightforward (because it relies on $\eta$ and assuming the physical locus of IIS), it produces the right order of magnitude, while $\eta, < \alpha_{01}(z;0,h_0) > \sim O(1)$. Hence we speculate that some stress measurements attributed to $A_D$ may really belong to $A_I$; this would support a role for angle-dependent stress measurements such as those of [47] in the future, and draws attention to the possibility that the neglect of a second source of ion-induced compressive stress at normal incidence can lead to overestimates of the ion-hammering effect for some systems.

5.4.2.5. Implications for $\theta_c$ for 250eV-2keV Ne$^+$,Ar$^+ \rightarrow \text{Ge}$

Figure 5.6. Here, we consider Ne$^+$, Ar$^+ \rightarrow \text{Ge}$ for 250eV-2keV; these systems are not observed to form patterns, at least up to about 75$^\circ$ [57]. Our model incorrectly predicts that these systems are pattern-forming. We discuss this inconsistency below.

In order to assess the validity of the present model and our hypothesized scaling of APF and IIS, we consider two systems which were not involved in the initial fitting, but which are known to strongly suppress pattern formation [57] up to at least 75$^\circ$. Using the fitted values of $\hat{K}_D$ and $\hat{K}_I$ and the same scaling arguments as before (Equations (5.42) and (5.46)), we sample 250eV, 750eV, 1250eV and 2000eV Ne$^+$ and Ar$^+ \rightarrow \text{Ge}$ and perform a cubic splines fit. Despite the unusual shape, the spline fit is faithful to the underlying
data. Interestingly, we do not obtain the expected strong stabilization up to $\sim 75^\circ$. Further investigation shows that we would require about two more orders of magnitude in $A_I$ or about two fewer orders of magnitude in $A_D$ in order to produce the expected $\theta_c \geq 75^\circ$.

Since the other systems under our consideration appear to be fairly well-described by the present parameter values and scaling arguments, we suspect that something more subtle is happening for these systems. This indicates the incompleteness of the present model; however, we have considered spatially-resolved IIS without spatially-resolved APF, whose distribution (and underlying mechanism) is currently unknown. It is possible that a more complete understanding of APF will lead to predictions for Ne$^+$,Ar$^+ \rightarrow$ Ge behaving as expected, and this is a clear topic for future work. We also note that even taking $\rho_a / \rho_c \rightarrow 0$ is not capable of reproducing $\theta_c \rightarrow 90^\circ$, suggesting that these observations are not explained by the possibility of $\rho_a / \rho_c$ substantially different from the values that we have used for the other systems under consideration here.

A natural hypothesis is that because of the smaller mass ratio between Ne$^+$ and Ge and Ar$^+$ and Ge than for other systems, the directionality of displacements due to momentum transfer from ion to target could be quite weak. This is consistent with discussion by [57]. We also observe that these systems exhibit much more backscattering (see [79, 62] for discussion of backscattering) than our other systems of interest. Backscattering could possibly lead to ions carrying more energy away, which might weaken pattern formation. We regard these systems as prime candidates for angle-dependent wafer curvature experiments, where we anticipate weak APF (observable as a stifled, or absent, compressive-to-tensile transition of in-plane stresses for increasing incidence angle).

5.4.3. On model reduction: spatial inhomogeneity is important

The expression for $\Omega_0$ is rather complicated, and it is natural to ask whether we could possibly use a simpler model to achieve the same (or similar) results. Here, we briefly consider a model that assumes uniform intensity throughout the bulk (i.e., no spatial resolution) but preserves the scaling argument. We find that there is a substantial increase in $l^2$ error in
Figure 5.7. Here, we have attempted to fit the model with the same scaling argument but without spatially-resolved IIS— that is, using the model of Chapter 4. We fit the model (Equation (5.37)) to $\theta_c$ across 250eV-2keV $\text{Ar}^+$, $\text{Kr}^+$, $\text{Xe}^+ \rightarrow \text{Si}$, and $\text{Kr}^+$, $\text{Xe}^+ \rightarrow \text{Ge}$ using two free parameters, $\hat{K}_I$ and $\hat{K}_D$, per target. For Si, we obtain $l^2 \approx 10.09$, $\hat{K}_D = 5.474 \times 10^{-6}$, $\hat{K}_I = 3.0769 \times 10^{-6}$. For Ge, we obtain $l^2 \approx 12.83$, $\hat{K}_D = 7.659 \times 10^{-6}$, $\hat{K}_I = 4.351 \times 10^{-6}$. Dots denote experimental data, and dashed lines are model predictions.

5.5. Discussion

5.5.1. Contributions

Explanation of $\theta_c$ for pattern-forming systems by a single model. In the present work, we have constructed a fairly simple, stress-based model consisting of anisotropic plastic
flow (APF) and ion-induced isotropic swelling (IIS). Unlike most previous work, we have considered the spatial distribution of IIS throughout the irradiated film—and unlike all previous work, we have considered its variation along two axes rather than one, which turns out to be essential in obtaining good agreement between theory and experiment. We have also allowed ourselves a simple, physically-plausible scaling argument for the IIS model: that IIS is correlated to the defect density produced in the ion-implanted region and the momentum of the incoming ion. We find that this enables a great deal of improvement in the fitting, and, because it is a physically-plausible mechanism which agrees with other experimental and computational work, we are inclined to accept that this is the correct physical mechanism to underlie our model of IIS which had previously been only phenomenological in nature.

With a somewhat more speculative scaling argument for APF, we have found that our model is capable of fitting the experimentally-observed values of $\theta_c$ to 8 different data points for irradiated Si, spanning three different ion species and an energy range of about 2keV, using 2 parameters. The same model is capable of fitting $\theta_c$ for 7 different data points for irradiated Ge, spanning two different ion species and a similar energy range. These fits occur with extremely low $l^2$ error, within the range of experimental uncertainty for $\theta_c$ in all cases ($\pm 2.5^\circ$). Importantly, this fit cannot be attributed to the fact that erosion also scales like sputter yield—attempting to replace APF with erosion in the model leads to an immediate deterioration of the fitting. Additionally, erosion is destabilizing at normal incidence and stabilizing at grazing incidence, while the opposite is true for APF, reducing the likelihood that they are somehow interchangeable.

### The importance of spatially-resolved models

We note the comparison between the cases of IIS spatially-resolved and IIS spatially-constant: while for Ge targets, the spatial resolution contributes only a small decrease in $l^2$ error, the decrease for Si is about 50%, a drastic reduction suggesting that not only do we need to understand how a mechanism scales, but we also need to understand where it operates. This is made explicit by the form of the IIS component of the dispersion relation (the term $\propto \Omega_0$ in Equation (5.37)), which
becomes more strongly negative with increasing mean depth $a$. Without an understanding of a physical locus for IIS, this observation could not be made, and even the hypothesis that IIS scales like $\text{MVDPI} \times \text{momentum}$, while useful for explaining $\theta_c$ in Ge targets, is insufficient to explain $\theta_c$ in Si targets.

While, in the present work, we have focused on IIS and its resolution in space, we have only briefly touched on APF— and only speculatively. However, the idea that the physical origin of a mechanism, and therefore its location in space, may strongly affect its contribution to linear stability is intriguing, and suggests an avenue for attempts at mechanism identification by comparison of theory and experiment: what would need to be true about APF in order to reconcile theory and experiment, and can these necessities be plausibly connected to some physical phenomenon? The development of a high-generality framework as has been explored in the present work and previous work by our group poses the interesting possibility of hypothesis-testing *en masse* different mechanisms that could explain APF and their implied spatial distributions. The ability of a single proposed mechanism to parsimoniously explain all data would lead to low-energy APF becoming a true, physical model rather than a phenomenological model, which would constitute a significant step towards first-principles parameter estimation.

**The importance and physical origins of IIS.** In our previous work (Chapters 2-4), we had developed a model of IIS in a fundamentally mechanism-agnostic fashion, having in mind a mathematically generic process that leads to localized decreases in density that become more severe with time. In the present work, the hypothesis that IIS is spatially-localized to the ion-implanted region and scales with momentum and defect-production per volume within that region appears to lead to superior fits for 15 different pattern-forming experimental systems that we have considered. This parsimony appears to suggest that the phenomenological model of IIS is physically-attributable to defect motility within the collision cascade. Such an explanation compares favorably with observations made throughout the literature: (1) that compressive stresses scale with ion implantation at low energies [102,
(2) that nuclear energy deposition is associated with swelling in irradiated Ge in the MeV range [104]; and (3) that there exists a complex interplay between stress generation and ion implantation [42, 46].

**Unification with previous work.** In this Chapter, we have connected observations on the scaling of stress due to [101], [97], and [82] with theoretical predictions made by a model of spatially-inhomogeneous ion-induced swelling. These observations have informed a scaling hypothesis which leads to surprisingly good agreement with experimental results across several areas of interest:

1. \( \theta_c \) for 15 low-energy systems: 250eV-2keV \( \text{Ar}^+ \), \( \text{Kr}^+ \), \( \text{Xe}^+ \) → Si, and \( \text{Kr}^+ \), \( \text{Xe}^+ \) → Ge;

2. Angle-dependent wavelength \( \lambda(\theta) \) for 250eV, 500eV \( \text{Ar}^+ \) → Si. Predictions are available for other systems based on the present model, although values of \( \gamma \) and \( \eta \) are more speculative;

3. Reproduction of the sudden increase in \( \theta_c \) (or, the strong suppression of ripples) in the vicinity of 1.2keV for \( \text{Ar}^+ \) → Si, which emerges generically for a stress that scales according to the Davis scaling [97] and the phase-change boundary condition proposed in Chapter 4;

4. Order-of-magnitude consistency between the parameter values implied by fitting the present model using \( \hat{K}_I \) and \( \hat{K}_D \) to experimental data and other parameter values from throughout the literature [43, 14, 28];

5. Localization of the origin of the compressive stress, hence a physical explanation for IIS, to the ion-implanted region, as suggested in simulations by [102, 33].

5.5.2. Open questions and future work

**Non-pattern-forming systems: \( \theta_c \) for 250eV-2keV \text{Ne}^+, \text{Ar}^+ \rightarrow \text{Ge.}** Despite the apparent, strong agreement with several previous parameter estimates and trends among the
data, the model described in the present work fails to predict a lack of pattern formation for 250eV-2keV Ne\(^{+}\),Ar\(^{+}\) \(\rightarrow\) Ge as described in [57], instead predicting \(\theta_c \sim 40^\circ\) when we use the same values of \(\tilde{K}_D\) and \(\tilde{K}_I\) for Ge as obtained here. However, there are indications that this is not a fundamental problem for the present approach. While IIS appears to be rooted in momentum and defect motility in the ion-implanted region, the physical origin for APF is still unknown. Since momentum appears to be a key component of both APF and IIS in the systems we have considered here, we anticipate that the same will hold for 250eV-2keV Ne\(^{+}\),Ar\(^{+}\) \(\rightarrow\) Ge. However, we expect momentum transfer to be weaker for Ne\(^{+}\), Ar\(^{+}\) \(\rightarrow\) Ge than for other projectile species due to a much smaller mass ratio. We also observe that for Ne and Ar on Ge, recoils and ion-implantation are significantly closer to the surface than for the other systems that we have considered; this may support the hypothesis that it is not merely momentum in absolute, but momentum transfer that governs APF, and, in particular, the directionality of momentum transfer compared to the ion-beam direction. This is consistent with the “directional momentum transfer” mechanism discussed elsewhere [82] which contributes to the “ion-hammering” effect. We therefore postulate that the absence of ripples in Ge irradiated by Ne\(^{+}\) and Ar\(^{+}\) is due to an absence of APF rather than an excess of IIS.

**Physical mechanism for APF.** As discussed above, further reconciliation of theory and experiment is to be expected when APF is better-understood as a mechanism. We hypothesize that it is, indeed, related to directional momentum transfer, as this would account well for the lack of pattern formation in Ne\(^{+}\) and Ar\(^{+}\) on Ge at low energies. More work is clearly needed. However, it is quite interesting that \(K_D \propto \frac{1}{\text{surface area}}\), which suggests that APF is somehow related to ion-implantation. Such behavior evokes comparison with a rich literature of eigenstrain problems originating from the work of Eshelby and others [105]. Hence the physical origin of low-energy APF may actually be in the short-lived, ellipsoidal distribution of inclusions produced by ongoing ion-implantation rather than the more classical single ellipsoidal inclusion. This would suggest that APF is really a nonlocal phenomenon.
and that it originates throughout the entire bulk at once, rather than being localized to the ion-implanted region. Such a hypothesis would appear to be consistent with the present results leading to rather strong agreement with experimental work despite having no spatial resolution of APF.
6.1. Introduction

In our previous work, we have focused primarily on the linear regime, where fluences are low. This is consistent with the strategy outlined elsewhere [6, 51]; because of the extreme complexity of ion-irradiated thin films at the nanoscale, it is possible for two fundamentally different mechanisms to exhibit similar behavior under some conditions, only to exhibit radically different behavior under others. Indeed, this has already happened, as, for a brief time, surface diffusion, athermal effective surface diffusion, and viscous surface energy relaxation coexisted as proposed regularization mechanisms; for more discussion, and to see an extensive review and history of this topic, we refer the reader to the following excellent reviews [10, 4, 5, 6, 51].

In light of this, a cautious approach therefore begins with the linear regime, where there exist at least a few points of comparison between any proposed model and experimental data: (1) $\theta_c$, (2) $\lambda(\theta)$, (3) in-plane stresses, and (4) the “jump” in surface stability for Ar$^+ \rightarrow$ Si near 1.2keV [22]. Once a model is capable of explaining these observations, it is natural to study the implications of that model in the nonlinear regime, where fluences are higher, and the linear, short-time description of thin film growth has failed.

In the previous few Chapters, we have developed a model that appears to be capable of producing good agreement between theory and the quantities just outlined— at least, if we are willing to incorporate a speculative, though empirically-informed, scaling argument for stress. We will therefore seek to develop the nonlinear analogue of that model, which will serve as a basis for studying several problems that exist in the nonlinear regime.
The main contribution of this Chapter is in the derivation of the nonlinear equations as a proof of concept, which lays the foundation for future work. We will also discuss possible applications of this approach. This body of work is incomplete at the time of writing.

6.1.1. Objectives of the present and near-future work

Here, we will derive weakly nonlinear equations describing surface evolution under the “anisotropic plastic flow” model. This opens up several possibilities:

1. Comparison of the resulting PDE with other PDE descriptions of surface evolution due to effective body force models [31, 33, 49], or crater functions [17], modifications thereof, and other prompt-regime mechanisms [20, 23]. The discovery of nonlinearities at leading order that occur in other models as part of a perturbative expansion, but which are important for experimentally-observed nonlinear evolution, would suggest the importance of stress-based mechanisms. Additionally, the discovery of novel nonlinearities not occurring in other models would be interesting, especially if they occur nearer to leading-order than in other models.

2. Comparison of 2D nonlinear PDEs with simulated GISAXS via Sinha’s equation [106], seeking better agreement with experimental observations than that found in [106], especially about the y-direction.

3. Comparison of the direction of ripple propagation with experiments and the diverse predictions of other models, both of the prompt-regime, crater function type or of the stress-based type [33, 49].

4. Determining the presence of absence of predicted coarsening behavior, or “kinetic roughening” or “secondary features”, which were not seen in [49] and other stress-related work. This is a natural place to leverage PETSc for high-precision, long-exposure simulations, as was done in [96].

This will also allow the inclusion of new knowledge about the complex interplay between interfacial geometry, boundary conditions, spatial variation of stress and stress evolution in
the linear regime, as obtained in the previous few Chapters; it would be highly unexpected for these effects to have no implications in the nonlinear regime.

We might seek the development of a stress-based, “bulk-active” analogue of the crater function framework, which may serve as an additional testing ground for stress-based theories and their interactions with erosive-redistributive theories, possibly supplying correction terms; this also serves to address observations that 250eV Ar$^+$ on Si is apparently dominated by redistribution rather than erosion, but even redistributive models can easily lead to incorrect predictions of wavelength and $\theta_c$ [24, 6]; this suggests a strong role for a comprehensive theory of stress evolution in low-energy irradiated thin films which is currently lacking in the literature.

We will also gain a means of performing a multi-scale analysis of the influence of the amorphous film on highly nonplanar geometries through a “projection” argument valid in the case of low-curvature surfaces, analogous to that of [107].

6.2. Model and theory

Below, we briefly summarize the model before beginning the weakly-nonlinear expansion.

6.2.1. Governing equations of the amorphous layer

Here, we present the derivation of a locally long-wave model for a thin film produced by the ion-beam amorphization of a surface whose constitutive law includes an anisotropic plastic flow response to the angle of beam inclination relative to the surface. We begin with momentum and mass conservation in the bulk, expressed as

$$\rho \left( \frac{\partial \vec{v}}{\partial t} + \vec{v} \cdot \nabla \vec{v} \right) = \nabla \cdot \mathbf{T}$$

$$\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \vec{v}) = 0,$$

respectively. In the present model, $\rho$ is not necessarily a constant due to the inclusion of swelling effects, which recent work in the linear regime [48, 50] has shown to be important in (1) thin-film stresses, (2) critical angle selection and (3) wavelength selection. As a
description of the density $\rho$, we take

$$\rho = \frac{\rho_a}{1 + \Delta},$$

(6.3)

where $\rho_a$ is the “basic” density of newly-amorphized a-Si as it enters the thin film. We note that, as discussed in [48, 50], it may be necessary to eventually include a term $\propto (p-p_0)$, although we have not done so here; this would abandon the “quasi-incompressibility” condition in favor of a more traditional treatment of compressibility, but would introduce additional complications beyond the scope of this preliminary work.

A parcel of matter sustains damage as it dwells within the amorphous layer, which is modeled by a forced advection equation

$$\frac{\partial \Delta}{\partial t} + \vec{v} \cdot \nabla \Delta = \alpha.$$  

(6.4)

In the above, $\alpha$ is a function, not necessarily constant in space, which describes the rate at which damage accumulates. In previous work (Chapter 5), connecting such a function to the distribution of ion deposition has led to significant improvements in modeling wavelengths and critical angles. Since such considerations were apparently necessary in the linear regime, we will include them here.

At the lower interface $z = g$, we apply the boundary conditions

$$\vec{v} \cdot \hat{n}_g = \left(1 - \frac{\rho_c}{\rho_a}\right) v_{I,g}$$

$$\vec{v} \cdot \hat{t}_{g,x} = 0$$

$$\vec{v} \cdot \hat{t}_{g,y} = 0$$

$$\Delta = 0,$$ 

(6.5)

where the first is a statement of mass conservation across the amorphous-crystalline boundary generalized to the case of a sharp change in density across that boundary. In the case of no density drop, it simply reduces to the typical no-penetration boundary condition. These boundary conditions, and others, are widely used within the solidification literature [85], but have not seen much application in the context of ion-irradiated nanopatterning outside...
of [20, 22, 23], where they are handled heuristically, and in the absence of bulk flow; see
the Discussion of Chapter 4. The second two boundary conditions are statements of no-slip
boundary conditions. We note that, in principle, we could have considered

\[ \vec{v} \cdot \hat{t}_{g,x} = \beta_x \hat{n}_g \cdot \nabla (\vec{v} \cdot \hat{t}_{g,x}) \]

\[ \vec{v} \cdot \hat{t}_{g,y} = \beta_y \hat{n}_g \cdot \nabla (\vec{v} \cdot \hat{t}_{g,y}) ; \] (6.6)

however, the ratio \( \frac{\beta_x}{\beta_y} \) is expected to be small, and the effect of the slip boundary should
be quite small based on our earlier work (Chapter 4). The condition

\[ \Delta = 0 \] (6.7)

asserts that a parcel of matter entering the amorphous layer from the amorphous-crystalline
interface has sustained just enough damage to amorphize, but not enough to have begun to
“swell”, i.e., to lose density greater than that which is lost during the amorphization process
itself.

At the upper interface \( z = h \), we apply a modified kinematic condition and stress balance,
respectively, as

\[ v_I = \vec{v} \cdot \hat{n} - V_h \left( \frac{\rho_c}{\rho_a} \right) \] (6.8)

\[ T \cdot \hat{n} = -\gamma \kappa \hat{n} ; \] (6.9)

the modified kinematic condition was originally derived in [48] in order to take into account
the loss of material at the upper interface due to erosion. Above, we use for our stress tensor

\[ T = -p I + 2\eta \left( \dot{\mathbf{E}} - f A \tau D(\theta) \right) , \] (6.10)

with \( \dot{\mathbf{E}} \) being the typical strain rate tensor. \( \tau \) is taken to be a function that may impart
spatial variation in the intensity of APF throughout the film: if \( \tau = \text{constant} \), there is no
spatial variation, while \( \tau \) could also be a linear function of the downbeam direction (as in
Chapter 3), or a spatially-resolved function of the collision cascade parameters (as in [7, 8,
9, 25] or Chapter 5). For the beam-injected stress, we have

\[
D(\theta) = R(-\theta)D(0)R(\theta) = \begin{bmatrix}
\frac{3}{2} \cos(2\theta) - \frac{1}{2} & 0 & \frac{3}{2} \sin(2\theta) \\
0 & 1 & 0 \\
\frac{3}{2} \sin(2\theta) & 0 & -\frac{3}{2} \cos(2\theta) - \frac{1}{2}
\end{bmatrix},
\]

rotation tensor

\[
R(\theta) = \begin{bmatrix}
\cos(\theta) & 0 & -\sin(\theta) \\
0 & 1 & 0 \\
\sin(\theta) & 0 & \cos(\theta)
\end{bmatrix},
\]

and

\[
D(0) = \begin{bmatrix}
1 & 0 & 0 \\
0 & 1 & 0 \\
0 & 0 & -2
\end{bmatrix}.
\]

6.2.2. Lubrication scaling

With the intention of exploiting the disparity in length and height scales in the thin-film geometry, we apply the standard lubrication scalings [108, 109, 110]. We will apply the scalings and nondimensionalization simultaneously as

\[
x = \frac{h_0 \tilde{x}}{\epsilon}; \quad y = \frac{h_0 \tilde{y}}{\epsilon}; \quad z = h_0 \tilde{z}; \quad t = \frac{h_0 \tilde{t}}{\epsilon u_0}; \quad u = u_0 \tilde{u}; \quad v = u_0 \tilde{v}; \quad w = \epsilon u_0 \tilde{w}
\]

where \( h_0 \) is a characteristic length scale, \( u_0 \) is a characteristic velocity, and \( \epsilon := Q = h_0 \sqrt{k_1^2 + k_2^2} \) is a nondimensionalized wave number. For the purposes of later performing an asymptotic expansion (hence exploiting the long-wave geometry) we impose \( \epsilon \ll 1 \). We also nondimensionalize the pressure, surface tension constant, upper and lower interfaces and ion flux as

\[
p = \frac{\eta u_0 \tilde{p}}{\epsilon h_0}; \quad \gamma = \frac{\eta u_0 \tilde{\gamma}}{\epsilon^3}; \quad h = h_0 \tilde{h}; \quad g = h_0 \tilde{g}; \quad f = \frac{u_0 \tilde{f}}{h_0}.
\]
**Bulk equations.** With these scalings and many applications of the chain rule, we obtain the nondimensionalized conservation of momentum (6.1):

\[
Re(\epsilon \ddot{u}_t + \epsilon \ddot{u}_x + \epsilon \ddot{v}_y + \epsilon \ddot{w}_z) = -\ddot{p}_z + 2\epsilon^2 \dddot{u}_{xx} \\
+ \epsilon^2 \ddot{u}_{yy} + \ddot{u}_{zz} + \epsilon^2 \ddot{v}_{xy} + \epsilon^2 \ddot{w}_{xz} + 2\tilde{f} A(\tilde{D}_{11}\tau_x + \epsilon \tilde{D}_{12}\tau_y + \tilde{D}_{13}\tau_z);
\]

\[
Re(\epsilon \ddot{v}_t + \epsilon \ddot{u}_x + \epsilon \ddot{v}_y + \epsilon \ddot{w}_z) = -\ddot{p}_y + \epsilon^2 \dddot{v}_{xx} + 2\epsilon^2 \ddot{v}_{yy} + \ddot{v}_{yy} + \epsilon^2 \ddot{u}_{xz} + \epsilon^2 \ddot{w}_{yz} \\
+ 2\tilde{f} A(\epsilon \tilde{D}_{21}\tau_x + \tilde{D}_{22}\tau_y + \tilde{D}_{23}\tau_z);
\]

\[
Re(\epsilon \ddot{w}_t + \epsilon^2 \ddot{u}_x + \epsilon^2 \ddot{v}_y + \epsilon^2 \ddot{w}_z) = -\ddot{p}_z + \epsilon^3 \dddot{w}_{xx} + 2\epsilon^2 \ddot{w}_{yy} + 2\epsilon \ddot{w}_{zz} + \epsilon \ddot{u}_{xz} + \epsilon \ddot{v}_{yz} \\
+ 2\tilde{f} A(\epsilon \tilde{D}_{31}\tau_x + \epsilon \tilde{D}_{32}\tau_y + \frac{\tilde{D}_{33}}{\epsilon}\tau_z)
\]

(6.16)

where \(Re\) is the dimensionless Reynolds number, \(\frac{\rho_0 h_0}{\eta}\). Nondimensionalization of conservation of mass leads to

\[
\ddot{\rho}_t + \left(\ddot{\rho}_x \dddot{u} + \ddot{\rho}_y \dddot{v} + \ddot{\rho}_z \dddot{w}\right) + \dddot{\rho} \left(\dddot{u}_x + \dddot{v}_y + \dddot{w}_z\right) = 0,
\]

(6.17)

and the forced advection equation describing the accumulation of damage in the amorphous layer leads to

\[
\Delta_t + (\dddot{u}_x + \dddot{v}_y + \dddot{w}_z) = \left(\frac{h_0}{\epsilon u_0}\right) \alpha.
\]

(6.18)

We will formally expand \(\alpha\) \((= \frac{1}{\epsilon})\) to its first term in \(f A_f\), since it is natural that the local swelling rate should be proportional to flux \(f\) and some parameter \(A_f\), which has previously (Chapter 5) been useful in reconciling theory and experiment in the linear regime. That is,

\[
\Delta_t + (\dddot{u}_x + \dddot{v}_y + \dddot{w}_z) = \left(\frac{f A_f h_0}{\epsilon u_0}\right) \alpha_1,
\]

(6.19)

where \(\alpha_1\) is dimensionless. We then consider \(\frac{f A_f h_0}{u_0} \ll 1\), so that \(\left(\frac{f A_f h_0}{\epsilon u_0}\right) \sim O(1)\), and we leave \(\alpha_1\) arbitrary so that it may be assigned later as needed. We also note that this assumption as to the form of \(\alpha\) is somewhat naïve; more realistically, we would expect \(\alpha\) to prohibit “infinite volumization”. As a quasi-static approximation, it is natural to consider the steady-state of \(\Delta\) given \(\alpha\), so that the influence of the ion-implantation is retained, even if the temporal evolution of the bulk IIS field is not.
Now taking the viscous limit $\text{Re} \to 0$ leads to the nondimensionalized momentum equations

\begin{align}
\tilde{p}_x &= 2\epsilon^2 \tilde{u}_{\tilde{z}\tilde{x}} + \epsilon^2 \tilde{u}_{\tilde{g}\tilde{g}} + \tilde{u}_{\tilde{z}\tilde{z}} + \epsilon^2 \tilde{v}_{\tilde{g}\tilde{z}} + \epsilon^2 \tilde{w}_{\tilde{g}\tilde{z}} + 2\tilde{f}A(\tilde{D}_{11}\tau_x + \epsilon \tilde{D}_{12}\tau_y + \tilde{D}_{13}\tau_z) \\
\tilde{p}_y &= \epsilon^2 \tilde{v}_{\tilde{z}\tilde{x}} + 2\epsilon^2 \tilde{v}_{\tilde{g}\tilde{g}} + \tilde{v}_{\tilde{z}\tilde{z}} + \epsilon^2 \tilde{u}_{\tilde{g}\tilde{y}} + \epsilon^2 \tilde{w}_{\tilde{g}\tilde{y}} + 2\tilde{f}A(\epsilon \tilde{D}_{21}\tau_x + \tilde{D}_{22}\tau_y + \tilde{D}_{23}\tau_z) \\
\tilde{p}_z &= \epsilon^3 \tilde{w}_{\tilde{z}\tilde{x}} + \epsilon^3 \tilde{w}_{\tilde{g}\tilde{g}} + 2\epsilon^2 \tilde{w}_{\tilde{z}\tilde{z}} + \epsilon^2 \tilde{v}_{\tilde{g}\tilde{z}} + \epsilon^2 \tilde{v}_{\tilde{g}\tilde{y}} + 2\tilde{f}A(\epsilon \tilde{D}_{31}\tau_x + \epsilon \tilde{D}_{32}\tau_y + \frac{\tilde{D}_{33}}{\epsilon}\tau_z)
\end{align}

We note that, like $\alpha$, we have not yet assumed any particular functional form of $\tau$, and the factors of $\epsilon$ in its coefficients are due to the scaling of the differential operators. We will now formally expand $\tau$ as

$$\tau = \tau_0 + \epsilon \tau_1 + \epsilon^2 \tau_2 + O(\epsilon^3)$$

so that we may proceed with the derivation up to $O(\epsilon^2)$ in full generality. With a given functional form of $\tau$, the corresponding $\tau_0 - \tau_2$ will be determined.

**Mass-conservation at amorphous-crystalline interface.** From the boundary conditions above, we obtain

\begin{align}
\tilde{u} + \epsilon^2 \tilde{w}_g &= 0 \\
\tilde{v} + \epsilon^2 \tilde{w}_g &= 0 \\
-\tilde{u}_g + \tilde{v}_g + \tilde{w} = \left(1 - \frac{\rho_c}{\rho_a}\right)\tilde{g}_g \\
\Delta &= 0
\end{align}

at $\tilde{z} = \tilde{g}$.

**Mass-conservation at free interface (kinematic condition).** The kinematic condition leads to

$$\tilde{h}_i = -\tilde{u}_h \tilde{z} - \tilde{v}_h \tilde{g} + \tilde{w} - \tilde{V}_h \frac{\rho_a}{\rho_c}.$$  

Now, in anticipation of needing the ion-induced, isotropic stresses to compete with the pressure, we scale the diagonal of (6.13) by $\frac{1}{\epsilon}$, thus introducing $D_{ij} = \tilde{D}_{ij} \times \{1 + (\frac{1}{\epsilon} - 1)\delta_{ij}\}$ where $\delta_{ij}$ is the Kronecker delta; this is consistent with discussion in [108], and also turns out...
to be necessary for obtaining the correct linearization of the resulting nonlinear equations
(see the Appendix). We note that, up until we consider the stress balance at the interface,
the physics involved in the model has been equivalent to that of the well-known Orchard
result for viscous leveling of thin films [27]. Where we depart from the scenario considered
by Orchard is in the inclusion of external stresses imposed by the ion beam (6.13), which
show up in the interface stress balance (6.9).

**Stress balance at free interface.** The nondimensionalized stress balance at the upper
interface \( \tilde{z} = \tilde{h} \) is:

\[
\left( -\tilde{e} \tilde{h}_x \left( -\frac{\tilde{p}}{\epsilon} + 2\epsilon \tilde{u}_x - 2\tilde{f} \tilde{A} \tilde{D}_{11} \tau \right) \right) - \tilde{e} \tilde{h}_y \left( \epsilon \tilde{u}_y + \epsilon \tilde{v}_x - 2\tilde{f} \tilde{A} \tilde{D}_{12} \tau \right) + \tilde{u}_x + \epsilon^2 \tilde{w}_x - 2\tilde{f} \tilde{A} \tilde{D}_{13} \tau \times \\
\left( 1 - (\tilde{h}_{xx}^2 + \tilde{h}_{yy}^2) \epsilon^2 \right) \\
= \frac{\tilde{\gamma}}{\epsilon^3} \left( -\epsilon \tilde{h}_y \left( \epsilon^2 \tilde{h}_{yy}(1 + \epsilon^2 \tilde{h}_{yy}) - 2\epsilon^4 \tilde{h}_{xx} \tilde{h}_{yy} + \epsilon^2 \tilde{h}_{xx}(1 + \epsilon^2 \tilde{h}_{yy}) \right) \left( 1 - 2(\tilde{h}_{xx}^2 + \tilde{h}_{yy}^2) \epsilon^2 \right), \right.
\]

(6.26)

\[
\left( -\tilde{e} \tilde{h}_y \left( -\frac{\tilde{p}}{\epsilon} + 2\epsilon \tilde{v}_y - 2\tilde{f} \tilde{A} \tilde{D}_{22} \tau \right) \right) - \tilde{e} \tilde{h}_x \left( \epsilon \tilde{v}_x + \epsilon \tilde{u}_y - 2\tilde{f} \tilde{A} \tilde{D}_{21} \tau \right) + \tilde{v}_y + \epsilon^2 \tilde{w}_y - 2\tilde{f} \tilde{A} \tilde{D}_{23} \tau \times \\
\left( 1 - (\tilde{h}_{xx}^2 + \tilde{h}_{yy}^2) \epsilon^2 \right) \\
= \frac{\tilde{\gamma}}{\epsilon^3} \left( -\epsilon \tilde{h}_x \left( \epsilon^2 \tilde{h}_{xx}(1 + \epsilon^2 \tilde{h}_{xx}) - 2\epsilon^4 \tilde{h}_{xx} \tilde{h}_{yy} + \epsilon^2 \tilde{h}_{xx}(1 + \epsilon^2 \tilde{h}_{yy}) \right) \left( 1 - 2(\tilde{h}_{xx}^2 + \tilde{h}_{yy}^2) \epsilon^2 \right), \right.
\]

(6.27)

\[
\left( -\frac{\tilde{p}}{\epsilon} + 2\epsilon \tilde{w}_z - 2\tilde{f} \tilde{A} \tilde{D}_{33} \tau \right) - \epsilon \tilde{h}_x \left( \epsilon^2 \tilde{w}_x + \tilde{u}_z - 2\tilde{f} \tilde{A} \tilde{D}_{31} \tau \right) - \epsilon \tilde{h}_y \left( \epsilon^2 \tilde{w}_y + \tilde{v}_z - 2\tilde{f} \tilde{A} \tilde{D}_{32} \tau \right) \times \\
\left( 1 - (\tilde{h}_{xx}^2 + \tilde{h}_{yy}^2) \epsilon^2 \right) \\
= \frac{\tilde{\gamma}}{\epsilon^3} \left( \epsilon^2 \tilde{h}_{yy}(1 + \epsilon^2 \tilde{h}_{yy}) - 2\epsilon^4 \tilde{h}_{xx} \tilde{h}_{yy} + \epsilon^2 \tilde{h}_{xx}(1 + \epsilon^2 \tilde{h}_{yy}) \right) \left( 1 - 2(\tilde{h}_{xx}^2 + \tilde{h}_{yy}^2) \epsilon^2 \right). \right.
\]

(6.28)

**Nonlinear evolution equations: full model.** Integrating bulk conservation of mass and
exploiting the Leibniz integral rule and boundary conditions (as in, for example, [108]), we
obtain the system
\[
\dot{h}_i = -\frac{\partial}{\partial \tilde{x}} \int_\tilde{y}^\hat{h} \tilde{u} d\tilde{z} - \frac{\partial}{\partial \tilde{y}} \int_\tilde{y}^\hat{h} \tilde{v} d\tilde{z} + f \tilde{A}_1 \int_\tilde{y}^\hat{h} \left( \frac{\alpha_1}{1 + \Delta} \right) d\tilde{z} - \dot{V}_h \left( \frac{\rho_c}{\rho_a} \right) + \left( 1 - \frac{\rho_c}{\rho_a} \right) \tilde{g}_i, \tag{6.29}
\]
\[
\Delta_i + \left( \tilde{u} \Delta_{\tilde{x}} + \tilde{v} \Delta_{\tilde{y}} + \tilde{w} \Delta_{\tilde{z}} \right) = f \tilde{A}_1 \alpha_1, \tag{6.30}
\]
\[
\tilde{g}(\tilde{x}, \tilde{y}, \tilde{t}) = \{(\tilde{x}, \tilde{y}) \text{ s.t. } \tilde{P}(\tilde{x}, \tilde{y}, \tilde{z}) = \tilde{P}_c \}, \tag{6.31}
\]
where, in dimensional form,
\[
P(x, y, z) = \int_{-\infty}^\infty \int_{-\infty}^\infty \left( \cos(\theta) + h_X \sin(\theta) \right) E_D(x, y, z; X, h(X, Y)) dX dY \tag{6.32}
\]
and
\[
E_D(x, y, z; X, h(X, Y)) = \frac{1}{(2\pi)^{3/2} \alpha \beta^2} \exp \left( -\frac{(\hat{z} - a)^2}{2\alpha^2} - \frac{\hat{x}^2 + \hat{y}^2}{2\beta^2} \right), \tag{6.33}
\]
where
\[
\hat{x} \to (x - X) \cos(\theta) + (z - h(X, Y)) \sin(\theta)
\]
\[
\hat{z} \to (x - X) \sin(\theta) - (z - h(X, Y)) \cos(\theta)
\]
\[
\hat{y} \to y
\]
by change of coordinates. In Equation (6.29) above, the first two terms are typical of lubrication models; the third emerges from the action of IIS in the bulk; the fourth is due to the modified kinematic condition which includes the effect of erosion; and the fifth is due to the phase-change boundary condition at the amorphous-crystalline interface. It is interesting to notice that the evolution of the upper interface depends non-trivially on the “damage field” \(\Delta\) and on the evolution of the amorphous-crystalline interface \(\tilde{g}\). The second equation tracks damage to the amorphous layer; again, it may be advantageous to use the steady state of \(\Delta\) given \(\alpha\) as a quasi-static approximation. Finally, we require a definition of the amorphous-crystalline interface \(\tilde{g}\); in the third equation, we will define it as we have done in Chapter 5, through a level curve of the deposition of some quantity (e.g., ions, power, momentum, etc.,) which can be modeled approximately as a bivariate Gaussian \([7, 8]\). We note that the “single-ellipse approximation” of our earlier work (Chapter 4),
\[
x_0 = a \sin(\theta) + 2 \left( \frac{(\alpha^2 - \beta^2) \sin(\theta) \cos(\theta)}{\sqrt{\alpha^2 \cos^2(\theta) + \beta^2 \sin^2(\theta)}} \right); \tag{6.35}
\]
\[
h_0 = a \cos(\theta) + 2 \left( \frac{\alpha^2 \cos^2(\theta) + \beta^2 \sin^2(\theta)}{\sqrt{\alpha^2 \cos^2(\theta) + \beta^2 \sin^2(\theta)}} \right),
\]
is also likely to be an appropriate simplification for $\tilde{g}$, although it neglects the possibility of local “flattening”.

From these equations, solutions of $\tilde{u}$ and $\tilde{v}$ in terms of $\tilde{h}$ and $\tilde{g}$ lead to a partial differential equation expressed completely in terms of the interfaces and the “damage field”, with all other scalar fields handled implicitly. With $\tilde{g}$ a function of $\tilde{h}$, we obtain a complete characterization of the nonlinear evolution of the surface in terms of interface relation, stress tensor, spatial variation of the stress tensor, and spatial variation of the swelling rate.

**Expansion of slowly-varying scalar fields.** We now expand formally in powers of $\epsilon$,

\[ \tilde{u} = \tilde{u}_0 + \epsilon \tilde{u}_1 + \epsilon^2 \tilde{u}_2 + O(\epsilon^3) \]
\[ \tilde{v} = \tilde{v}_0 + \epsilon \tilde{v}_1 + \epsilon^2 \tilde{v}_2 + O(\epsilon^3) \]
\[ \tilde{w} = \tilde{w}_0 + \epsilon \tilde{w}_1 + \epsilon^2 \tilde{w}_2 + O(\epsilon^3) \]
\[ \tilde{p} = \tilde{p}_0 + \epsilon \tilde{p}_1 + \epsilon^2 \tilde{p}_2 + O(\epsilon^3) \]
\[ \tau = \tau_0 + \epsilon \tau_1 + \epsilon^2 \tau_2 + O(\epsilon^3) \]
\[ \alpha_1 = \alpha_{10} + \epsilon \alpha_{11} + \epsilon^2 \alpha_{12} + O(\epsilon^3) \]
\[ \Delta = \Delta_0 + \epsilon \Delta_1 + \epsilon^2 \Delta_2 + O(\epsilon^3) \]

and we will solve the leading-order equations, as the resulting nonlinear behavior is already nontrivial at $O(1)$.

**Leading-order equations:** At leading order in $\epsilon$, we have the following in the bulk:

\[ \tilde{p}_{0,\tilde{x}} = \tilde{u}_{0,\tilde{x},\tilde{x}} + 2f \tilde{A}_D \left( D_{11} \tau_{0,\tilde{x}} + D_{13} \tau_{0,\tilde{z}} \right) \]  
(6.36)

\[ \tilde{p}_{0,\tilde{y}} = \tilde{v}_{0,\tilde{x}} + 2f \tilde{A}_D \left( D_{22} \tau_{0,\tilde{y}} + D_{23} \tau_{0,\tilde{z}} \right) \]  
(6.37)

\[ \tilde{p}_{0,\tilde{z}} = 2f \tilde{A}_D D_{33} \tau_{0,\tilde{z}} \]  
(6.38)

\[ \tilde{u}_{0,\tilde{x}} + \tilde{v}_{0,\tilde{y}} + \tilde{w}_{0,\tilde{z}} = \frac{f \tilde{A}_I \alpha_{01}}{1 + \Delta_0} \]  
(6.39)

\[ \Delta_{0\tilde{t}} + \left( \tilde{u}_{0,\Delta_{0,\tilde{x}}} + \tilde{v}_{0,\Delta_{0,\tilde{y}}} + \tilde{w}_{0,\Delta_{0,\tilde{z}}} \right) = f \tilde{A}_I \alpha_{01} \]  
(6.40)
At $\tilde{z} = \tilde{g}$, we have
\begin{align*}
\tilde{u}_0 &= \tilde{v}_0 = 0 \quad (6.41) \\
\tilde{w}_0 &= \left(1 - \frac{\rho_c}{\rho_a}\right)\tilde{g}_\xi \quad (6.42) \\
\Delta_0 &= 0 \quad (6.43)
\end{align*}

and at $\tilde{z} = \tilde{h}$, we have
\begin{align*}
-\tilde{h}_x(-\tilde{p}_0 - 2\tilde{f}A_D\tau_0\tilde{D}_{11}) + \tilde{u}_{0,z} - 2\tilde{f}A_D\tau_0\tilde{D}_{13} &= \tilde{\gamma}\tilde{h}_x(\tilde{h}_{xx} + \tilde{h}_{yy}) \quad (6.44) \\
-\tilde{h}_y(-\tilde{p}_0 - 2\tilde{f}A_D\tau_0\tilde{D}_{22}) + \tilde{v}_{0,z} - 2\tilde{f}A_D\tau_0\tilde{D}_{23} &= \tilde{\gamma}\tilde{h}_y(\tilde{h}_{xx} + \tilde{h}_{yy}) \quad (6.45) \\
-\tilde{p}_0 - 2\tilde{f}A_D\tau_0\tilde{D}_{33} &= \tilde{\gamma}(\tilde{h}_{xx} + \tilde{h}_{yy}) \quad (6.46)
\end{align*}

From the above, we may obtain a full solution for $u_0$ and $v_0$, which are sufficient to study the leading-order behavior of the nonlinear system. It is interesting to notice that, to leading order, $\tilde{u}_0$ and $\tilde{v}_0$ may be determined in the bulk without consideration of $\Delta$; the same is not true at higher orders in $\epsilon$, where solution of the flow field is complicated by the need to have already obtained $\tilde{w}_0$ at O(1).

6.2.3. Simplification: constant APF and uniform density

**O(1):** At leading order in $\epsilon$, we have the following in the bulk:
\begin{align*}
\tilde{p}_{0,\tilde{x}} &= \tilde{u}_{0,\tilde{z},\tilde{z}} \quad (6.47) \\
\tilde{p}_{0,\tilde{y}} &= \tilde{v}_{0,\tilde{z},\tilde{z}} \quad (6.48) \\
\tilde{p}_{0,\tilde{z}} &= 0 \quad (6.49) \\
\tilde{u}_{0,\tilde{x}} + \tilde{v}_{0,\tilde{y}} + \tilde{w}_{0,\tilde{z}} &= 0 \quad (6.50)
\end{align*}

At $\tilde{z} = \tilde{g}$, we have
\begin{align*}
\tilde{u}_0 &= \tilde{v}_0 = \tilde{w}_0 = 0, \quad (6.51)
\end{align*}
and at $\tilde{z} = \tilde{h}$, we have

\[ -\tilde{h}_z(-\tilde{p}_0 - 2\tilde{f}A\tilde{D}_{11}) + \tilde{u}_{0,\tilde{z}} - 2\tilde{f}A\tilde{D}_{13} = \tilde{\gamma}\tilde{h}_z(\tilde{h}_{\tilde{x}\tilde{x}} + \tilde{h}_{\tilde{y}\tilde{y}}) \quad (6.52) \]

\[ -\tilde{h}_y(-\tilde{p}_0 - 2\tilde{f}A\tilde{D}_{22}) + \tilde{v}_{0,\tilde{z}} - 2\tilde{f}A\tilde{D}_{23} = \tilde{\gamma}\tilde{h}_y(\tilde{h}_{\tilde{x}\tilde{x}} + \tilde{h}_{\tilde{y}\tilde{y}}) \quad (6.53) \]

\[ -\tilde{p}_0 - 2\tilde{f}A\tilde{D}_{33} = \tilde{\gamma}(\tilde{h}_{\tilde{x}\tilde{x}} + \tilde{h}_{\tilde{y}\tilde{y}}) \quad (6.54) \]

We may solve these equations by observing that (6.38) implies that $\tilde{p}_0 = c(x, y)$. Then (6.46) gives the value of $\tilde{p}_0$ explicitly; also, (6.44) and (6.45) may both be reduced to provide conditions for $\tilde{u}_{0,\tilde{z}}$ and $\tilde{v}_{0,\tilde{z}}$. (6.36) and (6.37) may each be integrated twice in $\tilde{z}$ to obtain terms that are at most quadratic in $\tilde{z}$, with $\tilde{p}_0$ appearing as a coefficient with $(\tilde{x}, \tilde{y})$-dependence.

There are four constants of integration which may be obtained by substituting the quadratic expressions into (6.41),(6.44) and (6.45). Then $\tilde{p}_0, \tilde{u}_0$ and $\tilde{v}_0$ are fully determined. We may integrate (6.39) once in $\tilde{z}$ to obtain $\tilde{w}_0$ as an expression in terms of $\tilde{u}_0, \tilde{v}_0$ and a constant of integration with possible $(\tilde{x}, \tilde{y})$-dependence. This constant may be determined by using (6.41).

**O(\epsilon):** At the next order in $\epsilon$, we have the following in the bulk:

\[ \tilde{p}_{1,\tilde{x}} = \tilde{u}_{1,\tilde{x},\tilde{z}} \quad (6.55) \]

\[ \tilde{p}_{1,\tilde{y}} = \tilde{v}_{1,\tilde{x},\tilde{z}} \quad (6.56) \]

\[ \tilde{p}_{1,\tilde{z}} = 0 \quad (6.57) \]

\[ \tilde{u}_{1,\tilde{x}} + \tilde{v}_{1,\tilde{y}} + \tilde{w}_{1,\tilde{z}} = 0. \quad (6.58) \]

At $\tilde{z} = \tilde{g}$, we have

\[ \tilde{u}_1 = \tilde{v}_1 = \tilde{w}_1 = 0, \quad (6.59) \]

and at $\tilde{z} = \tilde{h}$, we have

\[ -\tilde{h}_z(-\tilde{p}_1) - \tilde{h}_y(-2\tilde{f}A\tilde{D}_{12}) + \tilde{u}_{1,\tilde{z}} = 0 \quad (6.60) \]

\[ -\tilde{h}_y(-\tilde{p}_1) - \tilde{h}_x(-2\tilde{f}A\tilde{D}_{21}) + \tilde{v}_{1,\tilde{z}} = 0 \quad (6.61) \]

\[ \tilde{p}_1 = 0 \quad (6.62) \]
The solution is easily obtained at this order. From (6.13), it is known that \( \tilde{D}_{21} = \tilde{D}_{12} = 0 \). Now (6.58) and (6.62 imply that \( \tilde{p}_1 = 0 \). Using (6.55), (6.57), and (6.58), we know that \( \tilde{u}_1 \) and \( \tilde{v}_1 \) are at most quadratic in \( \tilde{z} \). Then the homogeneous boundary conditions for \( \tilde{u}_1 \) and \( \tilde{v}_1 \) imply that each are 0, and likewise \( \tilde{w}_1 \); i.e., there is no \( O(\epsilon) \) correction term.

**O(\( \epsilon^2 \)):** At \( \epsilon^2 \), we have the following in the bulk:

\[
\tilde{p}_{2,\tilde{z}} = \tilde{u}_{0,\tilde{x},\tilde{z}} + \tilde{u}_{0,\tilde{y},\tilde{g}} + \tilde{u}_{2,\tilde{z},\tilde{z}} \tag{6.63}
\]

\[
\tilde{p}_{2,\tilde{y}} = \tilde{v}_{0,\tilde{x},\tilde{z}} + \tilde{v}_{0,\tilde{y},\tilde{g}} + \tilde{v}_{2,\tilde{z},\tilde{z}} \tag{6.64}
\]

\[
\tilde{p}_{2,\tilde{z}} = \tilde{w}_{0,\tilde{z},\tilde{z}} \tag{6.65}
\]

\[
\tilde{u}_{2,\tilde{z}} + \tilde{v}_{2,\tilde{y}} + \tilde{w}_{2,\tilde{z}} = 0. \tag{6.66}
\]

At \( \tilde{z} = \tilde{g} \), we have

\[
\tilde{u}_2 = \tilde{v}_2 = \tilde{w}_2 = 0, \tag{6.67}
\]

and at \( \tilde{z} = \tilde{h} \), we have

\[
\gamma(-\tilde{h}_z) \left( \tilde{h}_x^2 \tilde{h}_g^2 - 2\tilde{h}_x \tilde{h}_g \tilde{h}_x,\tilde{g} + \tilde{h}_g^2 \tilde{h}_x,\tilde{x} \right) - 2(\tilde{h}_x^2 + \tilde{h}_g^2)(\tilde{h}_x,\tilde{x} + \tilde{h}_g,\tilde{g}) \right) = 0
\]

As before, we may integrate (6.65) to obtain an expression for \( \tilde{p}_2 \) in terms of \( \tilde{w}_{0,\tilde{z}} \) and an unknown function of \((\tilde{x}, \tilde{y})\). This may be substituted into (6.70) to solve for the unknown
function, and so $\tilde{p}_2$ is known. Then we may integrate each of (6.63) and (6.64) twice in $\tilde{z}$ to obtain expressions for $\tilde{u}_2$ and $\tilde{v}_2$ involving a total of four integration constants with possible $(\tilde{x}, \tilde{y})$-dependence. These “constants” may be obtained from substitution into (6.67), (6.68) and (6.69). If desired, we may also obtain $\tilde{w}_2$ from (6.66), although this is unnecessary for our purposes.

**Restriction to a single axis.** From the above, a weakly-nonlinear equation on two axes is obtained. Restricting to the $x$-axis, we obtain in dimensionless quantities

\[
\tilde{h}_0 = -3\tilde{f}A_D(\tilde{g} - \tilde{h}) \left( (\tilde{g}_x - \tilde{h}_x)(\sin(2\theta) - 2\cos(2\theta)\tilde{h}_x) + \cos(2\theta)\tilde{h}_{\tilde{x}\tilde{x}}(\tilde{h} - \tilde{g}) \right)
\]

\[
-\tilde{\gamma}(\tilde{g} - \tilde{h})(\tilde{h}_{\tilde{x}\tilde{x}}(2(\tilde{g}_x - \tilde{h}_x)\tilde{h}_x + (\tilde{g} - \tilde{h})\tilde{h}_{\tilde{x}\tilde{x}}) - (\tilde{g} - \tilde{h})(\tilde{g}_x - 2\tilde{h}_x)\tilde{h}_{\tilde{x}\tilde{x}\tilde{x}})
\]

\[
+\frac{\tilde{\gamma}}{3}(\tilde{g} - \tilde{h})^3\tilde{h}_{\tilde{x}\tilde{x}\tilde{x}},
\]

and restoring dimensionality, if needed, is simple.

**A simple specialization.** We would like to consider the influence of ion, target and energy on the nonlinear evolution equations. In principle, one should then evaluate $P(x, z)$ at each step, determine suitable level curves to define the amorphous-crystalline interface. $P(x, z)$ should also fully account for energy loss due to backscattering, recoil production, and localized phase-change (especially at higher energies). While these efforts are appropriate for a highly-detailed analysis where the “final model” is certain, we consider the present work only a plausibility study, the first extension into the nonlinear regime of the work begun in [32, 48]. We will therefore favor simplifications that (1) minimally allow for some sense of ion, target, and energy dependence and (2) allow at least *some* analytical insight.

For simplicity, we may consider the single-axis case where

\[
g(x, t) = h(x - x_0(\theta), t) - h_0(\theta);
\]

that is, the lower interface is fully determined by the upper-interfacial geometry up to a horizontal and vertical shift. In the long-wave case, we have also

\[
g(x, t) \approx h(x, t) - x_0(\theta) \frac{\partial h}{\partial x} - h_0(\theta),
\]
which is convenient. While the above is true to leading order for vanishingly-small perturbations, it is possible for $g(x, t)$ to appear as a slightly “reduced amplitude” or “flattened” version of $h(x, t)$ depending on the choice of function $P(x, z)$ whose level curves are identified with the amorphous-crystalline interface, where the “flattening” depends weakly on the wavenumber. This would lead to an amorphous-crystalline interface of the form

$$g(x, t) \approx q_0(\theta) \left( h(x, t) - x_0(\theta) \frac{\partial h}{\partial x} \right) - h_0(\theta), \quad (6.74)$$

which was obtained in Chapter 5. This effect is due to flux dilution across sloped surfaces producing regions in the bulk where the level curves of deposited energy (or ions, etc.,) do not track the free interface exactly; if flux dilution is (nonphysically) neglected for the purposes of $P(x, z)$, or in the case of vanishingly-small perturbations, this flattening should not occur (and we have simply $q_0(\theta) \to 1$). We refer the reader to details of calculation in Chapter 5 and its Appendix.

Under this specialization of the amorphous-crystalline interface, and restoring dimensionality (hence dropping tildes), we obtain

$$h_t = C_{APF} + C_{SER}, \quad (6.75)$$

where

$$C_{APF} = -3fA \left( h_0 - (q_0 - 1)h + q_0x_0h_x \right) \left( -\cos(2\theta)(h_0 - (1 + q_0)h + q_0x_0h_x)h_{xx} \right. \\
\left. - (2\cos(2\theta)h_x - \sin(2\theta)) \left( (q_0 - 1)h_x + q_0x_0h_{xx} \right) \right) \quad (6.76)$$

is the term due to anisotropic plastic flow and

$$C_{SER} = -\frac{\gamma}{3\eta} \left( h_0 - (q_0 - 1)h + q_0x_0h_x \right) \left( 2\left( -((q_0 - 1)h_x) + q_0x_0h_{xx} \right) \times \\
\left( 3h_xh_{xx} + (h_0 - (q_0 - 1)h + q_0x_0h_x)h_{xxx} \right) \right. \\
\left. + (h_0 - (q_0 - 1)h + q_0x_0h_x) (3h^2_{xx} - (q_0 - 4)h_xh_{xxx} \right. \\
\left. + q_0x_0h_xh_{xxx} + (h_0 - (q_0 - 1)h + q_0x_0h_x)h_{xxxx} \right) \right) \quad (6.77)$$

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is the term due to surface energy relaxation (i.e., the nonlinear equivalent of the term introduced in the linear stability case by Orchard [27]).

6.3. Simulation of weakly nonlinear evolution equation

With the numerical method described in the Appendix (briefly: second-order discretizations in space and time, Newton’s method to solve the resulting nonlinear system of equations, and fourth-order numerically-approximated Jacobian), we perform a few simple simulations of the above, Equations (6.75)-(6.77), one of which we show in Figure 6.1 as “proof of concept”.

![Figure 6.1](image)

Figure 6.1. Simulation of nonlinear evolution due to Equations (6.75)-(6.77) in the fictitious case that $q_0 = 1$, $h_0 = 3\text{nm}$, $x_0 = 1\text{nm}$, $fA = .1\frac{1}{s}$, $\gamma = 1.36\frac{1}{\text{m}^2}$, $\eta = .6\ \text{GPa}\cdot\text{s}$. Horizontal and vertical length scales above are in nm. Initial conditions are Gaussian white noise with mean 0, standard deviation $\frac{1}{10}$. We have taken discretization $\Delta x = \frac{1}{2}$, $\Delta t = \frac{1}{100}$.

6.4. Application to a radially-symmetric nanopore wall irradiated from above

6.4.1. APF in a multi-scale problem

Above, we have derived equations for the weakly-nonplanar stress-driven evolution of an ion-irradiated thin film — that is, the dynamics of an amorphous layer evolving about an approximately planar patch of surface. Such amorphous layers are often only a few nanome-
ters in depth; for example, 250eV Ar$^+$ → Si produces an amorphous layer of about 3.2nm under normal-incidence irradiation [78]. However, the amorphous layer exists atop a crystalline substrate, which may be strongly nonplanar at the scale of hundreds or thousands of nanometers. This naturally draws attention to a multiscale problem: how will the dynamics of the thin, amorphous layer influence the evolution of a much larger-scale structure? Here, as a relatively simple toy problem, we will consider a low-curvature nanopore irradiated uniformly from above, and we will develop the evolution equation of the nanopore in terms of erosion and the stress-driven dynamics.

We will now distinguish between the local coordinates native to the weakly-nonlinear evolution equation and the global coordinates native to the large-scale nanostructure: the latter we will denote as $H$ and $X$, and the notation of the former will be retained as above. For simplicity, we will consider only the “left” wall of the radially-symmetric nanopore (so that we avoid the need to track $\text{sgn}(H_X)$); due to radial symmetry, this is sufficient.

We observe that while the local film thickness is small relative to the curvature, the local $x$-derivative is approximated by the global arc-length derivative,

$$\frac{\partial}{\partial x} \to \frac{1}{\sqrt{1 + H_X^2}} \frac{\partial}{\partial X} = \frac{\partial}{\partial S}; \quad (6.78)$$

a much fuller derivation is given in either of [10, 17]. The “vertical” velocity of $h$ from the local frame is then identified with the normal velocity of $H$,

$$h_t \to \frac{H_t}{\sqrt{1 + H_X^2}} \quad (6.79)$$

and, similarly,

$$h \to \frac{H}{\sqrt{1 + H_X^2}}, \quad (6.80)$$

reflecting that heights with respect to $z$ are now taken as projections along $\hat{k}$, the vertical axis in global coordinates.

We apply the transformations described above to Equations (6.75)-(6.77) and append erosion, resulting in

$$\frac{H_t}{\sqrt{1 + H_X^2}} = C_{APF,\text{global}} + C_{SER,\text{global}} - \cos(\theta_{loc})Y(\theta_{loc}), \quad (6.81)$$

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with
\[
C_{APF,\text{global}} = -3 f A \left( h_0 - (q_0 - 1) \frac{H}{\sqrt{1 + H^2_X}} + q_0 x_0 \frac{\partial}{\partial S} \frac{H}{\sqrt{1 + H^2_X}} \right) \\
\times \left( -\cos(2\theta) \left( h_0 - (-1 + q_0) \frac{H}{\sqrt{1 + H^2_X}} + q_0 x_0 \frac{\partial}{\partial S} \frac{H}{\sqrt{1 + H^2_X}} \right) \frac{\partial^2}{\partial S^2} \frac{H}{\sqrt{1 + H^2_X}} \\
- (2 \cos(2\theta) \frac{\partial}{\partial S} \frac{H}{\sqrt{1 + H^2_X}} - \sin(2\theta)) \left( - ((q_0 - 1) \frac{\partial}{\partial S} \frac{H}{\sqrt{1 + H^2_X}} + q_0 x_0 \frac{\partial^2}{\partial S^2} \frac{H}{\sqrt{1 + H^2_X}} \right) \right) 
\]

is the term due to anisotropic plastic flow and
\[
C_{SER,\text{global}} = -\frac{\gamma}{3\eta} \left( h_0 - (q_0 - 1) \frac{H}{\sqrt{1 + H^2_X}} + q_0 x_0 \frac{\partial}{\partial S} \frac{H}{\sqrt{1 + H^2_X}} \right) \\
\times \left( 2 \left( - ((q_0 - 1) \frac{\partial}{\partial S} \frac{H}{\sqrt{1 + H^2_X}} + q_0 x_0 \frac{\partial}{\partial S} \frac{H}{\sqrt{1 + H^2_X}} \right) (3 \frac{\partial}{\partial S} \frac{H}{\sqrt{1 + H^2_X}} \frac{\partial^3}{\partial S^3} \frac{H}{\sqrt{1 + H^2_X}} \\
+ (h_0 - (q_0 - 1) \frac{H}{\sqrt{1 + H^2_X}} + q_0 x_0 \frac{\partial}{\partial S} \frac{H}{\sqrt{1 + H^2_X}} \right) (3 \frac{\partial^2}{\partial S^2} \frac{H}{\sqrt{1 + H^2_X}})^2 \\
- (q_0 - 4) \frac{\partial}{\partial S} \frac{H}{\sqrt{1 + H^2_X}} \frac{\partial^3}{\partial S^3} \frac{H}{\sqrt{1 + H^2_X}} \\
+ q_0 x_0 \frac{\partial^2}{\partial S^2} \frac{H}{\sqrt{1 + H^2_X}} \frac{\partial^3}{\partial S^3} \frac{H}{\sqrt{1 + H^2_X}} \\
+ (h_0 - (q_0 - 1) \frac{H}{\sqrt{1 + H^2_X}} + q_0 x_0 \frac{\partial}{\partial S} \frac{H}{\sqrt{1 + H^2_X}} \frac{\partial^4}{\partial S^4} \frac{H}{\sqrt{1 + H^2_X}} \right) \right). 
\]

In the above, it is important to note that $h_0, x_0, q_0$ and $\eta$ depend locally on $\theta$ (recalling the “ion-enhanced fluidity” phenomenon described elsewhere [46, 28], where defects produced by ion-implantation and their bimolecular annihilation directly modify the film’s ability to relax stress); we therefore require some notion of $\theta_{\text{loc}}$ in the global setting. This is obtained simply by realizing that the local angle of incidence of the beam is the angle between the surface normal and the beam in global coordinates. Taking the beam with direction away from the surface $\vec{b} = <\sin(\phi), \cos(\phi)>$ and $\phi = 0$ due to uniform irradiation into the pore, and a patch of surface on the “left” wall of the nanopore with surface normal $\vec{N} = \frac{<-H_X,1>}{\sqrt{1 + H^2_X}},$
we find

$$\cos(\theta_{\text{loc}}) = \frac{1}{\sqrt{1 + H_X^2}}. \quad (6.84)$$

Right-angle trigonometry then supplies

$$\sin(\theta_{\text{loc}}) = \frac{|H_X|}{\sqrt{1 + H_X^2}}. \quad (6.85)$$

Since $fA$ and $\frac{1}{\eta}$ both scale with $\cos(\theta_{\text{loc}})$, we may eliminate the divisor on the left-hand side of Equation (6.81).

### 6.4.2. On wall velocity

We would like to understand whether the evolving amorphous layer is capable of influencing the velocity of a nanopore wall. We follow the technique of [107, 111, 112]: we will take Equation (6.81) following the elimination of the divisor on the left-hand side with the flux-dilution on the right, differentiate both sides with respect to $X$, and seek traveling waves in the slope field $B = H_X$. That is, we take

$$H_t = C_{APF,\text{global}} + C_{SER,\text{global}} - Y(\theta_{\text{loc}}), \quad (6.86)$$

differentiate and convert to $B = H_X$ as the dynamical variable,

$$B_t = \frac{\partial}{\partial X} C_{APF,\text{global}} + \frac{\partial}{\partial X} C_{SER,\text{global}} - \frac{\partial}{\partial X} Y(\theta_{\text{loc}}). \quad (6.87)$$

Then we consider solutions of the form $B(X - ct) = B(p)$, so that we have

$$-cB_p = \frac{\partial}{\partial p} C_{APF,\text{global}} + \frac{\partial}{\partial p} C_{SER,\text{global}} - \frac{\partial}{\partial p} Y(\theta_{\text{loc}}). \quad (6.88)$$

In order to isolate velocity of propagation $c$, we integrate both sides from $p = -\infty$ to $p = \infty$, and we obtain

$$-c(B_L - B_R) = C_{APF,\text{global}}|_\infty^{-\infty} + C_{SER,\text{global}}|_\infty^{-\infty} - Y(B_L) + Y(B_R), \quad (6.89)$$

hence

$$c = -\frac{C_{APF,\text{global}}|^{B_R}_{B_L} + C_{SER,\text{global}}|^{B_R}_{B_L}}{(B_L - B_R)} + \frac{Y(B_L) - Y(B_R)}{(B_L - B_R)} \quad (6.90).$$
and the question of wall velocity is reduced, essentially, to limit evaluation. Because we are concerned with the “left wall” of a radially-symmetric nanopore, we have the slope \( B_L = 0 \) at \( X = -\infty \), and the limit at \( B = B_R \) remains.

We encounter numerous expressions of the form

\[
\left( \frac{1}{\sqrt{1 + H_X^2}} \frac{\partial}{\partial X} \right)^n \frac{H}{\sqrt{1 + H_X^2}}
\]

throughout the PDE prior to seeking traveling wave solutions. Since we have chosen to express the traveling wave equation in terms of the slope field, we consider

\[
H(x, t) = \int_{\alpha}^{X} B(\tilde{X}, t) d\tilde{X} + H(\alpha, t),
\]

where \( \alpha \) is a constant we will choose strategically.

This introduces a problem not seen in [111, 112]: the appearance of undifferentiated \( H \) throughout the traveling wave equation, which will occur as a result of the usual product rule from calculus. Hence the traveling wave equation is, on its face, an integro-differential boundary value problem with limits at infinity, and the velocity is given by a mix of derivatives of \( B \) and improper integrals of \( B \).

However, a simplification is available because we are primarily concerned with a specific geometry: that of the nanopore. If we assume, as in [107, 111, 112] that as \( X \to \infty \), \( B \to B_R, B_X \to 0, B_{XX} \to 0 \), i.e., that the wall is eventually linear with specified slope \( B_R \), tractability is restored. In summary, we consider

\[
H(X, t) = \int_{\alpha}^{X} B(\tilde{X}, t) d\tilde{X} + H(\alpha, t)
\]

and we seek solutions \( B(X, t) \to B(X - ct) \), so that

\[
H(X, t) = \int_{\alpha}^{X} B(\tilde{X} - ct) d\tilde{X} + H(\alpha, t).
\]

The substitution \( \tilde{X} - ct \to p \) leads to

\[
H(X, t) = \int_{0}^{p} B(\tilde{p}) d\tilde{p} + H(ct, t)
\]
after choosing $\alpha = ct$. Now we observe that if we have a traveling wave solution, then $H(ct, t)$ is a constant. Also, as $p \to \infty$, we are assured that $B \to B_R$, so that the nanopore wall becomes linear. Then we consider

$$\int_0^p B(\tilde{p})d\tilde{p} = \int_0^\beta B(\tilde{p})d\tilde{p} + B_R(p - \beta)$$

(6.96)

by taking $B \approx B_R$ beyond $p = \beta$. When $q_0 = 1$, it is then tedious but ultimately straightforward to calculate

$$C_{SER,global}^{B_R}|_{B_L} = C_{APF,global}^{B_R}|_{B_L} = 0,$$

(6.97)

which implies that thin-film viscous effects will not affect wall velocity. We note that $q_0 \neq 1$ is a much more complicated case and will require a more rigorous treatment of the amorphous-crystalline interface than is appropriate for the present work, which is of a more preliminary nature.

6.4.3. On selected wall geometries

Following from [107, 111, 112], we note that Equation 6.88 can be integrated to produce an integro-differential equation. This may be equipped with boundary conditions $B(+\infty) = B_R, B(-\infty) = B_L, B'(+\infty) = 0, B'(-\infty) = 0, B''(+\infty) = 0, B''(-\infty) = 0$ and solved numerically to study whether the presence of APF can alter the admissibility of a given wall geometry as a solution, which could possibly modify the results of [112]. The need to numerically solve a higher order, nonlinear integro-differential equation with boundary conditions at $\pm \infty$ will pose an interesting problem.

6.5. Conclusions

6.5.1. Contribution

Here, we have derived a set of nonlinear equations that describe the long-time evolution of amorphous thin films with a generalized treatment of the amorphous-crystalline interface and the possibility of spatially-inhomogeneous mechanism strength. This provides a staging
point for comparing SEM and TEM images from experiments to theoretical predictions. Our simulations on one axis are broadly compatible with the structures observed under high fluences [62, 48].

We note that even if it turns out to be the case that stress-induced nonlinear patterning is not interesting in its own right, it is possible that stress-based techniques could serve as a pre-patterning step to more complex procedures such as those described in [111, 112, 106], where it is suggested that small-scale structures can be “collided” to produce desired geometries not otherwise attainable.

6.5.2. Future directions

The present work is preliminary to many possible future directions. While we have shown the general form of a system of equations appropriate for the nonlinear evolution of a stress-driven thin film, we have only derived and studied a much-simplified case where we neglect isotropic swelling and the influence of the phase-change at the amorphous-crystalline boundary. Both of these effects will certainly influence nonlinear evolution, especially when the $\Delta$ scalar field is coupled with the ongoing irradiation process: while isotropic swelling will continue to proceed isotropically, it may not occur at the same rate across all regions of the amorphous bulk, especially as nonlinear evolution leads to potentially complex topographies. We therefore identify several areas of future progress:

**On the amorphous-crystalline interface in the nonlinear regime.** In the discussion of the nanopore wall above, as well as the statement of the one-dimensional evolution equation, we have neglected the phase-change boundary condition introduced in Chapter 4. It would be interesting to study the evolution equation with this condition included, especially when $x_0, h_0, q_0$ are derived from a fuller treatment of the amorphous-crystalline boundary than the small-perturbation limit of Chapters 4 and 5 (although we have calculated elsewhere that the expressions for $x_0, h_0, q_0$ should be valid for long-wave but non-vanishing surface variation as well).
Furthermore, although the amorphous-crystalline interface certainly tracks the free interface in a manner seemingly well-described by our previous work, it is possible that at low energies, grazing incidence, small fluxes, or high temperatures (including those induced by the irradiation process itself, see [44, 47]) recrystallization is rapid enough to compete with the amorphization process and introduce a delayed response, or other phenomena. For strictly planar boundaries, there already exists a body of work that has studied the precise relationship between energy deposition, the amorphization threshold, and the growth rate of the boundary: [86, 88, 89, 90, 91, 92], and others. In principle, it should be possible to unify the treatment of the nonplanar boundaries introduced in this thesis with the differential equations proposed by [86, 88, 89, 90, 91, 92], where our treatment of the amorphous-crystalline interface is effectively the “rapid-amorphization” limit of a fluence-dependent system of delay differential equations. Whether or not the results lead to significant alteration of the present nonlinear equations for experimentally-typical fluxes and energies remains to be seen, but they may be mathematically interesting nonetheless.

**On spatially-resolved nonlinear IIS.** Above, we have derived general weakly-nonlinear equations capable of including spatially-resolved IIS as an additional, evolving scalar field $\Delta$. This leads to additional complications and the need to consider a two-field model, which requires a numerical solution. However, we might anticipate that the results would be interesting; in [49], a stress-based nonlinear equation was derived based on the *effective body force* class of models [31, 33], which produced interesting surface morphologies when a downbeam-varying stress was introduced. While in Chapter 5 we showed that spatially-resolved APF is apparently unnecessary to explain most of the linear-regime data for pattern-forming systems across a range of energies, we require spatially-resolved IIS. Hence a natural next question is whether spatially-resolved IIS substantially modifies the behavior of our nonlinear evolution equations. We note that the work of [49] was done with a flat amorphous-crystalline interface, obscuring the possible interactions between bulk stresses and that interface (which were shown to be important throughout the rest of this thesis).


7.1. Contribution

Here, we will briefly summarize overall contributions from this work:

1. Chapter 2 demonstrates that the swelling mechanism proposed in [48] continues to contribute to thin film stability outside of the analytically-tractable “small swelling-rate” limit, and suggests that the form of the dispersion relation at higher swelling rates is consistent with that at lower swelling rates. This work resulted in one publication [50].

2. Chapter 3 develops a very general expression for growth rates of long-wave perturbations to amorphous films where both anisotropic plastic flow (APF) and ion-induced isotropic swelling (IIS) are active. The growth rate calculation accommodates arbitrary spatial distributions of each mechanism and handles the amorphous-crystalline interface in general, and suggests a previously-unappreciated role for the shape of the amorphous-crystalline interface in $\theta_c$ selection. At the time of writing, this work has been accepted for publication.

3. Chapter 4 develops a simple means of connecting the amorphous-crystalline interface to parameters that are easily obtained from SRIM [78] or other simulations tools. We also introduce a phase-change boundary condition based on the change in density across the amorphous-crystalline interface; to our knowledge, this condition has not been previously explored in the context of ion-induced nanopattern formation. We show that it is capable of strongly influencing $\theta_c$ and may account for the observed “nonuniversality” of $\theta_c$. It also suggests the possibility of a rapid increase in $\theta_c$ in the
vicinity of 1keV $\text{Ar}^+ \to \text{Si}$, which is experimentally observed, but lacks a consensus explanation at the time of writing.

4. In Chapter 5, we take full advantage of the capability, originally developed in Chapter 3, to assign spatial variation to each of APF and IIS, which we seek to connect to the distribution of ions within the irradiated film. We show that this “spatially-resolved” treatment of IIS via the ion-implantation distribution allows for a nonlinear fit of $\theta_c$ for 8 different experiments involving irradiated Si and 7 different experimental systems involving irradiated Ge with two fit parameters allowed per target species. We then show that the fit parameters that produce agreement with $\theta_c$ across all of these systems also produce surprisingly strong agreement with angle-dependent wavelengths $\lambda(\theta)$, even though $\lambda(\theta)$ was not involved in the nonlinear fit. The resulting values also lead to order-of-magnitude agreement with other experimentally-observed values, such as in-plane stresses found by [44, 47]. Taken together, this provides strong evidence for ion-induced stresses as a major contributor to nanoscale pattern formation for at least the noble-gas ion irradiation of monatomic, Group IV semiconductor targets.

5. In Chapter 6, we derive the general nonlinear evolution equations for the composite model of APF and IIS with arbitrary amorphous-crystalline interface and the phase-change boundary condition developed earlier. We perform some simple simulations of these equations in one spatial dimension and consider the implications of the model for a certain multiscale problem of interest.

7.2. Future directions

We see several opportunities for future work to follow on advances made here.

1. We have suggested a scaling argument for IIS and APF that appear to lead to reasonable agreement with $\theta_c$ and $\lambda(\theta)$ for multiple systems in the 250eV-2keV energetic range. While we suspect that IIS physically originates from the interstitial distribution, our attempts at “localizing” APF to the distribution of ions or recoils in the amorphous layer lead to worse agreement with experimental results. This suggests
that either APF is somehow a nonlocal phenomenon, or it is connected to some other quantity besides ions and recoils; however, the ion and recoil distributions serve as rather robust approximations for energy deposition and “damage” in a general sense. These results are interesting and prompt a more detailed analysis. Identification of the underlying physics of APF remains a high research priority.

2. As a simplification, we have not considered the possibility that APF and IIS interact. This is partially because we are, at the present time, uncertain as to the underlying physical cause of APF. However, observation of trends discussed in Chapter 5 leads us to wonder whether APF and IIS may be competitive: that is, APF is somehow weakened by the presence of IIS. This suspicion is motivated by the observation that when the distribution of implanted ions is relatively close to the free interface, patterns are strongly suppressed, as in Ne\(^+\), Ar\(^+\) \(\rightarrow\) Ge. This is not accounted for by the model presented in Chapter 5, which seems to correctly identify several trends and provide near-quantitative agreement across multiple systems.

3. The approach to spatial resolution of general, deposited quantities in the bulk is suitable to binary problems and reactive materials. It is not difficult to imagine multiple scalar fields distributed according to integrals similar to those in Chapter 5 where the resulting scalar fields are allowed to react, advect, and diffuse in response to, e.g., energy deposited. This approach is also amenable to allowing stressed material to sputter away, as was done in [42, 46].

4. The work of [28] studied the relative contributions of different terms in a full-spectrum linear dispersion relation to GISAXS-inferred growth rates. This theoretical-experimental collaboration advanced the hypothesis that erosion and stress-based phenomena, such as APF, coexist and govern film growth together. However, the results of [28] were performed on the basis of the amorphous-crystalline interface as a vertical translation of the free interface, in the absence of any IIS, and with the standard no-slip, no-penetration boundary conditions. It would be interesting to revisit these results with full-spectrum linear dispersion relations for APF and spatially-resolved IIS with the BCA-informed amorphous-crystalline interface shape, as well as the phase-change
boundary condition considered in Chapter 4. We anticipate that the inclusion of these new effects would substantially modify the conclusions of [28]. We note that spatially-resolved IIS is unlikely to admit a closed-form solution in its full-spectrum analysis (observing the difficulty of obtaining the full-spectrum result from [48] for the constant-IIS case), which will likely necessitate a numerical or semi-analytical approach. We anticipate that this numerical work, alone, will be highly nontrivial but potentially quite valuable, and connecting the numerical work to the nonlinear fitting technique used in [28] would also be significant.

5. The work in this thesis appears to strongly support a stress-based explanation for several phenomena observed in low-energy ion-bombardment of amorphizable materials. However, especially with respect to APF, no first-principles model has been established. For this reason, it would be interesting—and we regard it as a high research priority—to determine the underlying cause for “phenomenological” APF. This work points in the direction of an implantation-based, nonlocal phenomenon (Chapter 5), which recalls the work of [105].

6. Essentially all of this thesis has involved the real part of the linear dispersion relation; we have given almost no consideration of the imaginary part, which is associated with ripple velocity [6]. Elsewhere [100], it has been suggested that the stress-based model of [33] would appear to lead to disagreement with experimental results. An immediate next step would be in the comparison of these experimental results with our own model.

7. We have derived the weakly-nonlinear evolution equations for a composite of APF and IIS. Subject to a few simplifications, we are able to obtain relatively benign nonlinear PDEs amenable to finite-differences and multivariate Newton’s method. Although not shown in this thesis, the parametric dependence of these equations on collision cascade parameters $a, \alpha, \beta$, passed into the equations via $x_0(\theta), h_0(\theta)$, can significantly modify thin film behavior, even without taking into consideration the phase-change boundary condition or the scaling of parameters with energy or momentum. These observations will likely form a rich testbed for comparisons of theory and experiment, especially given recent work by [99, 100], which suggests the insufficiency of the redistributive-
erosive framework for nonlinear evolution. In their work, there are early indications that something like viscous flow may be influential. While they note that at least one other stress-based model [33] appears to give a wrong prediction of translation velocity, it would be interesting to see what the model described in Chapter 6 suggests.

8. We have not yet fully explored the implications of the work from Chapter 6 for multiscale problems. In particular, while we have ruled out viscous flow effects on wall velocity for a simplified case that excludes IIS and the phase-change boundary condition, these effects may be rather significant. It also appears rather likely that the parameter values associated with APF and IIS will be capable of modifying the geometries that admit traveling wave solutions in the slope field, even if they do not directly modify the wall velocity. This may lead to an exploitable feature of the system.
Appendix A
Appendix to Chapter 2

A.1. Long-wave limit, arbitrary swelling rate

As a simple first step in determining whether the swelling mechanism continues to produce a regime of stabilizing wave numbers for arbitrary swelling rates, we consider an alternate second linearization of the $O(\epsilon)$ equations, in small wavenumber $Q$, rather than in small swelling rate $A$, in order to understand the long-wave behavior of the swelling mechanism in general. We expand these equations in a standard asymptotic series for small $Q$ by applying the following set of ansatzes

$$
\rho_1(z) = \sum_{i=0}^{\infty} Q^i \rho_{1i}(z); \quad u_1(z) = \sum_{i=0}^{\infty} Q^i u_{1i}(z); \quad w_1(z) = \sum_{i=0}^{\infty} Q^i w_{1i}(z)
$$

$$
p_1(z) = \sum_{i=0}^{\infty} Q^i p_{1i}(z); \quad a_1(z) = \sum_{i=0}^{\infty} Q^i a_{1i}(z); \quad \Sigma = \sum_{i=0}^{\infty} Q^i \Sigma_i,
$$

deriving new sets of differential equations in the coefficients and solving them. The results will then provide good approximations to the $\rho_1, u_1, w_1, p_1, a_1$ and $\Sigma$ that we seek when $Q \approx 0$. Although the system, in principle, will admit multiple solutions for $\Sigma$, we are usually concerned only with the most positive branch, since this will determine the pattern formation that is actually observed. This suggests that we should begin the analysis by seeking perturbation solutions near $\Sigma_0 = 0$. We now provide a brief summary of the solution up to $O(Q^2)$ with this assumption about $\Sigma_0$. We will refer to this as the “main branch”.

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At O(1): The \( i = 0 \) term may be computed by setting \( Q \to 0, \Sigma_0 \to 0 \) for equations (2.9) - (2.19). This leads to the bulk equations

\[
-\frac{A}{\psi^3} w_{10} + \frac{w'_{10}}{\psi} + \psi \rho'_{10} + \frac{A}{\psi} \rho_{10} = 0 \quad (A.1)
\]

\[
\psi a'_{10} + \frac{A}{\psi} w_{10} = 0 \quad (A.2)
\]

\[
\psi \rho_{10} + \frac{a_{10}}{\psi} = 0 \quad (A.3)
\]

\[
u'_{10} = 0 \quad (A.4)
\]

\[
2w''_{10} = p'_{10}, \quad (A.5)
\]

with

\[
a_{10} = -h_{11} A \quad (A.6)
\]

\[
u_{10} = 0 \quad (A.7)
\]

\[
w_{10} = -h_{11} A \quad (A.8)
\]

at \( z=0 \), and

\[
w_{10} = -(1 + 2A) \rho_{10} \quad (A.9)
\]

\[
u'_{10} = 0 \quad (A.10)
\]

\[
-p_{10} + 2w'_{10} = 0 \quad (A.11)
\]

at \( z=1 \). It is clear that \( u_{10} \) is at most a polynomial. From equations (A.2) and (A.3), we may obtain \( a_{10} \) in terms of \( \rho_{10} \), and then \( w_{10} \) is known in terms of the \( \rho_{10} \) by substitution. These relationships may be applied to (A.1) to obtain a differential equation purely in terms of \( \rho_{10} \), which is readily solvable. Having obtained \( \rho_{10} \), we know \( a_{10} \) from (2.11) and \( w_{10} \) from having already identified \( w_{10} \) in terms of \( a_{10} \). Then \( p_{10} \) may be obtained from (A.5), and the boundary conditions are easy to match. We obtain the following solution set at O(1) for the main branch:
\[ w_{10} = \frac{-Ah_{11}}{\sqrt{1 + 2Az}} \]  
\[ u_{10} = 0 \]  
\[ p_{10} = \frac{2A^2h_{11}}{(1 + 2Az)^{3/2}} \]  
\[ \rho_{10} = \frac{Ah_{11}}{(1 + 2Az)^{3/2}} \]  
\[ a_{10} = \frac{-Ah_{11}}{\sqrt{1 + 2Az}} \]  
\[ \Sigma_0 = 0 \]  

**At O(Q):** We obtain the bulk equations

\[ \Sigma_1 \rho_{10} - \frac{A}{\psi^3} w_{11} + \frac{w'_{11}}{\psi} + \psi \rho'_{11} + \frac{A\rho_{11}}{\psi} = 0 \]  
\[ \Sigma_1 a_{10} + \psi a'_{11} + \frac{A w_{11}}{\psi} = 0 \]  
\[ \psi \rho_{11} + \frac{a_{11}}{\psi} = 0 \]  
\[ u''_{11} + iw'_{10} = -2iAh_{21} \sqrt{1 + 2A} \]  
\[ p'_{11} = 2w''_{11} \]

with boundary conditions at z=0:

\[ a_{11} = 0 \]  
\[ u_{11} = 0 \]  
\[ w_{11} = 0 \]

and boundary conditions at z=1:

\[ w_{11} = \Sigma_1 h_{21} - (1 + 2A)\rho_{11} \]  
\[ u'_{11} + iw_{10} = \frac{-2iAh_{21}}{\sqrt{1 + 2A}} \]  
\[ -p_{11} + 2w'_{11} = 0 \]
from which we obtain

\[ a_{11} = w_{11} = \rho_{11} = p_{11} = \Sigma_1 = 0 \]  (A.29)

\[ u_{11} = i(h_{11} + \frac{2A(h_{11} - h_{21})z}{\sqrt{1 + 2A}} - h_{11}\sqrt{1 + 2Az}) \]  (A.30)

As before, the primary “trick” is in exploiting the linearized equation of state. Here, we use

equation (A.22) to determine that \( p_{11} \) is a constant, and (A.21) can be integrated twice to provide the \( u_{11} \) in terms of the \( O(1) \) results. (A.20) gives a relationship between \( a_{11} \) and \( \rho_{11} \) which can be substituted into (A.19) to obtain \( w_{11} \) in terms of \( O(1) \) results and \( \rho_{11} \). This

may be substituted into (A.18) to arrive at a differential equation in terms of \( \rho_{11} \) which is solvable in closed form. We may then back-substitute to obtain the solutions for all other

quantities.

At \( O(Q^2) \): We obtain the bulk equations

\[ \Sigma_2 \rho_{10} + \Sigma_1 \rho_{11} - \frac{Aw_{12}}{\psi^3} + \frac{1}{\psi}(iu_{11} + w'_{12}) + \psi \rho'_{12} + \frac{A}{\psi} \rho_{12} = 0 \]  (A.31)

\[ \Sigma_2 a_{10} + \Sigma_1 a_{11} + \psi a'_{12} + \frac{Aw_{12}}{\psi} = 0 \]  (A.32)

\[ \psi \rho_{12} + \frac{a_{12}}{\psi} = 0 \]  (A.33)

\[ -2u_{10} + u''_{12} + iw'_{11} = ip_{11} \]  (A.34)

\[ -w_{10} + 2w''_{12} + iu'_{11} = p'_{12} \]  (A.35)

with boundary conditions at \( z=0 \):

\[ a_{12} = 0 \]  (A.36)

\[ u_{12} = 0 \]  (A.37)

\[ w_{12} = 0 \]  (A.38)

\[ (A.39) \]
and boundary conditions at z=1:

\[ w_{12} = h_{21} \Sigma_{12} - (1 + 2A)\rho_{12} \]  
\[ u_{12}' + iw_{11} = 0 \]  
\[ -p_{12} + 2w_{12}' = 0 \]

from which we may finally obtain \( \Sigma_{2} \). The solution method is similar to the first two orders of \( Q \). From (A.35) we obtain \( p_{12} \) in terms of known quantities. (A.34) may be integrated twice to obtain \( u_{12} \) in terms of known quantities. Then we may solve for \( a_{12} \) in terms of \( \rho_{12} \) and known quantities using (A.34). Using (A.33) as before, we obtain \( w_{12} \) in terms of \( \rho_{21} \) and known quantities, which may be substituted into (A.31) to arrive at a differential equation that is solvable for \( \rho_{21} \), and the other quantities may be obtained by back-substitution. The boundary conditions, as before, are easy to handle although the calculations are somewhat lengthy. It is at \( O(Q^2) \) that we obtain our first non-zero solution in the small-Q approximation of \( \Sigma \).

**Results.** Therefore we have in the long-wave limit, to leading order in \( Q \),

\[ \Sigma \approx Q^2(2(-2A^2 + A - \sqrt{2A+1} + 1)h_{21} + 5(\sqrt{2A+1} - 1)h_{11} + A(-2A + 6\sqrt{2A+1} - 11)h_{11}) \]
\[ 3A((2A+1)h_{11} + \sqrt{2A+1}(h_{21} - h_{11})) \]

which is proportional to \(-Q^2\), and this gives us a good approximation of the main branch of the dispersion relation for small wave numbers. As described above, the dependence on incidence angle can be explored by re-scaling the film depth, wave number, and linearized interface perturbations \( (h_0, Q, h_{11}, h_{21} \) respectively) using Eqs. (2.32)-(2.34). Dividing numerator and denominator by \( h_{21} \), applying those substitutions, and then performing a Taylor expansion for \( Q \approx 0 \), we deduce the angle- and swelling rate-dependent long-wave solution

\[ \text{Re}[\Sigma(Q, \theta; A)] \approx \frac{-A + \sqrt{2A+1} - 1}{A} Q^2 \cos^2(\theta) \]

and

\[ \text{Im}[\Sigma(Q, \theta; A)] \approx -\frac{\sqrt{2A+1} + A(2\sqrt{2A+1} - 3) - 1}{3A\sqrt{2A+1}} Q^3 \sin(\theta) \cos^3(\theta) \]
Figure A.1. The $A$-dependent, angle-independent coefficient from (a) the real part and (b) the imaginary part of the long-wave dispersion relation, from equations (A.44) and (A.45) which are much simpler forms accounting for the growth and translation, respectively, of long-wave perturbations. We note that a second Taylor expansion in $A$ for $A \approx 0$ for (A.44) and (A.45) reproduces the result given in [48],
\[
\text{Re}[\Sigma(Q, \theta, 0)] \approx -\frac{1}{2}AQ^2 \cos^2(\theta). \quad (A.46)
\]
We note that for $\theta = 0$, the imaginary part drops out of our result entirely, consistent with previous results at normal incidence. It is clear that for all values of $A$, small wave numbers are stable, reflecting that the swelling mechanism induces a pocket of stability for some range of long wave numbers at all angles of incidence. Hence the stabilizing effect of the swelling mechanism is established for arbitrary swelling rates, angle of incidence and \textit{at least} long waves. For interest, we have plotted the $A$-dependent coefficients of equations (A.44) and (A.45) above in Figure 2.
Appendix B
Appendix to Chapter 3

B.1. Details of linear stability analysis

Here, we provide a complete solution for the general case of bulk stress modification by APF and IIS.

B.1.1. General equations

Before beginning the analysis, it is convenient to compute in advance the following, as described in the main text:

\[ \alpha(z; 0 + \epsilon g_1, h_0 + \epsilon h_1) \approx \alpha_0(z; 0, h_0) + \epsilon \alpha_\epsilon(z; 0, h_0), \]  
\[ \tau(z; 0 + \epsilon g_1, h_0 + \epsilon h_1) \approx \tau_0(z; 0, h_0) + \epsilon \tau_\epsilon(z; 0, h_0), \]

and it will be understood throughout that when \( \alpha_0, \tau_0, \alpha_\epsilon \) or \( \tau_\epsilon \) appear, the arguments except for \( z \) will be taken as those about which the \( \epsilon \) expansion was performed. Here, we also note the general momentum balance equations in the bulk for the reader’s reference:

\[ -p_x + \eta \{ 2u_{xx} + u_{yy} + v_{xy} + u_{zz} + w_{xz} \} = 2f A_D \eta \{ D_{11} \frac{d\tau}{dx} + D_{13} \frac{d\tau}{dz} \} \]
\[ -p_y + \eta \{ u_{xy} + v_{xx} + 2v_{yy} + v_{zz} + w_{yz} \} = 2f A_D \eta \frac{d\tau}{dy} \]
\[ -p_z + \eta \{ u_{xz} + v_{yz} + w_{xx} + w_{yy} + 2w_{zz} \} = 2f A_D \eta \{ D_{31} \frac{d\tau}{dx} + D_{33} \frac{d\tau}{dz} \} \]

We note that we have not assumed standard incompressibility in order to simplify the above momentum balance equations (i.e., allowing cancellation of several mixed partials).
We will consider only the projected down-beam direction, as this is sufficient for a study of critical angle selection; hence we strip \( y \)-dependence from the following calculations.

\[ \text{B.1.1.1. Steady state and first expansion: } \epsilon \text{-small perturbations} \]

Following the application of the steady-state and translation-invariance assumptions \((\frac{\partial}{\partial x} \to 0, \frac{\partial}{\partial t} \to 0)\), linearization about small perturbations produces the steady-state equations

\[
\begin{align*}
\frac{\partial}{\partial z}(\rho_0 w_0) &= 0 \\
\eta u_{0zz} &= 2\eta f A_D D_{13}\tau_{0,z} \\
p_{0z} + 2\eta w_{0zz} &= 2\eta f A_D D_{33}\tau_{0,z} \\
\rho_0 &= \frac{\rho^*}{1 + \Delta_0} \\
w_0\Delta_{0z} &= \alpha_0(z;0,h_0).
\end{align*}
\]  

(B.4)

At \( z=0 \), we have

\[
\begin{align*}
\Delta_0 &= 0 \\
u_0 &= 0 \\
w_0 &= V,
\end{align*}
\]  

(B.5)

representing the steady-state equations of the no-slip and no-penetration conditions respectively in the downward-translating frame. At \( z=h_0 \), we have

\[
\begin{align*}
0 &= w_0 - V\frac{\rho^*}{\rho_0} \\
u_{0,z} &= 2f A_D \tau_0(z;0,h_0) D_{13} \\
p_0 &= 2\eta\{w_{0z} - f A_D \tau_0(z;0,h_0) D_{33}\},
\end{align*}
\]  

(B.6)

where the first equation is the steady-state equation of the modified kinematic condition, and the second two equations are due to the steady-state stress balance \( T_0 \cdot \hat{n}_0 = 0 \) at the
upper interface. We also find the linearized equations at $O(\epsilon)$ as

$$
\rho_1 t + \rho_0 u_1 x + \rho_0 w_1 z + \rho_1 w_0 z + \rho_1 w_0 = 0
$$

$$
-p_{1x} + \eta \{2u_{1xx} + u_{1zz} + w_{1z}\} = 2fA_D \eta \{D_{11} \tau_{\epsilon,xx} + D_{13} \tau_{\epsilon,zz}\}
$$

$$
-p_{1z} + \eta \{w_{1xx} + 2w_{1zz} + u_{1z}\} = 2fA_D \eta \{D_{31} \tau_{\epsilon,xx} + D_{33} \tau_{\epsilon,zz}\}
$$

(B.7)

$$
\rho_1 = \frac{-\rho^* \Delta_1}{(1 + \Delta_0)^2}
$$

$$
\Delta_{1t} + u_0 \Delta_{1x} + w_0 \Delta_{1z} + w_1 \Delta_{0z} = \alpha_\epsilon
$$

in the bulk, representing conservation of mass and momentum; the equation of state; and the advection of the volume-change respectively. Now at $z = 0$,

$$
\Delta_{0,z}(z; 0, h_0) g_1 + \Delta_1(z; 0, h_0) = 0
$$

$$
u_{0z} g_1 + u_1 = 0
$$

(B.8)

$$w_{0z} g_1 + w_1 = 0.
$$

At $z = h_0$,

$$h_{1t} = w_1 - u_0 h_{1x} + h_1 w_{0z} + \frac{V \rho^*}{\rho_0} \{\rho_0 h_1 + \rho_1\}
$$

$$2\eta \{\frac{1}{2}(u_{1z} + w_{1x}) - fA_D \tau_{\epsilon} D_{13}\} - h_{1x} T_{01}^{11} = 0
$$

(B.9)

$$-p_1 + 2\eta \{w_{1z} - fA_D \tau_{\epsilon} D_{33}\} + T_{0z}^{33} = 0,
$$

where the first equation is due to the linearization of the kinematic condition and the second two are due to the linearization of the stress balance,

$$
T_0 \cdot \hat{n}_1 + \left[ \frac{\partial T_0}{\partial z} \cdot \hat{n}_1 + T_1 \right] \cdot \hat{n}_0 = \vec{0}.
$$

(B.10)

Here, we have $\hat{n}_0 = < 0, 0, 1 >$ and $\hat{n}_1 = < -h_{1x}, -h_{1y}, 0 >$. $T_{01}^{11}$ denotes the upper-left component of steady-state stress tensor $T_0$, and $T_{0z}^{33}$ denotes the bottom-right component. The component indices are denoted as superscripts to distinguish them from the subscripts which elsewhere denote terms in the expansion.

**B.1.1.2. Second expansion: small perturbative wavenumber $k$**

Expanding the scalar fields above in normal modes as described in the main text, we now seek the long-wave linear dispersion relation, since we anticipate that pattern formation in a
unary material should be governed by a so-called Type II bifurcation [6]. This prompts the expansion of the dispersion relation $\sigma \approx 0$ in small wavenumber $k$ as

$$\sigma = 0 + k\sigma_1 + k^2\sigma_2 + O(k^3),$$

(B.11)

and we obtain the following systems at each order in $k$.

At $O(1)$:

$$\rho_0 \tilde{w}_{10} + \rho_0 \tilde{u}'_{10} + \tilde{\rho}_{10} w_0 + \tilde{\rho}_{10} w_{0z} = 0$$

$$\tilde{\omega}'_{10} = 2f A_D D_{13} \tau_{e,z}$$

$$-\tilde{\rho}'_{10} + 2\eta \tilde{w}''_{10} = 2f A_D \eta \{D_{33} \tau_{e,z}\}$$

(B.12)

$$\tilde{\rho}_{10} = \frac{-\rho^* \Delta_{10}}{(1 + \Delta_0)^2}$$

$$w_0 \tilde{\Delta}'_{10} + \tilde{\omega}_{10} \Delta_{0z} = \alpha_\epsilon.$$

At $z = 0$,

$$\Delta_{0z} \tilde{g}_1 + \tilde{\Delta}_{10} = 0$$

$$\tilde{u}_{10} + u_{0z} \tilde{g}_1 = 0$$

$$\tilde{w}_{10} + w_{0z} \tilde{g}_1 = 0$$

(B.13)

At $z = h_0$,

$$\tilde{w}_{10} + h_1 w_{0z} + \frac{V \rho^*}{\rho_0^2} \{\rho_0 \tilde{h}_1 + \tilde{\rho}_{10}\} = 0$$

$$2\eta \{\tilde{w}'_{10} - f A_D \tau_e D_{13}\} = 0$$

$$-\tilde{p}_{10} + 2\eta \{\tilde{w}'_{10} - f A_D \tau_e D_{33}\} + T_{0z}^{33} = 0$$

(B.14)

At $O(k)$:

$$\sigma_1 \tilde{\rho}_{10} + i \rho_0 \tilde{w}_{10} + \rho_0 \tilde{w}_{11} + \rho_0 \tilde{u}'_{11} + \tilde{\rho}'_{11} w_0 + \tilde{\rho}_{11} w_{0z} = 0$$

$$-i \tilde{p}_{10} + \eta \{\tilde{w}''_{11} + i \tilde{u}'_{11}\} = 2f A_D \eta i \{D_{11} \tau_e\}$$

$$-\tilde{p}'_{11} + \eta \{2 \tilde{w}''_{11} + i \tilde{u}'_{11}\} = 2f A_D \eta i \{D_{31} \tau_e\}$$

(B.15)

$$\tilde{\rho}_{11} = \frac{-\rho^* \Delta_{11}}{(1 + \Delta_0)^2}$$

$$\sigma_1 \tilde{\Delta}_{10} + i u_0 \tilde{\Delta}_{10} + w_0 \tilde{\Delta}'_{11} + \tilde{\omega}_{11} \Delta_{0z} = 0$$
At $z = 0$,\[\begin{align*}
\tilde{\Delta}_{11} &= 0 \\
\tilde{u}_{11} &= 0 \\
\tilde{w}_{11} &= 0
\end{align*}\] (B.16)

At $z = h_0$,\[\begin{align*}
\sigma_1 \tilde{h}_1 &= \tilde{w}_{11} - u_0 i \tilde{h}_1 + \frac{V \rho^*}{\rho_0^2} \tilde{\rho}_{11} \\
2\eta \left\{ \frac{1}{2} [\tilde{u}_{11}' + i \tilde{w}_{10}] \right\} - i \tilde{h}_1 T_{011} &= 0 \\
- \tilde{p}_{11} + 2 \eta \tilde{w}_{11}' &= 0
\end{align*}\] (B.17)

At $O(k^2)$:\[\begin{align*}
\sigma_1 \tilde{\rho}_{11} + \sigma_2 \tilde{\rho}_{10} + i \rho_0 \tilde{u}_{11} + \rho_0 \tilde{w}_{12} + \rho_0 \tilde{w}_{12}' + \tilde{\rho}'_{12} w_0 + \tilde{\rho}_{12} w_{0z} &= 0 \\
- i \tilde{p}_{11} + \eta \left\{ -2 \tilde{u}_{10} + \tilde{u}_{12}'' + i \tilde{w}_{11}' \right\} &= 0 \\
- \tilde{p}_{12}' + \eta \left\{ - \tilde{w}_{10} + 2 \tilde{w}_{12}'' + i \tilde{u}_{11}' \right\} &= 0 \\
\tilde{\rho}_{12} &= \frac{- \rho^* \tilde{\Delta}_{12}}{[1 + \Delta_0]^2} \\
\sigma_1 \tilde{\Delta}_{11} + \sigma_2 \tilde{\Delta}_{10} + i u_0 \tilde{\Delta}_{11} + w_0 \tilde{\Delta}_{12}' + \tilde{w}_{12} \Delta_{0z} &= 0
\end{align*}\] (B.18)

At $z=0$,\[\begin{align*}
\tilde{\Delta}_{12} &= 0 \\
\tilde{u}_{12} &= 0 \\
\tilde{w}_{12} &= 0
\end{align*}\] (B.19)

and at $z=h_0$,\[\begin{align*}
\sigma_2 \tilde{h}_1 &= \tilde{w}_{12} + \frac{V \rho^*}{\rho_0^2} \tilde{\rho}_{12} \\
\eta \left\{ \tilde{u}_{12}' + i \tilde{w}_{11} \right\} &= 0 \\
- \tilde{p}_{12} + 2 \eta \tilde{w}_{12}' &= 0
\end{align*}\] (B.20)

\textit{B.1.1.3. Third expansion: small swelling rate fA}$_1$

It quickly becomes apparent that a fully-general solution of the above equations, resulting in the closed-form linear dispersion relation, is not mathematically tractable. In particular,
by combining mass conservation, age-advection and the equation of state, we may obtain
the ordinary differential equation,

$$(1 + \Delta_0)\Delta_0 = \frac{\rho^*}{a_1}\alpha_0(z; 0, h_0).$$

(B.21)

Integration in $z$ leads immediately to

$$\frac{\Delta_0^2}{2} + \Delta_0 - \frac{\rho^*}{c_1} \left( \int_0^z \alpha_0(\tilde{z}; 0, h_0) d\tilde{z} + c_2 \right) = 0,$$

(B.22)

and we obtain

$$\Delta_0 = -1 + \sqrt{1 + \frac{2}{V} \int_0^z \alpha_0(\tilde{z}; 0, h_0) d\tilde{z}}$$

(B.23)

when boundary conditions are considered. A solution may then be determined for $w_0$ in

terms of $\Delta_0$. However, these solutions, as they appear in the $O(\epsilon)$ equations, result in a

system of ordinary differential equations with complicated, nonlinear coefficients, even for

the simple case of $\alpha_0$ linear in $z$.

Bearing in mind that we expect the swelling rate in the traveling frame to be fairly small

(that is, there isn’t much swelling prior to a parcel of matter being sputtered away), and

observing from previous results [50] that the effect of even large swelling rates is highly self-

similar at all wave numbers, and uniformly stabilizing for long waves, we consider a second

expansion in a small mean swelling rate, $fA_I$. Hence we take

$$\alpha_0 = \alpha_{00} + fA_I\alpha_{01} + ...$$

$$\alpha_\epsilon = \alpha_{\epsilon 0} + fA_I\alpha_{\epsilon 1} + ...$$

$$\rho_0 = \rho_{00} + fA_I\rho_{01} + ...$$

$$\rho_\epsilon = \rho_{\epsilon 0} + fA_I\rho_{\epsilon 1} + ...$$

$$\sigma_1 = \sigma_{10} + fA_I\sigma_{11} + ...$$

$$\tilde{\rho}_{10} = \tilde{\rho}_{100} + fA_I\tilde{\rho}_{111} + ...$$

(B.24)

Now, because the equations for the leading order terms are obvious from the above (simply

by appending a “0” to the subscript of each term), we shall only write out explicitly the
equations expanded in \( fA_I \). Because the stress tensor associated with anisotropic plastic flow is assumed, here, to be independent of swelling rate, there is no need for us to expand, say, \( \tau \rightarrow \tau_0 + fA_I \tau_1 \). In a more careful exploration of the interplay between volumization and stress-free strain through local changes in the density, such an expansion would be required.

**Steady state at O(\( fA_I \)):**

\[
\frac{\partial}{\partial z} (\rho_{00} w_{01} + \rho_{01} w_{00}) = 0 \\
\eta u_{01zz} = 0 \\
-p_{01z} + 2\eta w_{01zz} = 0 \\
\rho_{01} = -\frac{\rho^* \Delta_{01}}{(1 + \Delta_{00})^2} \\
w_{00} \Delta_{01z} + w_{01} \Delta_{00z} = \alpha_{01}(z; 0, h_0).
\]

At \( z=0 \):

\[
\Delta_{01} = 0 \\
u_{01} = 0 \\
w_{01} = 0.
\]

At \( z=h_0 \),

\[
w_{01} + V \rho^* \frac{\rho_{01}}{\rho_{00}^2} = 0 \\
u_{01z} = 0 \\
-p_{01} + 2\eta w_{01z} = 0.
\]

At O(\( fA_I \)):

\[
\rho_{00z} \ddot{w}_{101} + \rho_{01z} \ddot{w}_{100} + \rho_{00} \dot{w}_{101z} + \rho_{01} \dot{w}_{100z} + \ddot{\rho}_{100z} w_{01} + \ddot{\rho}_{101z} w_{00} + \ddot{\rho}_{100} w_{01z} + \ddot{\rho}_{101} w_{00z} = 0 \\
\ddot{u}_{101zz} = 0 \\
-p_{101z} + 2\eta \ddot{w}_{101zz} = 0 \\
w_{00} \ddot{\Delta}_{101z} + w_{01} \ddot{\Delta}_{100z} + \ddot{w}_{100} \Delta_{01z} + \ddot{w}_{101} \Delta_{00z} = \alpha_{e1} \\
\ddot{\rho}_{101} = -\frac{\rho^* \{ \Delta_{00} \ddot{\Delta}_{101} + \ddot{\Delta}_{101} - 2\Delta_{01} \ddot{\Delta}_{100} \}}{(1 + \Delta_{00})^3}
\]

(B.28)
At z=0:
\[
\Delta_{01z} g_1 + \ddot{\Delta}_{101} = 0
\]
\[
\ddot{u}_{101} + u_{01z} g_1 = 0
\]
\[
\ddot{w}_{101} + w_{01z} g_1 = 0.
\]  \(\text{(B.29)}\)

At z=h_0:
\[
\ddot{w}_{101} + \ddot{h}_1 w_{01z} + V \rho^* \left\{ \rho_{00}(\rho_{01z} h_1 + \rho_{101}) - 2 \rho_{01}(\rho_{00z} h_1 + \rho_{100}) \right\} = 0
\]
\[
\ddot{u}_{101} = 0
\]
\[
-\ddot{p}_{101} + 2 \eta \dddot{w}_{101} + T_{01z}^{33} = 0.
\]  \(\text{(B.30)}\)

At O(kfA_1):
\[
\sigma_{10} \ddot{\rho}_{101} + \sigma_{11} \ddot{\rho}_{100} + i[\rho_{00} \ddot{u}_{101} + \rho_{01} \ddot{u}_{100}] + [\rho_{00} \ddot{w}_{111} + \rho_{01} \ddot{w}_{110}]
\]
\[
+ [\rho_{00} \dddot{w}_{111} + \rho_{01} \dddot{w}_{110}] + [\ddot{\rho}_{110} w_{01z} + \dddot{\rho}_{111} w_{00z}] + [\ddot{\rho}_{110} w_{01z} + \dddot{\rho}_{111} w_{00z}] = 0
\]
\[
- i \dddot{\rho}_{101} + \eta \{ \dddot{u}_{111} + i \dddot{u}_{101} \} = 0
\]
\[
- \dddot{p}_{111} + \eta \{ 2 \dddot{w}_{111} + i \dddot{u}_{101} \} = 0
\]  \(\text{(B.31)}\)

At z=0:
\[
\ddot{\Delta}_{111} = 0
\]
\[
\ddot{u}_{111} = 0
\]
\[
\ddot{w}_{111} = 0.
\]  \(\text{(B.32)}\)

At z=h_0:
\[
\sigma_{11} \ddot{h}_1 = \ddot{w}_{111} - u_{01} \dddot{h}_1 + V \rho^* \left\{ \rho_{00} \ddot{\rho}_{111} - 2 \rho_{01} \ddot{\rho}_{110} \right\}
\]
\[
- i \dddot{h}_1 T_{01}^{11} + \eta \{ \dddot{u}_{111} + i \dddot{w}_{101} \} = 0
\]
\[
- \dddot{p}_{111} + 2 \eta \dddot{w}_{111} = 0
\]  \(\text{(B.33)}\)
At $O(k^2 \beta_1)$:

\[
\begin{align*}
\sigma_{10}\tilde{\rho}_{111} + \sigma_{11}\tilde{\rho}_{110} + [\sigma_{20}\tilde{\rho}_{101} + \sigma_{21}\tilde{\rho}_{100}] + i[\rho_{00}\tilde{u}_{111} + \rho_{01}\tilde{u}_{110}] \\
+ [\rho_{00}z\tilde{w}_{121} + \rho_{01}z\tilde{w}_{120}] + [\rho_{00}\tilde{w}_{121}' + \rho_{01}\tilde{w}_{120}'] + [\tilde{p}_{120}w_{01} + \tilde{p}_{121}w_{00}] + [\tilde{p}_{120}w_{01z} + \tilde{p}_{121}w_{00z}] = 0 \\
- i\tilde{p}_{111} + \eta\{-2\tilde{u}_{101} + \tilde{u}_{121}'' + i\tilde{w}_{111}'\} = 0 \\
- \tilde{p}_{121} + \eta\{-\tilde{w}_{101} + 2\tilde{w}_{121}'' + i\tilde{u}_{111}'\} = 0 \\
\sigma_{10}\tilde{\Delta}_{111} + \sigma_{11}\tilde{\Delta}_{110} + \sigma_{20}\tilde{\Delta}_{101} + \sigma_{21}\tilde{\Delta}_{100} + i[u_{00}\tilde{\Delta}_{111} + u_{01}\tilde{\Delta}_{110}] \\
+ w_{00}\tilde{\Delta}_{121}' + w_{01}\tilde{\Delta}_{120}' + \tilde{w}_{120}\Delta_{01z} + \tilde{w}_{121}\Delta_{00z} = 0 \\
\tilde{\rho}_{121} = -\rho^* \frac{\Delta_{00}\tilde{\Delta}_{121} + \tilde{\Delta}_{121} - 2\Delta_{01}\tilde{\Delta}_{120}}{(1 + \Delta_{00})^3}
\end{align*}
\]

(B.34)

At $z=0$:

\[
\tilde{\Delta}_{121} = 0 \\
\tilde{u}_{121} = 0
\]

(B.35)

At $z=h_0$:

\[
\sigma_{21}\tilde{h}_1 = \tilde{w}_{121} + V \rho^* \left( \frac{\rho_{00}\tilde{w}_{121} - 2\rho_{01}\tilde{w}_{120}}{\rho_{00}^3} \right) \\
\tilde{u}_{121}' + i\tilde{w}_{111} = 0
\]

(B.36)

\[
-\tilde{p}_{121} + 2\eta\tilde{w}_{121}' = 0
\]

B.1.2. Solution

Because of the lengthy calculations and the potential significance of the claims made in the present work, we include here the details of calculation leading to the theoretical steady-state stress and the dispersion relation.

**Steady state solutions at** $O(1)$. Observe that $w_{00}\Delta_{00z} = 0$ implies that either $w_{00} = 0$ or $\Delta_{00z} = 0$. If $w_{00} = 0$, we have no means of determining either $\Delta_{00}$ or $\rho_{00}$, but this solution appears physically unreasonable so we disregard it. On the other hand, $\Delta_{00z} = 0$ implies $\Delta_{00} = c_0$, and the first boundary condition at $z = 0$ implies $c_0 = 0$. Then $\rho_{00} = \rho^*$,
leading to \( w_{00z} = 0 \) in the steady-state mass conservation equation in the bulk. Then \( w_{00} \) is a constant, and the boundary conditions at both interfaces imply \( w_{00} = V \), the erosion rate.

Steady-state momentum conservation in the bulk implies that

\[
\begin{align*}
  u_{00z} &= 2fA_D D_{13} \tau_0 + c_1 
  \rightarrow u_{00} = 2fA_D D_{13} \int_0^z \tau_0 d\tilde{z} + c_1 z + c_2 \\
  -p_{00} &= 2\eta fA_D D_{33} \tau_0 + c_3.
\end{align*}
\]

(B.37)

Observe that the third boundary condition at the upper interface implies that \( c_3 = 0 \), and the second boundary conditions at both the upper and lower interfaces imply \( c_1 = c_2 = 0 \). Thus we have solution set

\[
\begin{align*}
  \Delta_{00} &= 0, \rho_{00} = \rho^*, \ w_{00} = V, \u0003 \\
  u_{00} &= 2fA_D D_{13} \int_0^z \tau_0 d\tilde{z}, p_{00} = -2\eta fA_D \tau_0 D_{33}.
\end{align*}
\]

(B.38)

**Steady state solutions at \( O(fA_I) \).** From the steady-state bulk equation for advection of volumization, including the knowledge that \( \Delta_{00} = 0 \) and \( w_{00} = V \) from the previous calculations (regardless of the spatial variation, which has been left general), we have

\[
\begin{align*}
  \Delta_{01z} &= \frac{1}{V} \alpha_{01} \rightarrow \Delta_{01} = \frac{1}{V} \int_0^z \alpha_{01} d\tilde{z} + c_4,
\end{align*}
\]

(B.39)

and the first boundary condition at \( z = 0 \) implies \( c_4 = 0 \). This leads immediately to the solution for \( \rho_{01} \),

\[
\rho_{01} = -\frac{\rho^*}{V} \int_0^z \alpha_{01} d\tilde{z}.
\]

(B.40)

Knowing \( \rho_{01} \) allows us to determine \( w_{01} \) from the steady state bulk mass conservation equation, and we obtain \( \rho^* w_{01} + \rho_{01} V = c_5 \), implying

\[
\begin{align*}
  w_{01} &= \int_0^z \alpha_{01} d\tilde{z} + c_5.
\end{align*}
\]

(B.41)

Consideration of the boundary conditions leads to \( c_5 = 0 \). Steady-state momentum conservation in the bulk leads to \( -p_{01} + 2\eta w_{01z} = 0 \), hence

\[
\rho_{01} = 2\eta \alpha_{01}.
\]

(B.42)
Finally, the other steady-state momentum conservation equation in the bulk leads to \(u_{01}\) being at most linear in \(z\), and the two homogeneous boundary conditions force \(u_{01} = 0\). We therefore have the solution set

\[
\Delta_{01} = \frac{1}{V} \int_0^z \alpha_{01} d\tilde{z} , \rho_{01} = \frac{-\rho^*}{V} \int_0^z \alpha_{01} d\tilde{z} ,
\]

\[
w_{01} = \int_0^z \alpha_{01} d\tilde{z} , u_{01} = 0 , p_{01} = 2\eta\alpha_{01}.
\]  

(B.43)

**General strategy.** Following the same pattern as above, the solution sets at all other orders may be obtained by checking the linearized “age-advection” equation, passing the solution for \(\Delta\) into the linearized equation of state to obtain \(\rho\), which may then be used to obtain \(w\) from the linearized conservation of mass. Pressure \(p\) is always obtained via integration of the \(z\)-component of linearized bulk momentum conservation equation after having determined \(w\), and \(u\) is always obtained by integration of the \(x\)-component of linearized bulk momentum conservation.

At \(\mathcal{O}(e^{1k^0}(fA_I)^0)\): We have

\[
w_{00} \tilde{\Delta}'_{100} = 0 \rightarrow \tilde{\Delta}_{100} = c_1
\]  

(B.44)

and boundary condition at \(z=0\), \(\Delta_{00z}\tilde{g}_1 + \tilde{\Delta}_{100} = 0\), implies \(\Delta_{100} = 0\), because \(\Delta_{00z} = 0\). Then we immediately have \(\rho_{100} = 0\). Mass conservation in the bulk yields

\[
\rho_{00} \tilde{w}'_{100} = 0 \rightarrow \tilde{w}_{100} = c_2
\]  

(B.45)

and the boundary condition \(\tilde{w}_{100} + w_{00z}\tilde{g}_1 = 0\) implies \(\tilde{w}_{100} = 0\), because \(w_{00z} = 0\). Then we have

\[
\tilde{p}_{100} = -2fA_D\eta D_{33}\tau_\epsilon + c_3
\]  

(B.46)

by integrating the \(z\)-component of bulk momentum conservation. We also have steady-state stress tensor to leading order in \(fA_I\), \(T_{00}\), whose bottom-right component is

\[
-p_{00} + 2\eta \{ w_{00z} - fA_D\tau_0(z;0,h_0)D_{33} \}.
\]  

(B.47)
With \( p_{00} = -2fA_D \eta \tau(z; 0, h_0)D_{33} \) and \( w_{00} = V \) from previous calculations, \( T_{00z}^{33} = 0 \). This combined with the boundary conditions implies \( c_3 = 0 \). Then integrating the \( x \)-component of bulk momentum conservation twice and satisfying the boundary conditions leads to

\[
\tilde{u}_{100} = 2fA_DD_{13} \int_0^z \tau_\epsilon d\tilde{z} - 2fA_DD_{13}\tau_0(0; 0, h_0)\tilde{g}_1. \tag{B.48}
\]

**At \( O(\epsilon^1 k^0 (fA_I)^1) \):** From linearized age-advection, we have

\[
\Delta_{101z} = \frac{1}{V} \alpha_{\epsilon_1}, \tag{B.49}
\]

and boundary condition at \( z = 0 \), \( \Delta_{01z}\tilde{g}_1 + \tilde{\Delta}_{101} = 0 \) implies

\[
\Delta_{101} = \frac{1}{V} \int_0^z \alpha_{\epsilon_1} d\tilde{z} - \alpha_{01}(0; 0, h_0)\frac{\tilde{g}_1}{V}. \tag{B.50}
\]

From the linearized equation of state, we have

\[
\tilde{\rho}_{101} = -\rho^* \tilde{\Delta}_{101}, \tag{B.51}
\]

leading immediately to

\[
\tilde{\rho}_{101} = -\frac{\rho^*}{V} \left( \int_0^z \alpha_{\epsilon_1} d\tilde{z} - \alpha_{01}(0; 0, h_0)\tilde{g}_1 \right). \tag{B.52}
\]

With the \( x \)-component of linearized bulk momentum conservation, we have that \( \tilde{u}_{101} \) is at most linear, and the homogeneous boundaries imply \( \tilde{u}_{101} = 0 \). Conservation of mass in the bulk leads to

\[
\rho_{00}\tilde{w}_{101z} + \tilde{\rho}_{101z}w_{00} = 0 \rightarrow \rho^*\tilde{w}_{101z} + V\tilde{\rho}_{101z} = 0, \tag{B.53}
\]

hence

\[
\tilde{w}_{101} = -\frac{\tilde{\rho}_{101}}{\rho^*} \frac{V}{c_2}, \tag{B.54}
\]

or

\[
\tilde{w}_{101} = \left( \int_0^z \alpha_{\epsilon_1} d\tilde{z} - \alpha_{01}(0; 0, h_0)\tilde{g}_1 \right) + c_2, \tag{B.55}
\]

and enforcement of the boundary condition at \( z=0 \)

\[
\tilde{w}_{101} + w_{01z}g_1 = 0 \rightarrow \tilde{w}_{101} + \alpha_{01}(z; 0, h_0)g_1 = 0 \tag{B.56}
\]
implies \( c_2 = 0 \). Finally, from the integrated z-component of bulk momentum conservation, we have

\[
\tilde{p}_{101} = 2\eta\tilde{w}_{101z} + c_3.
\]

The bottom-right component of the steady-state stress tensor at \( O(\alpha) \), \( T_{01}^{33} \), is

\[
-p_{01} + 2\eta w_{01z} = 0,
\]

which implies \( c_3 = 0 \), hence

\[
\tilde{p}_{101} = 2\eta\alpha_1.
\]

**At \( O(\epsilon^1 k^1 (f A_I)^0) \):** From the linearized age-advection equation, we have

\[
w_{00} \tilde{\Delta}_{110} = 0 \rightarrow \tilde{\Delta}_{110} = c_4,
\]

and the boundary conditions imply \( c_4 = 0 \), hence \( \tilde{\Delta}_{110} = 0 \). From the linearized equation of state, this implies \( \tilde{\rho}_{110} = 0 \), and linearized bulk mass conservation becomes

\[
i\rho^* \tilde{u}_{100} + \rho^* \tilde{w}_{110} = 0 \rightarrow \tilde{w}_{110}' = -i\tilde{u}_{100}.
\]

Integration leads to

\[
\tilde{w}_{110} = -2i f A_D D_{13} \left[ \int_0^z \int_{\tilde{z}^0}^{\tilde{z}} \tau_{i\tilde{z}d\tilde{z}} - \tau_0(0; 0, h_0) \tilde{g}_1 z \right] + c_5,
\]

and boundary condition at \( z = 0, \tilde{w}_{110} = 0 \) implies \( c_5 = 0 \). Then from the x-component of bulk momentum conservation, we have

\[
-i\tilde{p}_{100} + \eta \{ \tilde{u}_{110}'' + i\tilde{w}_{100}' \} = 2f A_D \eta i D_{11} \tau_\epsilon
\]

\[
\rightarrow \quad \tilde{u}_{110}'' = 2f A_D i(D_{11} - D_{33}) \tau_\epsilon.
\]

Integrating twice leads to

\[
\tilde{u}_{110} = 2f A_D i(D_{11} - D_{33}) \int_0^z \int_{\tilde{z}^0}^{\tilde{z}} \tau_{i\tilde{z}d\tilde{z}} + c_6 z + c_7,
\]
and we must enforce boundary conditions. At $z=0$, $\tilde{u}_{110} = 0$, implying $c_7 = 0$, and at $z = h_0$,

$$\eta \tilde{u}'_{110} + \eta i \tilde{w}'_{100} - i \tilde{h}_1 T_{00}^{11} = 0 \quad \rightarrow \quad (B.65)$$

$$\eta \left[ 2 f A_D i (D_{11} - D_{33}) \int_0^h \tau_\epsilon d\tilde{z} + c_6 \right] + 2 f A_D \eta i \tilde{h}_1 \tau_0(z; 0, h_0)(D_{11} - D_{33}) = 0,$$

by recognizing that

$$T_{00}^{11} = -p_{00} - 2 f A_D \eta \tau_0(z; 0, h_0) D_{11} = -2 \eta f A_D \tau_0(z; 0, h_0)(D_{11} - D_{33}). \quad (B.66)$$

Then we find that

$$c_6 = -2 f A_D i \tilde{h}_1 \tau_0(h_0; 0, h_0)(D_{11} - D_{33}) - 2 f A_D i (D_{11} - D_{33}) \int_0^{h_0} \tau_\epsilon d\tilde{z}, \quad (B.67)$$

which we will carry symbolically. Finally, we seek $\tilde{p}_{110}$. From the $z$-component of bulk momentum conservation, we have

$$-\tilde{p}'_{110} + \eta (2 \tilde{u}''_{110} + i \tilde{u}_{100}) = 2 f A_D \eta i D_{31} \tau_\epsilon$$

$$\rightarrow \quad \rightarrow \quad -\tilde{p}'_{110} = 2 f A_D \eta i (D_{31} + D_{13}) \tau_\epsilon. \quad (B.68)$$

One integration leads to

$$\tilde{p}_{110} = -2 f A_D \eta i (D_{31} + D_{13}) \int_0^h \tau_\epsilon d\tilde{z} + c_7. \quad (B.69)$$

From the boundary condition at $z = h_0$,

$$-\tilde{p}_{110} + 2 \eta \tilde{w}'_{110} = 0, \quad (B.70)$$

hence

$$c_7 = 2 f A_D \eta i (D_{31} - D_{13}) \int_0^{h_0} \tau_\epsilon d\tilde{z} + 4 f A_D \eta i D_{13} \tau_0(0; 0, h_0) \tilde{g}_1, \quad (B.71)$$

which we will carry symbolically. Last, we compute $\sigma_{10}$ from the compatibility condition due to the linearized kinematic condition at the upper interface,

$$\sigma_{10} \tilde{h}_1 = \tilde{w}_{110} - u_{00} \tilde{h}_1 + \frac{V \rho^*}{\rho_{00}^*} \tilde{p}_{110}, \quad (B.72)$$

which leads to

$$\sigma_{10} = -\frac{2 f A_D i D_{13}}{\tilde{h}_1} \left[ \int_0^{h_0} \int_0^h \tau_\epsilon d\tilde{z} - \tau_0(0; 0, h_0) \tilde{g}_1 h_0 \right] - 2 f A_D i D_{13} \int_0^{h_0} \tau_0 d\tilde{z}. \quad (B.73)$$

Note the appearance of the interface relation term, $\tilde{g}_1$. 178
At $O(\epsilon^1 k^1 (f A_f)^1)$: Looking at the age-advection equation and simplifying, we obtain

$$\sigma_{10} \Delta_{101} + i u_{00} \Delta_{101} + w_{00} \Delta'_{111} + \tilde{w}_{110} \Delta_{01z} = 0.$$  \hspace{1cm} (B.74)

Note the appearance of the interaction term $u_{00} \Delta_{101}$, which involves the spatial variation functions of both anisotropic plastic flow and isotropic swelling. This leads to

$$\tilde{\Delta}_{111} = - \frac{1}{V} \int_0^z \left[ \sigma_{10} \Delta_{101} + i u_{00} \Delta_{101} + \tilde{w}_{110} \frac{\alpha_{01}}{V} \right] \Big|_{z=z} dz + c_8,$$  \hspace{1cm} (B.75)

and the boundary condition $\tilde{\Delta}_{111} = 0$ at $z = 0$ implies that $c_8 = 0$. The linearized equation of state immediately reduces to

$$\tilde{\rho} = - \rho^* \tilde{\Delta}_{111}.$$  \hspace{1cm} (B.76)

Bulk mass conservation leads to

$$\sigma_{10} \tilde{\rho}_{101} + i \rho_{01} \tilde{u}_{100} + \rho_{01z} \tilde{w}_{110} + \rho_{00} \tilde{w}'_{111} + \rho_{01} \tilde{w}'_{110} + \tilde{\rho}'_{111} u_{00} = 0$$  \hspace{1cm} (B.77)

after simplification. Then we have

$$\tilde{w}'_{111} = \left( \frac{1}{V} \int_0^z \alpha_{01} d\tilde{z} \right) \tilde{w}'_{110} + \frac{\alpha_{01}}{V} \tilde{w}_{110} + \left( \frac{i}{V} \int_0^z \alpha_{01} d\tilde{z} \right) \tilde{u}_{100} - \sigma_{10} \frac{\tilde{\rho}_{101}}{\rho^*} - \frac{V}{\rho^*} \tilde{\rho}'_{111},$$  \hspace{1cm} (B.78)

hence

$$\tilde{w}_{111} = \int_0^z \left[ \left( \frac{1}{V} \int_0^z \alpha_{01} d\tilde{z} \right) \tilde{w}'_{110} + \frac{\alpha_{01}}{V} \tilde{w}_{110} + \left( \frac{i}{V} \int_0^z \alpha_{01} d\tilde{z} \right) \tilde{u}_{100} - \sigma_{10} \frac{\tilde{\rho}_{101}}{\rho^*} - \frac{V}{\rho^*} \tilde{\rho}'_{111} \right] \Big|_{z=z} dz + c_9,$$  \hspace{1cm} (B.79)

and the boundary condition $\tilde{w}_{111} = 0$ at $z = 0$ implies $c_9 = 0$. From the z-component of bulk momentum conservation, the knowledge that $\tilde{u}_{101} = 0$, and one integration, we have

$$\tilde{p}_{111} = 2 \eta \tilde{w}'_{111} + c_{10},$$  \hspace{1cm} (B.80)

and the boundary condition at $z = h_0$ implies $c_{10} = 0$. We now seek $\tilde{u}_{111}$ by looking at the x-component of bulk momentum conservation,

$$-i \tilde{p}_{101} + \eta \{ \tilde{u}_{111}'' + i \tilde{w}'_{101} \} = 0$$  \hspace{1cm} \rightarrow

$$\tilde{u}_{111}'' = \frac{i}{\eta} \tilde{p}_{101} - i \tilde{w}'_{101} = i \alpha_{e_1},$$  \hspace{1cm} (B.81)
where the final equality follows from simplification. Hence

\[
\tilde{u}_{111} = \int_0^z \int_0^{\tilde{z}} i\alpha_1 d\tilde{z} d\tilde{z} + c_{11} z + c_{12}.
\]  
(B.82)

From the boundary condition \(\tilde{u}_{111} = 0\) at \(z = 0\), \(c_{12} = 0\). From the boundary condition at \(z = h_0\),

\[
i\tilde{h}_1 p_{01} + \eta [\tilde{u}_{111}' + i\tilde{w}_{101}] = 0,
\]  
(B.83)

we arrive at

\[
c_{11} = i \left[ \alpha_{01}(0; 0, h_0) \tilde{g}_1 - 2\alpha_{01}(h_0; 0, h_0) \tilde{h}_1 - 2 \int_0^{h_0} \alpha_1 d\tilde{z} \right],
\]  
(B.84)

which we will carry symbolically. Finally, from the linearized kinematic condition and simplification, we obtain

\[
\sigma_{11} = \frac{1}{h_1} \left( \tilde{w}_{111} - V\tilde{\Delta}_{111} \right) |_{z = h_0}.
\]  
(B.85)

At \(O(\epsilon^1 k^2 (f A_t)^0)\): From the linearized age-advection equation and the boundary condition at \(z = 0\), we find that \(\tilde{\Delta}_{120} = 0\), which immediately implies that \(\tilde{\rho}_{120} = 0\). Linearized bulk mass conservation and the boundary condition at \(z = 0\) for \(\tilde{w}_{120}\) then imply

\[
\tilde{w}_{120} = -i \int_0^z \tilde{u}_{110} d\tilde{z}.
\]  
(B.86)

From the \(z\)-component of bulk momentum conservation and simplification, we have

\[
-\tilde{p}_{120}' + \eta \{2\tilde{u}_{120}' + i\tilde{u}_{110}'\} = 0
\]

\[
\rightarrow
\]

\[
\tilde{p}_{120}' = -i\eta \tilde{u}_{110}'
\]  
(B.87)

\[
\rightarrow
\]

\[
\tilde{p}_{120} = -i\eta \tilde{u}_{110} + c_{13}.
\]

From the boundary condition at \(z = h_0\), \(-\tilde{p}_{120} + 2\eta \tilde{w}_{120} = 0\), we find that \(c_{13} = -i\eta \tilde{u}_{110} |_{z = h_0}\),

\[
\tilde{p}_{120} = -i\eta [\tilde{u}_{110} + \tilde{u}_{110} |_{z = h_0}].
\]  
(B.88)
Now we seek \( \tilde{u}_{120} \). From the x-component of bulk momentum conservation, we have
\[
-i\tilde{p}_{110} + \eta\{-2\tilde{u}_{100} + \tilde{u}''_{120} + i\tilde{w}'_{110}\} = 0
\]
\[
\rightarrow \quad \tilde{u}_{120} = \int_0^z \int_0^z \left[ \frac{i}{\eta} \tilde{p}_{110} + 2\tilde{u}_{100} \right] d\tilde{z} d\tilde{z} - i \int_0^z \tilde{w}_{110} d\tilde{z} + c_{14} z + c_{15}.
\]
From the boundary condition \( \tilde{u}_{120} = 0 \) at \( z = 0 \), \( c_{15} = 0 \). From the boundary condition \( \tilde{u}'_{120} + i\tilde{w}_{110} = 0 \) at \( z = h_0 \), we have
\[
c_{14} = -\int_0^{h_0} \left[ \frac{i}{\eta} \tilde{p}_{110} + 2\tilde{u}_{100} \right] d\tilde{z},
\]
which will be carried symbolically. Finally, using the linearized kinematic condition and the knowledge that \( \tilde{\rho}_{120} = 0 \), at \( z = h_0 \) we find
\[
\sigma_{20} = \frac{\tilde{w}_{120}}{h_1} = -\frac{i}{h_1} \int_0^{h_0} \tilde{u}_{110} d\tilde{z}.
\]

At \( O(\epsilon^1 k^2(fA_s)^1) \): We note that at this order, we require only \( \tilde{w}_{121} \) and \( \tilde{\rho}_{121} \) in order to determine \( \sigma_{21} \) and complete our analysis. From the linearized age-advection equation with simplifications, we have
\[
\sigma_{10} \tilde{\Delta}_{111} + \sigma_{20} \tilde{\Delta}_{101} + w_{00} \tilde{\Delta}'_{121} + \tilde{w}_{120} \tilde{\Delta}_{01z} + iu_{00} \tilde{\Delta}_{111} = 0.
\]
Isolating \( \tilde{\Delta}'_{121} \), integrating once and applying the boundary condition at \( z = 0 \) implies
\[
\tilde{\Delta}_{121} = -\frac{1}{V} \int_0^z \left[ \sigma_{10} \tilde{\Delta}_{111} + \sigma_{20} \tilde{\Delta}_{101} + \tilde{w}_{120} \tilde{\Delta}_{01z} + iu_{00} \tilde{\Delta}_{111} \right] d\tilde{z}.
\]
From the linearized equation of state and the knowledge that \( \Delta_{00} = 0 \) and \( \tilde{\Delta}_{120} = 0 \), we immediately find
\[
\tilde{\rho}_{121} = -\rho^* \tilde{\Delta}_{121}.
\]
Now we seek \( \tilde{w}_{121} \) using the linearized bulk mass conservation equation. We find
\[
\sigma_{10} \tilde{\rho}_{111} + \sigma_{20} \tilde{\rho}_{101} + i [\rho_{00} \tilde{u}_{111} + \rho_{01} \tilde{u}_{110}] + \rho_{01z} \tilde{w}_{120} + \rho_{00} \tilde{w}_{121} + \rho_{01} \tilde{w}'_{120} + \rho_{121} w_{00} = 0.
\]
Rearrangement, integration and enforcement of the boundary condition for \( \tilde{w}_{121} \) at \( z = 0 \) implies

\[
\tilde{w}_{121} = -\frac{1}{\rho_{00}} \int_{0}^{z} \left[ \sigma_{10} \tilde{\rho}_{111} + \sigma_{20} \tilde{\rho}_{101} + i \rho_{00} \tilde{u}_{111} + i \rho_{01} \tilde{u}_{110} + \rho_{01} \tilde{w}_{120} + \rho_{01} \tilde{w}'_{120} + \tilde{\rho}'_{121} w_{00} \right] d\tilde{z}.
\]

(B.96)

Finally, we use the linearized kinematic condition at \( z = h_0 \) to determine \( \sigma_{21} \) and complete the analysis. With \( \tilde{\rho}_{120} = 0 \), we have

\[
\sigma_{21} = \frac{1}{h_1} \left( \tilde{w}_{121} - V \tilde{\Delta}_{121} \right) \bigg|_{z = h_0}.
\]

(B.97)

**Construction of general dispersion relation: general form.** Recall that

\[
\sigma \approx 0 + k \left( \sigma_{10} + f A_I \sigma_{11} \right) + k^2 \left( \sigma_{20} + f A_I \sigma_{21} \right).
\]

(B.98)

Collecting all terms, we find

\[
\sigma_{10} = -\frac{2 f A_D i D_{13}}{h_1} \left[ \int_{0}^{h_0} \int_{0}^{z_1} \tau_\epsilon(z_2) d z_2 d z_1 - \tau_0(0) \tilde{g}_1 h_0 + \tilde{h}_1 \int_{0}^{h_0} \tau_0(z) d z \right];
\]

(B.99)

\[
\sigma_{11} = \frac{1}{V h_1} \int_{0}^{h_0} \left[ -2 f A_D i D_{13} \alpha_{01}(z) \left[ \int_{0}^{z} \int_{0}^{z_1} \tau_\epsilon(z_2) d z_2 d z_1 - \tau_0(0) \tilde{g}_1 z \right] + \sigma_{10} \left[ \int_{0}^{z} \alpha_{\epsilon 1}(z_1) d z_1 - \alpha_{01}(0) \tilde{g}_1 \right] \right] d z;
\]

(B.100)

\[
\sigma_{20} = \frac{2 f A_D (D_{11} - D_{33})}{h_1} \int_{0}^{h_0} \left[ \int_{0}^{z} \int_{0}^{z_1} \tau_\epsilon(z_2) d z_2 d z_1 - z \left[ \tau_0(h_0) \tilde{h}_1 + \int_{0}^{h_0} \tau_\epsilon(z) d z \right] \right] d z;
\]

(B.101)

and
Linearization of the spatial variation functions. In the course of the linear stability analysis, we are naturally required to perform a linearization of the spatial-variation associated with each bulk mechanism. In the present work, we will derive these profiles from angle-dependent spatial variation functions associated with the generation of each respective bulk mechanism. In the spirit of generality, we carry out our linear stability analysis for arbitrary functions, which forms a reusable framework for later analyses. We will therefore consider expansions

\[ \tau(z; ...) = \tau_0(z; ...) + \epsilon \tau_\epsilon(z; ...) + O(\epsilon^2) \]

\[ \alpha(z; ...) = \alpha_0(z; ...) + \epsilon \alpha_\epsilon(z; ...) + O(\epsilon^2) \]

in anticipation of these forms arising within the linear stability analysis. Here, the (...) denotes any other quantities carried within the linearization. For example, linearizing as \( g = 0 + \epsilon g_1 \) and \( h = h_0 + \epsilon h_1 \) leads to

\[ \tau(z; g, h) = \tau(z; 0, h_0) + \epsilon \left( \frac{\partial \tau}{\partial h} h_1 + \frac{\partial \tau}{\partial g} g_1 \right) \bigg|_{h=h_0, g=0} \]

hence

\[ \tau_0(z) = \tau(z; 0, h_0); \quad \tau_\epsilon(z) = \left( \frac{\partial \tau}{\partial h} h_1 + \frac{\partial \tau}{\partial g} g_1 \right) \bigg|_{h=h_0, g=0}, \]

and the same convention will be adopted for \( \alpha_0(z) \) and \( \alpha_\epsilon(z) \). In the Appendix, we discuss the necessity of an additional expansion of \( \alpha \) in the limit of small swelling rate \( fA_I \) relative
to dwell-time, which will lead to

\[
\alpha_0 \rightarrow \alpha_{00} + fA_I\alpha_{01},
\]

\[
\alpha_\varepsilon \rightarrow \alpha_{\varepsilon 0} + fA_I\alpha_{\varepsilon 1},
\]

(B.106)

with \(\alpha_{00} \rightarrow 0, \alpha_{\varepsilon 0} \rightarrow 0\) in the small-\(fA_I\) limit.

**B.2. Nondimensionalization and small cross-terms limit**

In the main text, we have claimed that a nondimensionalization in the limit of small “dwell-times” for each stressed parcel of matter in the amorphous bulk produces a simplification which is mathematically convenient while still physically relevant for the 250eV Ar\(^+\) \(\rightarrow\) Si system. To show this, consider the nondimensionalization of all length scales by \(\hat{z} = \frac{z}{h_0}\) and all time scales by \(\hat{t} = \frac{V}{h_0}t\). This leads to

\[
\Sigma = \frac{\sigma h_0}{V} \approx 0 + (kh_0) \left( \frac{\sigma_{10}}{V} + \frac{fA_I\sigma_{11}}{V} \right) + (kh_0)^2 \left( \frac{\sigma_{20}}{h_0V} + \frac{fA_I\sigma_{21}}{h_0V} \right),
\]

(B.107)

where \(\sigma\) is the linear dispersion relation, \(k\) is the wavenumber, and \(fA_I\) is the baseline magnitude of the isotropic swelling mechanism across the film. The analysis above leads to

\[
\frac{\sigma_{10}}{V} = -\left( \frac{fA_Dh_0}{V} \right) \frac{2iD_{13}}{h_1} \int_0^1 \int_0^{\hat{z}_1} \tau_\varepsilon(\hat{z}_2)d\hat{z}_2d\hat{z}_1 - \tau_0(0)\hat{g}_1 + \hat{h}_1 \int_0^1 \tau_0(\hat{z})d\hat{z};
\]

(B.108)

\[
fA_I\sigma_{11} = \left( \frac{\hat{\alpha}h_0}{V} \right) \frac{1}{h_1} \int_0^1 \left[ -2\left( \frac{fA_Dh_0}{V} \right) iD_{13} \alpha_{01}(\hat{z}) \left[ \int_0^{\hat{z}} \int_0^{\hat{z}_1} \tau_\varepsilon(\hat{z}_2)d\hat{z}_2d\hat{z}_1 - \tau_0(0)\hat{g}_1 \hat{z} \right] 
+ \left( \frac{\sigma_{10}}{V} \right) \left[ \int_0^{\hat{z}} \alpha_{\varepsilon 1}(\hat{z}_1)d\hat{z}_1 - \alpha_{01}(0)\hat{g}_1 \right] \right]d\hat{z};
\]

(B.109)

\[
\frac{\sigma_{20}}{h_0V} = \frac{2(D_{11} - D_{33})}{h_1} \left( \frac{fA_Dh_0}{V} \right) \int_0^1 \left[ \int_0^{\hat{z}} \int_0^{\hat{z}_1} \tau_\varepsilon(\hat{z}_2)d\hat{z}_2d\hat{z}_1 - \hat{z} \left[ \tau_0(1)\hat{h}_1 + \int_0^1 \tau_\varepsilon(\hat{z})d\hat{z} \right] \right]d\hat{z};
\]

(B.110)

and
\[ \frac{f_{\text{A} \sigma_{21}}}{h_0 V} = - \left( \frac{f_{\text{A} h_0}}{V} \right) \frac{1}{h_1} \int_0^1 \left[ \frac{\sigma_{10}}{V} \int_0^{\tilde{z}_1} \alpha_{e1}(\tilde{z}_2) d\tilde{z}_2 - \alpha_{01}(0) \tilde{g}_1 \right] \left( \frac{\sigma_{10}}{V} \right) \right. \\
+ 2 \left( \frac{f_{\text{A} h_0}}{V} \right) i D_{13} \int_0^{\tilde{z}_1} \tau_0(\tilde{z}_2) d\tilde{z}_2 \right) \\
- 2 \left( \frac{f_{\text{A} h_0}}{V} \right) i D_{13} \alpha_{01}(\tilde{z}_1) \left( \int_0^{\tilde{z}_1} \int_0^{\tilde{z}_2} \tau_3(\tilde{z}_3) d\tilde{z}_2 - \tau_0(0) \tilde{g}_1 \tilde{z}_1 \right) \right] d\tilde{z}_1 \\
- \frac{\sigma_{20}}{h_0 V} \left[ \int_0^{\tilde{z}_1} \alpha_{e1}(\tilde{z}_1) d\tilde{z}_1 - \alpha_{01}(0) \tilde{g}_1 \right] \\
- \left[ \int_0^{\tilde{z}_1} \int_0^{\tilde{z}_2} \alpha_{e1}(\tilde{z}_2) d\tilde{z}_2 d\tilde{z}_1 + \tilde{z} \left( \alpha_{01}(0) \tilde{g}_1 - 2 \alpha_{01}(1) \tilde{h}_1 - 2 \int_0^{\tilde{z}_1} \alpha_{e1}(\tilde{z}) d\tilde{z} \right) \right] \\
- \left( \frac{f_{\text{A} h_0}}{V} \right) 2 \alpha_{01}(\tilde{z}) (D_{11} - D_{33}) \left[ \int_0^{\tilde{z}} \left( \int_0^{\tilde{z}_1} \int_0^{\tilde{z}_2} \tau_3(\tilde{z}_3) d\tilde{z}_2 d\tilde{z}_1 \right. \right. \\
\left. \left. \left. - \tilde{z}_1 \tau_0(1) \tilde{h}_1 + \int_0^{\tilde{z}_1} \tau_3(\tilde{z}) d\tilde{z} \right) d\tilde{z}_1 \right] \right] d\tilde{z}.
\] (B.111)

From the above, it is clear that \( \sigma_{10} \propto \left( \frac{f_{\text{A} h_0}}{V} \right) \) and \( \sigma_{20} \propto \left( \frac{f_{\text{A} h_0}}{V} \right) \), while \( \frac{f_{\text{A} \sigma_{11}}}{h_0 V} \) and \( \frac{f_{\text{A} \sigma_{21}}}{h_0 V} \) have several cross-terms each. The elimination of these cross-terms and reinstating dimensional quantities leads to the “small cross-terms” result cited in the main text.

**B.3. Legendre polynomials in downbeam-crossbeam coordinates**

![Diagram](image.png)

Figure B.1. Schematic depicting the construction of depth-dependence at a point \( S \) in the film depth in Cartesian \( x, z \) coordinates by mapping the influence of an ion that enters the surface at the point \( Q \) to the point \( S \). In the limit of small slopes, this geometric argument leads to analytically-tractable forms.
Choose an arbitrary patch of surface \((X, h(X))\) on a surface \((x, h(x))\), identified with point \(P\) in Figure 2 and examine some distance \(z\) below its surface, \((X, h(X) - z)\), identified with \(S\). Given an irradiation angle \(\theta\) (from the left) and under the assumption of no deflection of that angle within the film, the point \((X, h(X) - z)\) ("\(S\)") is associated with precisely one location on a nearby patch of surface from which the deposition occurring at \((X, h(X) - z)\) originates. Denote this associated patch of surface as the point \((x_1, h(x_1))\), identified with the point \(Q\), the “entry point” at which an ion passes through the surface before initiating a collision cascade. Construct a triangle using the points \((X, h(X) - z)\) (\(S\)), \((x_1, h(x_1))\) (\(Q\)) and an additional point determined by the intersection of the vertical line running through \(x_1\) and the horizontal line \(h(X) - z\) (\(R\)), i.e., the triangle \(\triangle RQS\), whose interior angle is the same as that of the angle of the incoming ion to the laboratory frame’s \(z\) axis. The lengths of this triangle can be easily determined exactly via right-angle trigonometry. With the Taylor expansion of the surface about \(x = X\), we may approximate near \(x = x_1\). and with \(x_1 = X - x_2\) by definition, we have

\[
h(x_1) = h(X - x_2) = h(X) - x_2 \frac{\partial h}{\partial x} + \ldots, \tag{B.112}
\]

which implies that line segment \(QR\) is approximately of length \((h(X) - z) - x_2 \frac{\partial h}{\partial x}\). We now seek the horizontal distance from the entry point \((x_1, h(x_1))\) to the point of interest, \((X, h(X) - z)\), the line segment \(RS\). This horizontal shift, \(x_2\), whose length is identified with the line segment \(RS\) is precisely the length of the right triangle opposite the interior angle \(\theta\) and is immediately given by

\[
x_2 = \frac{\tan(\theta)(h(X) - z)}{1 + \tan(\theta) \frac{\partial h}{\partial x}} \tag{B.113}
\]

Finally, the hypotenuse (the line segment \(QS\), equivalent to the downbeam distance, which we will denote \(\tilde{z}\), has the form

\[
\tilde{z} = \sec(\theta) \left( h(X) - z - x_2 \frac{\partial h}{\partial x} \right) \bigg|_X \tag{B.114}
\]

which can be used to compute the influence of an arbitrary Legendre polynomial defined in downbeam coordinates. That is, we may express the spatial variation along the line
containing $P$ and $S$ in terms of a down-beam stress profile originally defined along the line containing $Q$ and $S$. For a given triplet of surface $h(X)$, ion-angle $\theta$ and point in the bulk $S$, there is exactly one patch of surface (the point $Q$) which determines the influence of the ion beam at the point $S$, hence the transformation is invertible and all depth-dependence throughout the entire film is uniquely assigned at each arbitrary point $S$ of the bulk. From the above argument, we find that in the limit of small slopes, the entire transformation is expressible in a manner suited to the present linear stability analysis. This construction is applicable to any arbitrary $(X, h(X))$; we now drop the dummy variable and consider

$$\tau(\hat{z}) = \sum_{k=0}^{N} \beta_k P_k(\hat{z}),$$

(B.115)

where $P_k(\hat{z})$ are the typical Legendre polynomials defined over $[-1,1]$. We may then transform to an arbitrary setting via

$$\hat{z} \rightarrow \frac{2\tilde{z} - (\hat{g} + \hat{h})}{\hat{h} - \hat{g}},$$

(B.116)

so that $\hat{g}$ is mapped to $-1$, and $\hat{h}$ is mapped to 1. We now set $\hat{g} = 0$ and $\hat{h} = h_0'$, the fixed downbeam distance as seen from downbeam coordinates. Hence

$$\tau(z) = \sum_{k=0}^{N} \beta_k P_k\left(\frac{2\tilde{z} - h_0'}{h_0'}\right),$$

(B.117)

where

$$\tilde{z} = \frac{(h - z)}{\cos(\theta) + h_x \sin(\theta)}$$

(B.118)

expresses the influence of an arbitrary Legendre polynomial, originally defined in downbeam coordinates, on an arbitrary location (in terms of the Cartesian $z$ axis, as opposed to the downbeam $\tilde{z}$) while the small-slopes approximation holds. For the reader’s convenience, we note that the typical $n^{th}$ Legendre polynomial $P_n(x)$ comprises an orthogonal family on the interval $[-1,1]$, and is easily computed by the Rodrigues formula,

$$P_n(x) = \frac{1}{2^n n! \ dx^n} \left[(x^2 - 1)^n\right].$$

(B.119)
The first three such polynomials are therefore

\begin{align*}
P_0(x) &= 1, \\
P_1(x) &= x, \\
P_2(x) &= \frac{1}{2} (3x^2 - 1).
\end{align*}

(B.120)

B.4. Previous depth-dependence considerations

The present work naturally evokes comparison to a previous discussion of depth-dependent stress due to [33]. However, several substantial differences exist. First, the fundamental form of the stress tensor, even before depth-dependence considerations, is different, and differently motivated. The present work bases its analysis on the phenomenological use of a stress tensor found in the nuclear stopping regime work of [37, 38, 39, 41, 59, 32], which draws analogy with the “pancake strain” model of anisotropic stress in irradiated films at higher, electronic stopping regime energies. On the other hand, that of [33] could be considered as an alternative phenomenological model seeking analogy with classical hydrodynamic instabilities [54], such as Bénard-Marangoni convection. (If anything, the existence of two such mathematically valid phenomenological models giving different predictions for the same physical system is an argument in favor of leveraging phenomenological models only to the extent necessary to develop true physically-grounded models—and then abandoning the phenomenological models.) It is therefore immediately unsurprising that we have obtained different results. Additionally, the present model allows an additional stabilizing mechanism via isotropic swelling, whereas that of most other hydrodynamic analyses of irradiated thin films consider only anisotropic stress (although not necessarily APF) due to the ion beam. Finally, we note that a simple way to determine which model (or family of models) is truer to the underlying physics would be to determine whether a Si wafer’s in-plane stress changes sign for some irradiation angle: the APF model is capable of producing such a change, while that of [33] is not. While the data of [47] appears to reflect such a sign-change, later experimental results strongly contraindicating this phenomenon would lead to a re-evaluation of the present theoretical work. In this part of the Appendix, we take a moment to detail some technical
differences between the present work and that of [33].

**Physical motivation and stress tensor form.** In both our work and that of [33], the influence of the ion-beam is treated as a viscous stress tensor with an extra term of the general form

\[
T = -pI + \eta \left( \nabla \vec{v} + (\nabla \vec{v})^T \right) + T_{\text{beam}} \tag{B.121}
\]

However, the specific forms of that irradiation term are meaningfully different between our respective analyses. In the present work, restricted to the \(x-z\) plane for the sake of comparison, we have

\[
T_{\text{beam, PW}} = -2fA_D \eta \tau(z; g, h) \times \begin{bmatrix}
\frac{3}{2} \cos(2\theta) - \frac{1}{2} & \frac{3}{2} \sin(2\theta) \\
\frac{3}{2} \sin(2\theta) & -\frac{3}{2} \cos(2\theta) - \frac{1}{2}
\end{bmatrix}, \tag{B.122}
\]

which has a fundamentally different physical meaning than that of [33]. In their work

\[
T_{\text{beam, MB}} = \tau_{zz}(h) \begin{bmatrix}
\cos(2\theta) & \sin(2\theta) \\
\sin(2\theta) & -\cos(2\theta)
\end{bmatrix}. \tag{B.123}
\]

The differences are subtle but impactful. First, \(T_{\text{beam, MB}}\) modifies the amorphous layer by contributing stress, and the resulting flow fields are a response to this contribution, while \(T_{\text{beam, PW}}\) is precisely the opposite: it deducts stress, permitting more strain per unit than would otherwise be possible. \(T_{\text{beam, PW}}\) is motivated by previous work on the “melt cycle” phenomenon in the electronic stopping regime, recently shown to lead to good experimental predictions even in the nuclear stopping regime, while \(T_{\text{beam, MB}}\) appeals to analogies with classical hydrodynamic stability, seeking essentially to motivate a modification of the viscous flow comparable to that of the well-known case of flow down an inclined plane, hence their terminology of an “effective body force”.

**Experimental comparison; wafer-curvature measurements.** The difference is not restricted to interpretation or motivation. We briefly consider differences in experimental predictions. The work of [47] has presented a first-of-its-kind angle-dependent measurement of ion-induced stresses, separating out the isotropic parts from the anisotropic parts. This
permits an immediate and revealing comparison of our respective models. The analysis of [33] results in an in-plane component of their steady-state stress tensor,

\[ T_{0,xx} = -p_0(z) + \tau_{zz}(0) \cos(2\theta), \]  

(B.124)

where

\[ p_0(z) = -\frac{\Delta \tau_{zz}}{R_0} z \cos^2(\theta) + \tau_{zz}(0) \cos(2\theta). \]  

(B.125)

As we have done in the present work, we may simply average across the film depth in order to compute \(< T_{0,xx} >\), which can be compared with wafer-curvature measurements of Perkinson [47]. This leads to

\[ < \tau_{0,xx} > = -\frac{1}{2} \Delta \tau_{zz} \cos^2(\theta) \]  

(B.126)

in the analysis of [33], compared with

\[ < T_{0}^{11} > = -6 f A_D \eta \cos(2\theta) - 2 f A_I \eta. \]  

(B.127)

in our own analysis. Revealingly, our stress tensor permits a change of sign for the in-plane steady-state stress component at some irradiation angle, while that of [33] does not. The work of Perkinson [47] clearly shows such a transition of in-plane stress behavior around 50 degrees, consistent with our stress tensor even in the absence of isotropic swelling, while that of [33] is incapable of making such a prediction unless \(\Delta \tau_{zz}\) itself is allowed to change signs with angle— which would be highly unexpected in an ion-irradiated system, as the energy deposited in the rotating Gaussian ellipsoid model of Sigmund is generally a fair approximation of true energy deposition [7, 8, 113, 114].

Construction of depth-dependence: down-beam versus laboratory coordinates

In [33], the depth-dependence about laboratory-coordinate \(z\) is constructed by a finite difference approximation of the change in stress about the downbeam coordinate, \(\partial_{z'} \tau_{zz} \approx \frac{\tau(h)-\tau(h_{ac})}{d_{z'}}\). However, in our opinion, coordinates in this construction seem to have been mixed: \(h\) and \(h_{ac}\) are treated as if they were in laboratory coordinates \((x, z)\) (indeed, they are later linearized), while \(d_{z'}\) is a true down-beam distance in the downbeam coordinate, a
fixed quantity. If this is a finite difference approximation in the downbeam direction and the film is subject to the assumption that the ion-track deposits its power directly along the downbeam axis, leading to amorphization up to some lower interface, then the difference between the relevant quantities $\tau(h)$ and $\tau(h_{ac})$ along the downbeam direction (which is actually of fixed length), is also fixed. This is because the symbols $h$ and $h_{ac}$ are fixed positions along the downbeam direction. As $h_{ac}$ is “pinned” to $h$ due to the connection between the two via the downbeam axis, we would not expect them to be subject to linearization later in the analysis: the difference between them is fully characterized by the assumption of diagonally-translated interfaces and exact. The downbeam film thickness $d_z$ is likewise a fixed value and it is seemingly unnecessary to compute it in terms of local incidence angles. This renders the expansion $\cos(\theta - \gamma) \approx \cos(\theta) + h_x \sin(\theta)$ unnecessary in equations (8)-(13) of [33].

If $\tau(h) - \tau(h_{ac})$ is taken as a fixed quantity, no linearization of these terms is necessary; likewise, $\cos(\theta - \gamma) \approx \cos(\theta) + h_x \sin(\theta)$ is also unnecessary. Then equations (A15)-(A19), the equations of mass and momentum conservation in the bulk and the stress balance at the upper interface, are substantially modified. Specifically, all terms with $h_1$ due to the previously-discussed linearizations are set to zero. We note that the $h_1$ in (A20) still appears: it is due to the linearization of the lower interface, not due to the linearization of the stress tensor. However, with these $h_1$ terms gone from the bulk and stress balance at the upper interface, the term $\Delta \tau_{zz}$ completely disappears from the real part of the linear dispersion relation (although not from the imaginary part; an influence due to the lower interface is retained there, which is carried from the solution of the horizontal flow field at the lower interface). With these modifications, the real part of the linear dispersion relation is

$$\text{Re}(\omega_q) = -\frac{48\tau_{zz}(0) R_0^2}{9\mu \sin^2(\theta)} \left( \sin^2(2\theta) + \cos(2\theta) \right),$$

(B.128)

which predicts $\theta_c \approx 65^\circ$ independent of spatial variation. The uniformity of this prediction is consistent with our own in the case of fixed downbeam film thickness, where we have assumed diagonally-translated interfaces as well.
C.1. Details of linear stability analysis

C.1.1. General equations

Here, we also note the general momentum balance equations in the bulk for the reader’s reference:

\[-p_x + \eta\{2u_{xx} + u_{yy} + v_{xy} + u_{zz} + w_{xz}\} = 2fA_D\eta\{D_{11}\frac{d\tau}{dx} + D_{13}\frac{d\tau}{dz}\}\]

\[-p_y + \eta\{u_{xy} + v_{xx} + 2v_{yy} + v_{zz} + w_{yz}\} = 2fA_D\eta\frac{d\tau}{dy}\]  \hspace{1cm} (C.1)

\[-p_z + \eta\{u_{xz} + v_{yz} + w_{xx} + w_{yy} + 2w_{zz}\} = 2fA_D\eta\{D_{31}\frac{d\tau}{dx} + D_{33}\frac{d\tau}{dz}\}\]

We note that we have not assumed standard incompressibility in order to simplify the above momentum balance equations (i.e., removing mixed partials). We will consider only the projected down-beam direction, as this is sufficient for a study of critical angle selection; hence we strip $y$-dependence from the following calculations.

C.1.1.1. Steady state and first expansion: $\epsilon$-small perturbations

Following the application of the steady-state and translation-invariance assumptions \((\frac{\partial}{\partial x} \rightarrow 0, \frac{\partial}{\partial t} \rightarrow 0)\), linearization about small perturbations produces the steady-state equa-
\[
\frac{\partial}{\partial z}(\rho_0 w_0) = 0
\]
\[
\eta u_{0zz} = 2\eta f_A D_{13} r_{0z}
\]
\[
-p_0 + 2\eta w_{0zz} = 2\eta f_A D_{33} \frac{\partial \tau_0}{\partial z}
\]  \(C.2\)
\[
\rho_0 = \frac{\rho_a}{1 + \Delta_0}
\]
\[
w_0 \Delta_{0z} = \alpha_0 (z; 0, h_0).
\]
At \(z=0\), we have
\[
\Delta_0 = 0
\]
\[
u_0 = \beta u_{0z}
\]  \(C.3\)
\[
w_0 = V \left( \frac{\rho_c}{\rho_a} \right),
\]
representing the steady-state equations of the no-slip and no-penetration conditions respectively in the downward-translating frame. At \(z=h_0\), we have
\[
0 = w_0 - V \frac{\rho_c}{\rho_0}
\]
\[
u_{0,z} = 2f_A D_{1} \tau_0 (z; 0, h_0) D_{13}
\]  \(C.4\)
\[
p_0 = 2\eta \{ w_{0z} - f_A D_{1} \tau_0 (z; 0, h_0) D_{33} \},
\]
where the first equation is the steady-state equation of the modified kinematic condition, and the second two equations are due to the steady-state stress balance \(T_0 \cdot \mathbf{n}_0 = 0\) at the upper interface. We also find the linearized equations at \(O(\epsilon)\) as
\[
\rho_1 t + \rho_0 u_1 x + \rho_0 z w_1 + \rho_0 w_1 z + \rho_1 z w_0 + \rho_1 w_0 z = 0
\]
\[
-p_1 x + \eta \{ 2u_{1xx} + u_{1zz} + w_{1xz} \} = 2f_A D \eta \{ D_{11} \tau_{\epsilon, x} + D_{13} \tau_{\epsilon, z} \}
\]
\[
-p_1 z + \eta \{ w_{1xx} + 2w_{1zz} + u_{1xx} \} = 2f_A D \eta \{ D_{31} \tau_{\epsilon, x} + D_{33} \tau_{\epsilon, z} \}
\]  \(C.5\)
\[
\rho_1 = -\rho^* \frac{\Delta_1}{(1 + \Delta_0)^2}
\]
\[
\Delta_{1t} + u_0 \Delta_{1x} + w_0 \Delta_{1z} + w_1 \Delta_{0z} = \alpha_\epsilon
\]
in the bulk, representing conservation of mass and momentum; the equation of state; and the advection of the volume-change respectively. Now at \( z = 0 \),

\[
\Delta_{0,z}(z; 0, h_0)g_1 + \Delta_1(z; 0, h_0) = 0
\]

\[
u_{0z}g_1 + u_1 + g_{1z}(w_0 - V) = \beta (g_1u_{0zz} + u_{1z} + g_{1x}(w_{0z} - u_{0z})) \tag{C.6}
\]

\[-g_{1z}u_0 + w_{0z}g_1 + w_1 = \left( \frac{\rho_a - \rho_c}{\rho_a} \right) g_{1t} \]

At \( z = h_0 \),

\[h_{1t} = w_1 - u_0h_{1x} + h_1w_{0z} + \frac{V\rho_c}{\rho_0^2} \{\rho_0h_1 + \rho_1\} - h_{1x}V_{e,h} \frac{\rho_c}{\rho_0} \]

\[2\eta\{\frac{1}{2}(u_{1z} + w_{1x}) - fA_D[\tau_hh_1 + \tau_gg_1]D_{13}\} - h_{1x}T_{011}^{11} = 0 \tag{C.7}
\]

\[-p_1 + 2\eta\{w_{1z} - fA_D[\tau_hh_1 + \tau_gg_1]D_{33}\} + T_{033}^{33} = 0, \]

where the first equation is due to the linearization of the kinematic condition and the second two are due to the linearization of the stress balance,

\[T_0 \cdot \hat{n}_0 + \left[ \frac{\partial T_0}{\partial z} \cdot h_1 + T_1 \right] \cdot \hat{n}_0 = \vec{0}. \tag{C.8}\]

Here, we have \( \hat{n}_0 = \langle 0, 0, 1 \rangle \) and \( \hat{n}_1 = \langle -h_{1x}, -h_{1y}, 0 \rangle \). \( T_{011}^{11} \) denotes the upper-left component of steady-state stress tensor \( T_0 \), and \( T_{033}^{33} \) denotes the bottom-right component. The component indices are denoted as superscripts to distinguish them from the subscripts which elsewhere denote terms in the expansion.

**C.1.1.2. Second expansion: small perturbative wavenumber \( k \)**

We now seek the long-wave linear dispersion relation, since we anticipate that pattern formation in this unary material should be governed by a so-called Type II bifurcation [6]. This prompts the expansion of the dispersion relation \( \sigma \approx 0 \) in small wavenumber \( k \) as

\[\sigma = 0 + k\sigma_1 + k^2\sigma_2 + O(k^3), \tag{C.9}\]

and we obtain the following systems at each order in \( k \).
At O(1):

\[
\rho_0 \tilde{w}_{10} + \rho_0 \tilde{w}'_{10} + \tilde{\rho}'_{10} w_0 + \tilde{\rho}_{10} w_{0z} = 0 \\
\tilde{u}''_{10} = 2 f A_D D_{13} \tau_{e,z} \\
-\tilde{p}'_{10} + 2 \eta \tilde{w}''_{10} = 2 f A_D \eta \{D_{33} \tau_{e,z}\} \\
\tilde{p}_{10} = \frac{-\rho_a \tilde{\Delta}_{10}}{(1 + \Delta_0)^2} \\
w_0 \tilde{\Delta}'_{10} + \tilde{w}_{10} \Delta_{0z} = \alpha_e. 
\]

At \( z = 0 \),

\[
\Delta_0 g_1 + \tilde{\Delta}_{10} = 0 \\
\tilde{u}_{10} + u_{0z} \tilde{g}_1 = \beta (\tilde{g}_1 u_{0zz} + \tilde{u}_{10z}) \quad (C.11) \\
\tilde{w}_{10} + w_{0z} \tilde{g}_1 = 0 
\]

At \( z = h_0 \),

\[
\tilde{w}_{10} + \tilde{h}_1 w_{0z} + \frac{V \rho^*}{\rho_0^2} \{\rho_0 \tilde{h}_1 + \tilde{\rho}_{10}\} = 0 \\
2 \eta \{\tilde{u}'_{10} - f A_D \tau_{e} D_{13}\} = 0 \quad (C.12) \\
-\tilde{p}_{10} + 2 \eta \{\tilde{w}'_{10} - f A_D \tau_{e} D_{33}\} + T_{0z}^{33} = 0 
\]

At O(k):

\[
\sigma_1 \tilde{p}_{10} + i \rho_0 \tilde{u}_{10} + \rho_{0z} \tilde{w}_{11} + \rho_0 \tilde{w}'_{11} + \tilde{\rho}'_{11} w_0 + \tilde{\rho}_{11} w_{0z} = 0 \\
-\tilde{p}_{11} + \eta \{\tilde{w}''_{11} + i \tilde{w}'_{11}\} = 2 f A_D \eta i \{D_{11} \tau_e\} \\
-\tilde{p}'_{11} + \eta \{2 \tilde{w}''_{11} + i \tilde{w}'_{11}\} = 2 f A_D \eta i \{D_{31} \tau_e\} \quad (C.13) \\
\tilde{p}_{11} = \frac{-\rho_a \tilde{\Delta}_{11}}{(1 + \Delta_0)^2} \\
\sigma_1 \tilde{\Delta}_{10} + i \rho_0 \tilde{\Delta}_{10} + w_0 \tilde{\Delta}'_{11} + \tilde{w}_{11} \Delta_{0z} = 0 
\]

At \( z = 0 \),

\[
\tilde{\Delta}_{11} = 0 \\
\tilde{u}_{11} + i \tilde{g}_1 (w_0 - V) = \beta (\tilde{u}_{11z} + i \tilde{g}_1 (w_{0z} - u_{0z})) \quad (C.14) \\
-\tilde{g}_1 u_0 + \tilde{w}_{11} = \left(\frac{\rho_a - \rho_c}{\rho_a}\right) \sigma_1 \tilde{g}_1 
\]
At $z = h_0$,
\[
\begin{align*}
\sigma_1 \tilde{h}_1 &= \tilde{w}_{11} - u_0 i \tilde{h}_1 + \frac{V \rho_c}{\rho_0^2} \tilde{\rho}_{11} \\
2\eta \left\{ \frac{1}{2} [\tilde{u}_{11}^' + i \tilde{w}_{10}] \right\} - i \tilde{h}_1 T_0^{11} &= 0 \\
-\tilde{p}_{11} + 2\eta \tilde{w}_{11}^' &= 0
\end{align*}
\] (C.15)

At $O(k^2)$:
\[
\begin{align*}
\sigma_1 \tilde{\rho}_{11} + \sigma_2 \tilde{\rho}_{10} + i \rho_0 \tilde{u}_{11} + \rho_{0z} \tilde{w}_{12} + \rho_0 \tilde{w}_{12}^' + \tilde{\rho}_{12} w_0 + \tilde{\rho}_{12} w_0^z &= 0 \\
- i \tilde{p}_{11} + \eta \left\{ -2 \tilde{u}_{10} + \tilde{u}_{12}^'' + i \tilde{w}_{11}^' \right\} &= 0 \\
- \tilde{p}_{12}^' + \eta \left\{ -\tilde{w}_{10} + 2 \tilde{w}_{12}^'' + i \tilde{u}_{11}^' \right\} &= 0 \\
\tilde{\rho}_{12} &= \frac{-\rho_a \tilde{\Delta}_{12}}{[1 + \Delta_0]^2} \\
\sigma_1 \tilde{\Delta}_{11} + \sigma_2 \tilde{\Delta}_{10} + i u_0 \tilde{\Delta}_{11} + w_0 \tilde{\Delta}_{12} + \tilde{w}_{12} \Delta_{0z} &= 0
\end{align*}
\] (C.16)

At $z=0$,
\[
\tilde{\Delta}_{12} = 0 \\
\tilde{u}_{12} = \beta \tilde{u}_{12z} \\
\tilde{w}_{12} = \left( \frac{\rho_a - \rho_c}{\rho_a} \right) \sigma_2 \tilde{g}_1,
\] (C.17)

and at $z=h_0$,
\[
\begin{align*}
\sigma_2 \tilde{h}_1 &= \tilde{w}_{12} + \frac{V \rho_c}{\rho_0^2} \tilde{\rho}_{12} \\
\eta \left\{ \tilde{u}_{12}^' + i \tilde{w}_{11} \right\} &= 0 \\
-\tilde{p}_{12} + 2\eta \tilde{w}_{12}^' &= 0
\end{align*}
\] (C.18)

C.1.1.3. Third expansion: small swelling rate $f A_f$

Bearing in mind that we expect the swelling rate in the traveling frame to be fairly small (that is, there isn’t much swelling prior to a parcel of matter being sputtered away), and observing from previous results [50] that the effect of even large swelling rates is highly self-similar at all wave numbers, and uniformly stabilizing for long waves, we consider a second
expansion in a small mean swelling rate, \( fA_I \). Hence we take

\[
\alpha = \alpha_0 + fA_I \alpha_1 + ... \\
\rho_0 = \rho_{00} + fA_I \rho_{01} + ... \\
... \\
\sigma_1 = \sigma_{10} + fA_I \sigma_{11} + ... \\
\tilde{\rho}_{10} = \tilde{\rho}_{100} + fA_I \tilde{\rho}_{111} + ... \\
...
\]

(C.19)

Now, because the equations for the leading order terms are obvious from the above (simply by appending a “0” to the subscript of each term), we shall only write out explicitly the equations expanded in \( fA_I \). Because the stress tensor associated with anisotropic plastic flow is assumed, here, to be independent of swelling rate, there is no need for us to expand \( \tau \to \tau_0 + fA_I \tau_1 \). In a more careful exploration of the interplay between volumization and stress-free strain through local changes in the density, such an expansion would be required.

**Steady state at \( O(fA_I) \):**

\[
\frac{\partial}{\partial z} (\rho_{00} w_{01} + \rho_{01} w_{00}) = 0 \\
\eta u_{01zz} = 0 \\
-p_{01z} + 2\eta w_{01zz} = 0 \\
\rho_{01} = \frac{-\rho_0 \Delta_{01}}{(1 + \Delta_{00})^2} \\
w_{00} \Delta_{01z} + w_{01} \Delta_{00z} = \alpha_1(z; 0, h_0).
\]

At \( z=0 \):

\[
\Delta_{01} = 0 \\
u_{01} = \beta u_{01z} \\
w_{01} = 0.
\]

(C.20)
At \( z = h_0 \),

\[
\begin{align*}
\dot{w}_{01} + V \rho_c \frac{\rho_{01}}{\rho_{00}^2} &= 0 \\
\dot{u}_{01z} &= 0 \\
-p_{01} + 2\eta \dot{w}_{01z} &= 0.
\end{align*}
\]  \tag{C.22}

At \( O(f_A I) \):

\[
\begin{align*}
\dot{\rho}_{00z} \dot{w}_{101} + \rho_{01z} \dot{w}_{100} + \rho_{00} \dot{w}_{101z} + \rho_{01} \dot{w}_{100z} + \dot{\rho}_{100z} \dot{w}_{01} + \dot{\rho}_{101z} \dot{w}_{00} + \dot{\rho}_{100} w_{01z} + \dot{\rho}_{101} w_{00z} &= 0 \\
\ddot{u}_{101zz} &= 0 \\
-p_{101z} + 2\eta \ddot{w}_{101zz} &= 0 \\
\dot{w}_{100} \ddot{\Delta}_{101z} + \dot{w}_{01} \ddot{\Delta}_{100z} + \dot{w}_{100} \Delta_{01z} + \dot{w}_{101} \Delta_{00z} &= \alpha e_1 \\
\ddot{\rho}_{101} &= -\rho_a \left\{ \Delta_{00} \ddot{\Delta}_{101} + \ddot{\Delta}_{101} - 2 \Delta_{01} \ddot{\Delta}_{100} \right\} \\
&\quad (1 + \Delta_{00})^3 \tag{C.23}
\end{align*}
\]

At \( z = 0 \):

\[
\begin{align*}
\Delta_{01z} \ddot{g}_1 + \ddot{\Delta}_{101} &= 0 \\
\ddot{u}_{101} + u_{01z} g_1 &= \beta (\ddot{g}_1 u_{01zz} + \ddot{u}_{101z}) \tag{C.24}
\end{align*}
\]

At \( z = h_0 \):

\[
\begin{align*}
\ddot{w}_{101} + \ddot{h}_1 w_{01z} + V \rho_c \left\{ \rho_{00} (\rho_{01z} h_1 + \ddot{\rho}_{101}) - 2 \rho_{01} (\rho_{00z} h_1 + \ddot{\rho}_{100}) \right\} \rho_{00}^3 &= 0 \\
\ddot{u}_{101} &= 0 \\
-p_{101} + 2\eta \ddot{w}_{101} + T_{01z}^{33} &= 0. \tag{C.25}
\end{align*}
\]
At $O(k f A_r)$:

$$
\sigma_{10}\tilde{\rho}_{101} + \sigma_{11}\tilde{\rho}_{10} + i[\rho_{00}\tilde{u}_{101} + \rho_0 \tilde{u}_{100}] + [\rho_{00}\tilde{w}_{111} + \rho_{01z}\tilde{w}_{110}]
+ [\rho_{00}\tilde{w}'_{111} + \rho_{01}\tilde{w}'_{110}] + [\tilde{\rho}'_{110}\tilde{w}_{01} + \tilde{\rho}'_{111}\tilde{w}_{00}] + [\tilde{\rho}_{110}\tilde{w}_{01z} + \tilde{\rho}_{111}\tilde{w}_{00z}] = 0
$$

$$
-\tilde{\rho}_{101} + \eta\{\tilde{u}''_{111} + i\tilde{u}'_{101}\} = 0
$$

$$
-\tilde{\eta}_{111} + \eta\{2\tilde{w}''_{111} + i\tilde{u}'_{101}\} = 0
$$

$$
\sigma_{10}\tilde{\Delta}_{101} + \sigma_{11}\tilde{\Delta}_{100} + i[u_{00}\tilde{\Delta}_{101} + u_{01}\tilde{\Delta}_{100}] + w_{00}\tilde{\Delta}'_{111} + w_{01}\tilde{\Delta}'_{110} + \tilde{w}_{110}\tilde{\Delta}_{01z} + \tilde{w}_{111}\tilde{\Delta}_{00z} = 0
$$

$$
\tilde{\rho}_{111} = -\rho_a\{\Delta_{00}\tilde{\Delta}_{111} + \tilde{\Delta}_{111} - 2\Delta_{01}\tilde{\Delta}_{110}\} / (1 + \Delta_{00})^3
$$

(C.26)

At $z=0$:

$$
\tilde{\Delta}_{111} = 0
$$

$$
\tilde{u}_{111} + i\tilde{g}_1 w_{01} = \beta(\tilde{u}_{111z} + i\tilde{g}_1(w_{01z} - u_{01z}))
$$

(C.27)

$$
-i\tilde{g}_1 u_{01} + \tilde{w}_{111} = \left(\frac{\rho_a - \rho_c}{\rho_a}\right) \sigma_{11}\tilde{g}_1.
$$

At $z=h_0$:

$$
\sigma_{11}\tilde{h}_1 = \tilde{w}_{111} - u_{01}i\tilde{h}_1 + \frac{V \rho_c\rho_{00}\tilde{\rho}_{111} - 2\rho_{01}\tilde{\rho}_{110}}{\rho_{00}^3}
$$

$$
-i\tilde{h}_1 T_{01}^{11} + \eta\{\tilde{u}'_{111} + i\tilde{w}'_{101}\} = 0
$$

$$
-\tilde{p}_{111} + 2\eta\tilde{w}'_{111} = 0
$$

(C.28)

At $O(k^2 f A_r)$:

$$
\sigma_{10}\tilde{\rho}_{111} + \sigma_{11}\tilde{\rho}_{110} + [\sigma_{20}\tilde{\rho}_{101} + \sigma_{21}\tilde{\rho}_{100}] + i[\rho_{00}\tilde{u}_{111} + \rho_0 \tilde{u}_{110}]
+ [\rho_{00}\tilde{w}_{121} + \rho_{01z}\tilde{w}_{120}] + [\rho_{00}\tilde{w}'_{121} + \rho_{01}\tilde{w}'_{120}] + [\tilde{\rho}'_{120}\tilde{w}_{01} + \tilde{\rho}'_{121}\tilde{w}_{00}] + [\tilde{\rho}_{120}\tilde{w}_{01z} + \tilde{\rho}_{121}\tilde{w}_{00z}] = 0
$$

$$
-\tilde{\rho}_{111} + \eta\{-2\tilde{u}_{101} + \tilde{u}'_{121} + i\tilde{w}'_{111}\} = 0
$$

$$
-\tilde{\rho}_{121} + \eta\{-\tilde{w}_{101} + 2\tilde{w}''_{121} + i\tilde{u}'_{111}\} = 0
$$

$$
\sigma_{10}\tilde{\Delta}_{111} + \sigma_{11}\tilde{\Delta}_{110} + \sigma_{20}\tilde{\Delta}_{101} + \sigma_{21}\tilde{\Delta}_{100} + i[u_{00}\tilde{\Delta}_{111} + u_{01}\tilde{\Delta}_{110}]
+ w_{00}\tilde{\Delta}'_{121} + w_{01}\tilde{\Delta}'_{120} + \tilde{w}_{120}\tilde{\Delta}_{01z} + \tilde{w}_{121}\tilde{\Delta}_{00z} = 0
$$

$$
\tilde{\rho}_{121} = -\rho_a\{\Delta_{00}\tilde{\Delta}_{121} + \tilde{\Delta}_{121} - 2\Delta_{01}\tilde{\Delta}_{120}\} / (1 + \Delta_{00})^3
$$

(C.29)
At $z=0$:
\[
\tilde{\Delta}_{121} = 0 \\
\tilde{u}_{121} = \beta \tilde{u}_{121,z} \\
\tilde{w}_{121} = \left( \frac{\rho_a - \rho_c}{\rho_a} \right) \sigma_{21} \tilde{g}_1.
\]

At $z=h_0$:
\[
\sigma_{21} \tilde{h}_1 = \tilde{w}_{121} + V \rho_c \left( \frac{\rho_{00} \tilde{\rho}_{121} - 2 \rho_{01} \tilde{\rho}_{120}}{\rho_{00}^3} \right) \\
\tilde{u}_{121} + i \tilde{w}_{111} = 0 \\
-\tilde{p}_{121} + 2 \eta \tilde{w}_{121}' = 0
\]

(C.31)

C.1.2. Solution

In the limit of small cross-terms, the same limit considered in Chapter 3, we obtain
\[
\sigma_{10} = \frac{-2 f A_D i D_{13}}{h_1} \int_0^{h_0} \left( \int_0^z \tau_1(z_1) dz_1 + \beta \left( \tilde{g}_1 \tau_{0,z}(0) + \tau_1(0) \right) - \tilde{g}_1 \tau_{0}(0) \right) dz \\
+ 2 f A_D i \beta D_{13} \tau_{0}(0) \frac{\tilde{g}_1}{h_1} \\
+ \sigma_{10} \left( 1 - \frac{\rho_c}{\rho_a} \right) \left( \frac{\tilde{g}_1}{h_1} \right) \\
-2 f A_D i D_{13} \left( \int_0^{h_0} \tau_0(z) dz + \beta \tau_{0}(0) \right)
\]

(C.32)
\[
\sigma_{11} = \left( 1 - \frac{\rho_c}{\rho_a} \right) \sigma_{11} \left( \frac{\tilde{g}_1}{h_1} \right); \\
\sigma_{20} = \frac{2 f A_D (D_{11} - D_{33})}{h_1} \int_0^{h_0} \int_0^z \int_0^{z_1} \tau_2(z_2) dz_2 dz_1 dz \\
- \frac{i}{h_1} \left( \frac{c_9 h_0^2}{2} + c_{10} h_0 \right) + \left( 1 - \frac{\rho_c}{\rho_a} \right) \sigma_{20} \left( \frac{\rho_c}{\rho_a} \right)
\]

(C.34)
and

where
\[
c_9 = -2i h_1 f A_D \tau_0(h_0) \left( D_{11} - D_{33} \right) - 2 f A_D i \left( D_{11} - D_{33} \right) \int_0^{h_0} \tau_2(z) dz; \\
c_{10} = i \tilde{g}_1 V \left( 1 - \frac{\rho_c}{\rho_a} \right) + \beta c_9
\]

(C.35)
and

\[
\sigma_{21} = \frac{1}{h_1} \int_{0}^{h_0} \left( \frac{\sigma_{20,0}}{V} \left( \frac{\rho_c}{\rho_a} \right) \left[ \int_{0}^{z} \alpha_{\epsilon_1}(z_1) dz_1 - \tilde{g}_1 \alpha_{01}(0) \right] + \int_{0}^{z} \int_{0}^{z_1} \alpha_{\epsilon_1}(z_2) dz_2 dz_1 - ic_{13}z - ic_{14} \right) + \frac{\tilde{w}_{120,0}}{V} \left( \frac{\rho_c}{\rho_a} \right) \alpha_{01}(z) dz + \left( 1 - \frac{\rho_c}{\rho_a} \right) \sigma_{21} \left( \frac{\tilde{g}_1}{h_1} \right),
\]

where

\[
c_{13} = -2i\tilde{h}_1 \alpha_{01}(h_0) - 2i \int_{0}^{h_0} \alpha_{\epsilon_1}(z) dz + i\tilde{g}_1 \alpha_{01}(0)
\]

\[
c_{14} = \beta \left[ c_{13} + i\tilde{g}_1 \alpha_{01}(0) \right];
\]

\[
\sigma_{20,0} = \frac{h_0 V \left( 1 - \frac{\rho_c}{\rho_a} \right) \left( \frac{\tilde{g}_1}{h_1} \right)}{1 - \left( 1 - \frac{\rho_c}{\rho_a} \right) \left( \frac{\tilde{g}_1}{h_1} \right)}
\]

\[
\tilde{w}_{120,0} = \tilde{g}_1 V \left( 1 - \frac{\rho_c}{\rho_a} \right) z + \left( 1 - \frac{\rho_c}{\rho_a} \right) \sigma_{20,0} \tilde{g}_1.
\]

### C.2. Details of modified interfacial and boundary conditions

#### C.2.1. Derivation of general mass-conserving boundary condition

It is worth recognizing that the widely-used “no-penetration” condition (i.e., \( w = 0 \)) at the crystalline-amorphous interface is fundamentally a statement of mass conservation at the interface: that is, no mass crosses the boundary. The typical \( w = 0 \) emerges as a result of this mass conservation at the interface provided that the density on each side of the interface is the same. Physically, it is well-known that, for example, a-Si and c-Si have different densities, varying by perhaps 3-8\% [87, 67, 22]. Here, we will consider a loosening of the typical no-penetration condition in favor of a generalized statement of mass-conservation at the interface which takes into account the change in density across that interface. We denote \( \rho_a, \tilde{v}_a \) as the density and velocity fields within the amorphous layer, and likewise \( c \) subscripts denote the same within the underlying crystalline substrate. Hence we consider the differential form of bulk mass conservation,

\[
\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \tilde{v}) = 0,
\]

\[(C.38)\]
where we integrate over a control volume $V$ at the lower interface:

$$
\int_V \left( \frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \vec{v}) \right) dV = 0.
$$

(C.39)

Allowing the control volume to move with the interface, hence time-invariant in the moving frame, and applying the typical Divergence Theorem, we may rewrite the above as

$$
\frac{\partial}{\partial t} \int_V \rho dV + \int_S \rho \vec{v} \cdot \hat{n} dS,
$$

(C.40)

where $\hat{n}$ is the vector normal to the interface $S$. Taking the limit as the control volume’s extent in the $\hat{n}$ direction approaches zero and taking the interface to move with velocity $v_{I,g} = \vec{v}_{I,g} \cdot \hat{n}_g$, we find

$$
\rho_c (\vec{v}_c - \vec{v}_{I,g}) \cdot \hat{n} - \rho_a (\vec{v}_a - \vec{v}_{I,g}) \cdot \hat{n} = 0,
$$

(C.41)

and rearrangement leads to

$$
\rho_a \vec{v}_a \cdot \hat{n} - \rho_c \vec{v}_c \cdot \hat{n} = (\rho_a - \rho_c) v_{I,g}.
$$

(C.42)

With $\vec{v}_c = \vec{0}$, which is also clearly nonphysical but is required to avoid needing to consider the stress tensor describing the crystalline (or possible polycrystalline, but not quite amorphous) solid, we have

$$
\rho_a \vec{v}_a \cdot \hat{n} = (\rho_a - \rho_c) v_{I,g}
$$

or, as a jump relation,

$$
[\rho \vec{v}] \cdot \hat{n} = [\rho] v_{I,g}
$$

and conversion to the traveling frame via

$$
\vec{v} \rightarrow \vec{v} - V \hat{k}.
$$

(C.44)

Then, we have

$$
v_{I,g} \rightarrow v_{I,g} - V \hat{k} \cdot \hat{n}_g
$$

(C.45)
due to ongoing erosion. These lead to

$$
\rho_a \vec{v}_a \cdot \hat{n}_g = (\rho_a - \rho_c) (v_{I,g} - V \hat{k} \cdot \hat{n}_g) + \rho_a V \hat{k} \cdot \hat{n}_g
$$

$$
\vec{v}_a \cdot \hat{n}_g = \left( \frac{\rho_a - \rho_c}{\rho_a} \right) (v_{I,g} - V \hat{k} \cdot \hat{n}_g) + V \hat{k} \cdot \hat{n}_g
$$

$$
\vec{v}_a \cdot \hat{n}_g = \left( 1 - \frac{\rho_c}{\rho_a} \right) v_{I,g} - \left( 1 - \frac{\rho_c}{\rho_a} \right) V \hat{k} \cdot \hat{n}_g + V \hat{k} \cdot \hat{n}_g
$$

We will now drop the subscript $a$ as it is clear that the only bulk velocity field under consideration is that of the amorphous layer. In principle, we have made the assumption that the motion of the amorphous bulk is much faster than that of the underlying crystalline substrate. It is also clear that when $\rho_a = \rho_c$ (i.e., there is no density drop across the interface), and if we assume the typical no-slip condition $u = 0$ at $z = g$, we recover

$$
\vec{v} = V \hat{k}.
$$

(C.47)

This is as was seen in [48].

C.2.2. Linearization of general mass-conserving boundary condition

From

$$
\vec{v} \cdot \hat{n}_g = \left( 1 - \frac{\rho_c}{\rho_a} \right) v_{I,g} + \left( \frac{\rho_c}{\rho_a} \right) V \hat{k} \cdot \hat{n}_g,
$$

we obtain the following expansions in Fourier modes. At $O(k^0)$,

$$
\tilde{g}_1 u_{0z} + \tilde{w}_{100} = 0.
$$

(C.49)

At $O(k^1)$,

$$
-i \tilde{g}_1 u_0 + \tilde{w}_{110} = \sigma_{10} \left( 1 - \frac{\rho_c}{\rho_a} \right) \tilde{g}_1.
$$

(C.50)

At $O(k^2)$,

$$
\tilde{w}_{120} = \sigma_{20} \left( 1 - \frac{\rho_c}{\rho_a} \right) \tilde{g}_1.
$$

(C.51)

Expansion in $f A_f$ is straightforward. Then we arrive at the boundary conditions in the main text. We note that this condition is more typical of the solidification theory literature,
and is featured prominently in Chapter 9 of [85] and elsewhere, while being generally absent from most other resources of continuum mechanics (where phase transitions are seldom of interest).

C.2.3. Linearization of slip-flow boundary condition

In the x-z plane, we have a Navier-type slip-flow condition,

\[ \vec{v} \cdot \hat{t} = \beta_s \hat{n} \cdot \nabla (\vec{v} \cdot \hat{t}), \]  

(C.52)

where \( \beta_s \) is a slip length. Linearization leads to steady-state

\[ u_0 = \beta u_{0z}, \]  

(C.53)

and at \( O(\epsilon) \), before expanding in modes,

\[ u_1 + u_{0z}g_1 + g_{1z}(w_0 - V) = \beta_s (g_1u_{zz} + u_{1z} + g_{1z}(w_{0z} - u_{0z})). \]  

(C.54)

Now expanding in modes, we have at \( O(\epsilon k^0) \):

\[ \tilde{u}_{100} + u_{0z}\tilde{g}_1 = \beta_s (\tilde{g}_1u_{0zz} + \tilde{u}_{100z}), \]  

(C.55)

at \( O(\epsilon k) \):

\[ \tilde{u}_{110} + i\tilde{g}_1(w_0 - V) = \beta_s (\tilde{u}_{110z} + i\tilde{g}_1(w_{0z} - u_{0z})), \]  

(C.56)

and at \( O(\epsilon k^2) \):

\[ \tilde{u}_{120} = \beta_s \tilde{u}_{120}'. \]  

(C.57)

The expansion in \( fA_I \) is similar. At steady-state, we have

\[ u_{01} = \beta_s u_{01z} \]  

(C.58)

Then, expanding, we have at \( O(\epsilon k^0 fA_I) \):

\[ \tilde{u}_{101} + u_{01z}\tilde{g}_1 = \beta_s (\tilde{g}_1u_{01zz} + \tilde{u}_{101z}), \]  

(C.59)

at \( O(\epsilon k^1 fA_I) \):

\[ \tilde{u}_{111} + i\tilde{g}_1w_{01} = \beta_s (\tilde{u}_{111z} + i\tilde{g}_1(w_{01z} - u_{01z})), \]  

(C.60)
and at $O(\epsilon k^2 f A_f)$:

$$\tilde{u}_{121} = \beta_s \tilde{u}_{121}'.$$

(C.61)

C.3. Review of Yamamura yield function and erosion velocity

In the present work, we require an estimate of the steady-state velocity $V$ at which the thin film translates downward (in the laboratory frame) due to erosion. We may compute $V$ by

$$V = f \cos(\theta) Y(\theta) \Omega,$$

(C.62)

where $f$ is the beam flux, $\theta$ is (as elsewhere) the beam angle, $Y(\theta)$ is the yield function, and $\Omega$ is atomic volume for the irradiated target. While many empirical and semi-empirical descriptions of $Y(\theta)$ exist [115, 116, 79, 62], we will make use of the semi-empirical yield function due to Yamamura [115], which we will summarize here for convenience. We take

$$Y(\theta) = Y(0) \sec f(\theta) \exp(-\Sigma(\sec(\theta) - 1)),$$

(C.63)

where

$$f = f_s \left[ 1 + 2.5\left(1 - \frac{\xi}{\xi}\right) \right]$$

(C.64)

and $f_s$ is an empirical ion-and-target-dependent fit parameter from tables in the text [115].

$$\xi = 1 - \left(\frac{E_{th}}{E}\right)^{1/2}$$

(C.65)

where $E_{th}$ is the threshold energy for the onset of sputtering, which can also be looked up from the tables in the text. $E$ is the beam energy expressed in eV.

$$\Sigma = f \cos(\theta_{opt}),$$

(C.66)

where

$$\theta_{opt} = 90^\circ - 286 \psi^{0.45}$$

(C.67)

is the angle, expressed in degrees, at which the erosion rate peaks, and

$$\psi(E) = \frac{\psi(1)}{\sqrt{E}}$$

(C.68)
is a function of beam energy expressed in eV. $\psi(1)$ may be looked up from tables in the text. $Y(0)$ is the number of target atoms sputtered per incoming ion, which can be computed from SRIM [78] or similar packages. Finally, $\Omega$ is the atomic volume of the target, which can be looked up from any one of many sources. With these quantities, we see that the yield function, hence the downward translation velocity, is a function of both irradiation angle and energy for a given ion-target combination. We also note that there is temperature dependence implicit in the above calculations: $\Omega$ is taken from values at 300K. Likewise, the experiments from which the empirical Yamamura parameters were fit were conducted at room temperature, and it should be expected that at very high or very low temperatures these parameters could change. Our present analysis is therefore restricted to near-room temperature.

**Atomic volume of Si and Ge.** We compute the atomic volume for Si as $12.1 \frac{\text{cm}^3}{\text{mol}} = .02 \frac{\text{nm}^3}{\text{atom}}$, and for Ge, $13.6 \frac{\text{cm}^3}{\text{mol}} = .02258 \frac{\text{nm}^3}{\text{atom}}$. 
Appendix D

Appendix to Chapter 5

D.1. Details of calculation

D.1.1. Linearization

From where the main text leaves off, we have taken

\[ h(X) = h_0 + \epsilon h_1(X), \]  

which leads to the expression for the integrand of the above,

\[ I_0 = \exp \left( - \frac{[(x - X) \sin(\theta) - (z - h_0) \cos(\theta) - a]^2}{2\alpha^2} - \frac{[(x - X) \cos(\theta) + (z - h_0) \sin(\theta)]^2}{2\beta^2} \right) \]  

Next we apply the linear stability ansatz to the above in spatial variable \( X \), retaining \( x \) as a fixed constant for the moment, being the \( x \) coordinate at which we wish to determine mean power deposition. Hence

\[ h_1(X) \rightarrow \tilde{h}_1 \exp(ikX + \sigma t). \]
We now wish to integrate the linearized quantity $I$ from $X = -\infty$ to $X = +\infty$ in order to account for the flux-diluted power deposition at a fixed $(x, z)$ due to ions entering through all points $(X, h(X))$. We make a change of variables $p := x - X$, so $dX = -dp$. The linearization combined with this change of variables allows us to consider

$$\exp(ikX) = \exp(ikx + \sigma t) \exp(-ikp);$$

this is useful because we expect the cancellation of a factor of $\exp(ikx + \sigma t)$ so that we can solve the linearized differential equations in the $z$ coordinate alone, as is typical in such calculations. Now this leads to

$$P(x, z) = \frac{\cos(\theta)}{2\pi \alpha \beta} \int_{-\infty}^{\infty} I_0 dp$$

$$+ \tilde{h}_1 \exp(ikx + \sigma t) \frac{\cos(\theta)}{2\pi \alpha^3 \beta^3} \left( c_1 \int_{-\infty}^{\infty} \exp(-ikp) I_0 dp + c_2 \int_{-\infty}^{\infty} p \exp(-ikp) I_0 dp \right)$$

$$+ O(\epsilon^2),$$

reducing the problem at $O(\epsilon)$ to that of a Fourier transform, where we have

$$c_1 = (z - h_0) \left( (\alpha^2 \sin^2(\theta) + \beta^2 \cos^2(\theta)) \right) + \cos(\theta) \alpha \beta^2 + ik \alpha^2 \beta^2 \tan(\theta)$$

$$c_2 = \cos(\theta) \sin(\theta) (\alpha^2 - \beta^2),$$

grouping the coefficients of each integral in variable $p$. Note that $c_1$ has $z$-dependence, which will become relevant later. For the moment, both $(x, z)$ are fixed. Now we complete the square inside of the integrand of all three integrals, bringing the extra factors of $\exp(-ikp)$ into the argument of the $\exp()$ in the second two, which will allow some simplification. We identify

$$A = \frac{\cos^2(\theta)}{2\beta^2} + \frac{\sin^2(\theta)}{2\alpha^2}$$

$$B = \frac{(z - h_0) \sin(\theta) \cos(\theta)}{\beta^2} - \frac{\sin(\theta) ((z - h_0) \cos(\theta) + a)}{\alpha^2} + ik$$

$$\tilde{B} = B - ik$$

$$C = \frac{(z - h_0)^2 \sin^2(\theta)}{2\beta^2} + \frac{((z - h_0) \cos(\theta) + a)^2}{2\alpha^2},$$

so that we may express

$$I_0 = \exp \left( \frac{\tilde{B}^2}{4A} - C \right) \exp \left( \left[ \frac{p\sqrt{A}}{2\sqrt{A}} + \frac{\tilde{B}}{2\sqrt{A}} \right]^2 \right).$$
We will then incorporate \(-ikp\) into \(I_0\) prior to evaluation. Consider that

\[
\exp(-ikp)I_0 = \exp(-ikp) \exp\left(\frac{\tilde{B}^2}{4A} - C\right) \exp\left(-\left[p\sqrt{A} + \frac{\tilde{B}}{2\sqrt{A}}\right]^2\right)
\]

\[
= \exp\left(\frac{\tilde{B}^2}{4A} - C\right) \exp\left(-\left[p^2 A + p(\tilde{B} + ik) + \frac{\tilde{B}^2}{4A}\right]\right)
\]

\[
= \exp\left(\frac{B^2}{4A} - C\right) \exp\left(-\left[p^2 A + pB + \frac{B^2}{4A}\right]\right)
\]

\[
= \exp\left(\frac{B^2}{4A} - C\right) \exp\left(-\left[p\sqrt{A} + \frac{B}{2\sqrt{A}}\right]^2\right),
\]

a fact which will enable us to conserve some effort. It soon becomes clear that

\[
\int_{-\infty}^{\infty} I_0 dp = \frac{\sqrt{\pi}}{\sqrt{A}} \exp\left(\frac{\tilde{B}^2}{4A} - C\right),
\]

\[
\int_{-\infty}^{\infty} \exp(-ikp)I_0 dp = \frac{\sqrt{\pi}}{\sqrt{A}} \exp\left(\frac{B^2}{4A} - C\right),
\]

\[
\int_{-\infty}^{\infty} p \exp(-ikp)I_0 dp = -\frac{B\sqrt{\pi}}{2A^{3/2}} \exp\left(\frac{B^2}{4A} - C\right)
\]

This simplifies the resulting expressions and introduces a pleasant symmetry. Finally, we have a closed-form expression after carrying out the integration in \(p\):

\[
P(x, z) = \frac{\cos(\theta)}{2\pi\alpha\beta} \left[\frac{\sqrt{\pi}}{\sqrt{A}} \exp\left(\frac{\tilde{B}^2}{4A} - C\right)\right] + 
\]

\[
\epsilon \tilde{h}_1 \exp(ikx + \sigma t) \frac{\cos(\theta)}{2\sqrt{\pi}A^2 B} \exp\left(\frac{B^2}{4A} - C\right) \left(c_1 - \frac{Bc_2}{2A}\right)
\]

\[+O(\epsilon^2),\]

which will enable the matching of mean power deposition for an arbitrary Gaussian ellipsoid to stress modification within the amorphous bulk. We remind the reader that both the argument of the exponential and the term \((c_1 - \frac{Bc_2}{2A})\) retain \(z\)-dependence, will be important because the above \(P(x, z)\) will provide \(\tau_0(z)\) and \(\tau_\epsilon(z)\), or \(\alpha_{10}(z)\) and \(\alpha_{1\epsilon}(z)\). These expressions must then be integrated according to Equations (5.14)-(5.19), which leads to the
final linear stability result associated with the Gaussian ellipse characterized by the triplet 
\( (a, \alpha, \beta) \), Equation (5.37).

D.1.2. Integral evaluations

We identify
\[
P_0 = \frac{\cos(\theta)}{2\pi \alpha \beta} \left[ \frac{\sqrt{\pi}}{\sqrt{A}} \exp \left( \frac{B^2}{4A} - C \right) \right],
\]
the leading-order part of the depth-dependence extracted from the bivariate Gaussian intensity field. We will also identify
\[
P_1 = \frac{\cos(\theta)}{2\sqrt{\pi} A \alpha^3 \beta^3} \exp \left( \frac{B^2}{4A} - C \right) \left( c_1 - \frac{Bc_2}{2A} \right),
\]
the \( O(\epsilon) \) part of the depth-dependence of the stress tensor following the Fourier transform of the surface in the small-perturbation limit. From the functional form of the dispersion relation described earlier, we will be required to carry out one integral of \( P_0 \) and the first through third integrals of \( P_1 \). While we could, in principle, write numerical evaluation of these integrals into the script that will later generate our theoretical predictions for \( \theta_c \), it is actually more practical to obtain a closed-form solution, even if expressed in terms of complicated constants. This is because when we seek \( \theta_c \), we must run a root-finding algorithm (in the present work, we use Brent’s method implemented via \texttt{opt.brentq} in Python); at each step, the dispersion relation (Equation (5.37)) must be evaluated. If we rely solely on numerical integral evaluations, the multiple-integrals involved in the dispersion relation and the need to take small step sizes (to suppress error) leads to enormous computational cost, and sweeping over large regions in parameter space becomes prohibitive. We are therefore committed to the present analytical approach: not only in order to gain insight as to the parametric dependence of the growth rates prescribed by the present stress-based model, but also for computational speed.

\( P_1 \) evaluations. It is useful to rewrite \( P_1 \):
\[
\frac{\cos(\theta)}{2\sqrt{\pi} A \alpha^3 \beta^3} \exp \left( R - \frac{Q^2}{4P} \right) \exp \left( \left[ (z - h_0)\sqrt{P} + \frac{Q}{2\sqrt{P}} \right]^2 \right) (E(z - h_0) + F), \quad (D.15)
\]
where constants (with respect to \(z\)) \(E\) and \(F\) are from the rearrangement of \(c_1 - \frac{Bc_2}{A}\), so that we have
\[
E = \alpha^2 \sin^2(\theta) + \beta^2 \cos^2(\theta) - \frac{c_2}{2A} \left( \frac{\sin(\theta) \cos(\theta)}{\beta^2} - \frac{\sin(\theta) \cos(\theta)}{\alpha^2} \right) \tag{D.16}
\]
and
\[
F = \frac{c_2}{2A} \left( \sin(\theta) \frac{a}{\alpha^2} - ik \right) + a \beta^2 \cos(\theta) + i k \alpha^2 \beta^2 \tan(\theta)
\]
and
\[
P = \frac{1}{4A} \left( \frac{\sin^2(\theta) \cos^2(\theta)}{\beta^4} - \frac{2 \sin^2(\theta) \cos^2(\theta)}{\beta^2 \alpha^2} + \frac{\sin^2(\theta) \cos^2(\theta)}{\alpha^4} \right) - \left( \frac{\sin^2(\theta)}{2 \beta^2} + \frac{\cos^2(\theta)}{2 \alpha^2} \right) ;
\]
\[
Q = \frac{1}{4A} \left( -2 \sin^2(\theta) \cos(\theta) \frac{a}{\beta^2 \alpha^2} + 2 \sin(\theta) \cos(\theta) \frac{ik}{\beta^2} - \frac{2ik}{\alpha^2} \sin(\theta) \cos(\theta) + 2a \sin^2(\theta) \cos(\theta) \frac{1}{\alpha^4} \right)
\]
\[\quad - \frac{a \cos(\theta)}{\alpha^2} ;
\]
\[
R = \frac{1}{4A} \left( \frac{a^2 \sin^2(\theta)}{\alpha^4} - 2ik \sin(\theta) \frac{a}{\alpha^2} - k^2 \right) - \frac{a^2}{2 \alpha^2} \tag{D.17}
\]
so that the problem is reduced to multiple integrations of the product of an affine linear function and a Gaussian. We will define
\[
\tilde{P} = P \bigg|_{k=0} , \tilde{Q} = Q \bigg|_{k=0} , \tilde{R} = R \bigg|_{k=0} , \tilde{F} = F \bigg|_{k=0} \tag{D.18}
\]
here for later use.

**\(P_0\) evaluation.** Similarly, it is useful to rewrite \(P_0\) prior to evaluation. We take
\[
P_0 = \frac{\cos(\theta)}{2 \sqrt{\pi A} \alpha \beta} \exp \left( \frac{\tilde{B}^2}{4A} - C \right) = 
\]
\[\frac{\cos(\theta)}{2 \sqrt{\pi A} \alpha \beta} \exp \left( \tilde{R} - \frac{Q^2}{4\tilde{P}} \right) \exp \left( \sqrt{\tilde{P}} \left[ (z - h_0) \sqrt{\tilde{P}} + \frac{Q}{2\sqrt{\tilde{P}}} \right]^2 \right) \tag{D.19}
\]
so that the problem is reduced to a single integration of a Gaussian.
Expression in physical parameters. We re-express $P, Q, R, E, F$ in terms of basic physical quantities $\theta, a, \alpha, \beta$. We find

$$P = \frac{s^2 c^2 \alpha^2 \beta^2}{2[\alpha^2 c^2 + \beta^2 s^2]} \left[ \frac{1}{\beta^2} - \frac{1}{\alpha^2} \right]^2 - \left[ \frac{s^2}{2\beta^2} + \frac{c^2}{2\alpha^2} \right];$$

$$Q = -s c a^2 \beta^2 \left[ \frac{1}{\beta^2} - \frac{1}{\alpha^2} \right] \left[ \frac{sa}{\alpha^2} - ik \right] - \frac{ac}{\alpha^2};$$

$$R = \frac{2[\alpha^2 c^2 + \beta^2 s^2]}{2[\alpha^2 c^2 + \beta^2 s^2]} \left[ \frac{sa}{\alpha^2} - ik \right]^2 - \frac{a^2}{2\alpha^2};$$

$$E = \left[ \frac{\alpha^2 s^2 + \beta^2 c^2}{\alpha^2 c^2 + \beta^2 s^2} \right] - \left[ \frac{c^2 s^2 \alpha^2 \beta^2 (\alpha^2 - \beta^2)}{\alpha^2 c^2 + \beta^2 s^2} \right] \left[ \frac{1}{\beta^2} - \frac{1}{\alpha^2} \right];$$

$$F = \frac{c s a^2 \beta^2 (\alpha^2 - \beta^2)}{\alpha^2 c^2 + \beta^2 s^2} \left[ \frac{sa}{\alpha^2} - ik \right] + a \beta^2 c + i k \alpha^2 \beta^2 \tan(\theta),$$

where $s$ denotes $\sin(\theta)$ and $c$ denotes $\cos(\theta)$. As before, the “∼” version of a quantity denotes that $k = 0$ there.

D.1.3. Real part of coefficient for IIS linear dispersion relation: $\frac{\rho_a}{\rho_c} = 1$

Carrying out the integrals described above and substituting into the linear dispersion relation (Equations (5.14)-(5.19)), we find

$$\text{Re}(\sigma_{IIS}) = -\frac{f A_i \cos(\theta)}{64(\sqrt{A} \sqrt{\pi} P^{7/2} \alpha^3 \beta^3)} \exp \left(-\tilde{Q} h_0 - \frac{\tilde{Q}^2}{4\tilde{P}} + \tilde{R} \right) \omega_3 k^2 + O(k^4) \quad (D.21)$$

where

$$\omega_3 =$$

$$-2 \exp \left(\frac{\tilde{Q}^2}{4\tilde{P}}\right) \sqrt{\tilde{P}} \left[ e^{\tilde{h}_0} \left( 2\tilde{F} \tilde{P}(2\tilde{h}_0 \tilde{P} + \tilde{Q}) + E(4\tilde{h}_0^2 \tilde{P}^2 - \tilde{Q}^2 + \tilde{P}(4 - 2\tilde{h}_0 \tilde{Q})) \right) + 8\tilde{h}_0^2 \tilde{P}^3 \alpha^2 \beta^2 \right.$$  

$$+ e^{\tilde{Q} h_0} \left[ E(-4\tilde{P} - 8\tilde{h}_0^2 \tilde{P}^2 + \tilde{Q}^2) - 2(\tilde{F} \tilde{P} \tilde{Q} + 8\tilde{h}_0^2 \tilde{P}^3 \alpha^2 \beta^2) \right]$$

$$+ \sqrt{\pi} e^{\tilde{Q} h_0} \left( 2\tilde{F} \tilde{P}(2\tilde{P} + 8\tilde{h}_0^2 \tilde{P}^2 - \tilde{Q}^2) \right)$$

$$+ E\tilde{Q}(-6\tilde{P} - 8\tilde{h}_0^2 \tilde{P}^2 + \tilde{Q}^2) \left( \text{erfi}(\frac{2\tilde{h}_0 \tilde{P} - \tilde{Q}}{2\sqrt{\tilde{P}}}) + \text{erfi}(\frac{\tilde{Q}}{2\sqrt{\tilde{P}}}) \right)$$

and we find that the imaginary part, $\text{Im}(\sigma_{IIS}) = 0$.

D.1.4. Real part of coefficient for IIS linear dispersion relation: $\frac{\rho_a}{\rho_c} \neq 1$
In the case where we evaluate Equations (5.14)-(5.19) *without* the simplification $\frac{\rho_a}{\rho_c} \approx 1$ for the IIS term, we have

$$\text{Re}(\sigma_{IIS}) = -\frac{f A_I \cos(\theta) \rho_a}{64(\sqrt{A} \sqrt{\pi} \rho_c^3 \tilde{P}^{\gamma/2} \alpha^5 \beta^5)} \exp(\tilde{R}) \omega_4 k^2 + O(k^4) \quad (D.23)$$

where

$$\omega_4 = 2(1 + \exp(h_0(h_0 \tilde{P} - \tilde{Q}))) h_0 \tilde{P}^{\gamma/2} \left( -2 \tilde{F} \tilde{P} \rho_c^2 + E \tilde{Q} \rho_c^2 + 2Eh_0 \tilde{P}(2\rho_a^2 - 2\rho_a \rho_c + \rho_c^2) 
+ 4_0 \tilde{P}^2(2\rho_a^2 - 2\rho_a \rho_c + \rho_c^2) \alpha \beta \right) + 2 \left( 1 + \exp(h_0(h_0 \tilde{P} - \tilde{Q})) \right) \sqrt{\tilde{P}} \times \left( E(4h_0 \tilde{P}(h_0 \tilde{P} + \tilde{Q})\rho_a^2 - 4h_0 \tilde{P}(h_0 \tilde{P} + \tilde{Q})\rho_a \rho_c + (-2 \tilde{P}(2 + 3h_0 \tilde{P}) + h_0 \tilde{P} \tilde{Q} + \tilde{Q}^2)\rho_c^2) 
- \exp(\frac{-\tilde{Q}^2}{4\tilde{P}}) \sqrt{\pi}(E(4h_0 \tilde{P}(2 \tilde{P} - \tilde{Q}^2)\rho_a(\rho_a - \rho_c) + 8h_0 \tilde{P}^2 \tilde{Q} \rho_c^2 - \tilde{Q}(-6 \tilde{P} + \tilde{Q}^2)\rho_c^2) 
+ 2 \tilde{P}(4h_0 \tilde{P} \tilde{Q} \rho_a^2 - 4h_0 \tilde{P} \tilde{Q} \rho_a \rho_c + (-2 \tilde{P}(1 + 4h_0 \tilde{P}) + \tilde{Q}^2)\rho_c^2) 
+ 4 \tilde{P}(\rho_a - \rho_c)(2h_0 \tilde{P} \rho_a - \tilde{Q} \rho_c)\alpha^2 \beta^2) \right) \left( \text{erfi}(\frac{2h_0 \tilde{P} - \tilde{Q}}{2\sqrt{\tilde{P}}}) + \text{erfi}(\frac{\tilde{Q}}{2\sqrt{\tilde{P}}}) \right). \quad (D.24)$$

D.1.5. Real part of coefficient for APF linear dispersion relation

For reference, we also include the part of the long-wave linear dispersion relation due to APF distributed due to the Gaussian ellipsoid model, even though it hasn’t been used in the main text (it turned out to be unnecessary, and appears to actually worsen fitting to $\theta_c$ across the 15 systems surveyed). We find:

$$\text{Re}(\sigma_{APF}) = -\frac{3f A_D \cos(\theta)}{64A^{3/2} \sqrt{\pi} \tilde{P}^{\gamma/2} \alpha^5 \beta^5} \left( \cos(2\theta)\omega_1 - \sin(2\theta)\omega_2 \right) k^2 + O(k^4), \quad (D.25)$$
where

$$\omega_1 =$$

$$2Ae^{\bar{R}}\alpha^2\beta^2 \left[4h_0^2\bar{P}^2 \left[2\sqrt{\bar{P}} \left(E - e^{h_0(\bar{h}_0 - \bar{Q})} E + 2\bar{P}\alpha^2\beta^2\right) + \sqrt{\pi}e^{-\bar{Q}^2/4\bar{P}} \left(2\bar{F}\bar{P}ight)

- E\bar{Q}) \left(\text{erfi}\left(\frac{2h_0\bar{P} - \bar{Q}}{2\sqrt{\bar{P}}}\right) + \text{erfi}\left(\frac{\bar{Q}}{2\sqrt{\bar{P}}}\right)\right)\right) + e^{-\bar{Q}^2/4\bar{P}} \left[2e^{\bar{Q}(4\bar{h}_0 + \bar{Q})} \sqrt{\bar{P}} \left(e^{\bar{Q}h_0}(4E\bar{P}

+ 2\bar{F}\bar{P} - E\bar{Q}^2) + e^{\bar{P}h_0}(2\bar{F}\bar{P} + \bar{Q}) + E(4\bar{P}(-1 + h_0^2\bar{P}) + 2h_0\bar{P}\bar{Q} + \bar{Q}^2))\right)

+ \sqrt{\pi}(2\bar{F}\bar{P}(2\bar{P} - \bar{Q}^2) + E\bar{Q}(\bar{Q}^2 - 6\bar{P})) \left(\text{erfi}\left(\frac{2\bar{P}h_0 - \bar{Q}}{2\sqrt{\bar{P}}}\right) + \text{erfi}\left(\frac{\bar{Q}}{2\sqrt{\bar{P}}}\right)\right)\right]\right] $$

and

$$\omega_2 = \left[32Ae^{\bar{P}h_0 - \bar{Q}h_0 + \bar{R}x_0h_0\bar{P}^7/2\alpha^4\beta^4} - e^{-\bar{Q}^2/4\bar{P}} + \bar{R}\left[2a\bar{P}\beta^2 + \bar{Q}(\alpha^2 - \beta^2)c\right] \times

\left[2e^{\frac{\bar{Q}(4\bar{h}_0 + \bar{Q})}{4\bar{P}}} \sqrt{\bar{P}} \left(e^{\bar{Q}h_0(-2\bar{F}\bar{P} + E\bar{Q})} + e^{\bar{P}h_0}(2\bar{F}\bar{P} - E(2\bar{P}h_0 + \bar{Q}))\right)

+ \sqrt{\pi}\left[2E\bar{P} + 2\bar{F}\bar{P}\bar{Q} - E\bar{Q}^2\right] \left(\text{erfi}\left(\frac{2\bar{P}h_0 - \bar{Q}}{2\sqrt{\bar{P}}}\right) + \text{erfi}\left(\frac{\bar{Q}}{2\sqrt{\bar{P}}}\right)\right]\right] \sin(\theta)

+ 2\sqrt{\pi} \left[\left(e^{\frac{(-2\bar{h}_0^2 + \bar{Q})^2}{4\bar{P}}} - e^{\bar{Q}^2/4\bar{P}}\right) \left((2E\bar{P} + 2\bar{F}\bar{P}\bar{Q} - E\bar{Q}^2)(\alpha^2 - \beta^2)c\right)

+ 2\sqrt{\pi}\sqrt{\bar{P}} \left(\text{erfi}\left(\frac{2\bar{P}h_0 - \bar{Q}}{2\sqrt{\bar{P}}}\right) + \text{erfi}\left(\frac{\bar{Q}}{2\sqrt{\bar{P}}}\right)\right)\left(- (\bar{F}\bar{P} - E\bar{Q})(\alpha^2 - \beta^2)c\right)

+ \bar{P}\bar{Q}\alpha^2\beta^2(c_2 - 2A\alpha^2\beta^2 \tan(\theta))\right)

+ e^{\frac{\bar{Q}(4\bar{h}_0 + \bar{Q})}{4\bar{P}}} \left(\left(2h_0\bar{P} - \bar{Q}\right) \left(e^{\bar{Q}h_0}(E\bar{Q} - 2\bar{F}\bar{P}) + e^{\bar{P}h_0}(2\bar{F}\bar{P} - E(2\bar{P}h_0 + \bar{Q}))\right)\right) \left(\alpha^2 - \beta^2\right)c

+ 2\bar{P}\left[e^{\bar{P}h_0}E - e^{\bar{Q}h_0}(E - 2\bar{F}\bar{P}h_0 + E\bar{Q}h_0)\right] \left(\alpha^2 - \beta^2\right)cs

+ 2\left(e^{\bar{P}h_0} - e^{\bar{Q}h_0}\right)\bar{P}\alpha^2\beta^2\left(c_2 - 2A\alpha^2\beta^2 \tan(\theta)\right)\right]\right].$$

The above is in the case that $\frac{\rho_a}{\rho_c} = 1$.

D.1.6. Expressions for mean in-plane stress
From the analysis of the main text, we have

\[ \tau_0(z; 0, h_0) = \cos(\theta); \]
\[ \alpha_{01}(z; 0, h_0) = \frac{\cos(\theta)}{2\sqrt{\pi A\alpha\beta}} \exp\left(\frac{-(z - h_0 + a \cos(\theta))^2}{2(\alpha^2 \cos^2(\theta) + \beta^2 \sin^2(\theta))}\right), \] (D.28)

where the \( \cos(\theta) \) originates from flux dilution along an unperturbed, flat free interface. Now computing the mean about the film depth, we obtain

\[ \langle \alpha_{01}(z; 0, h_0) \rangle = \frac{\cos(\theta)}{2h_0} \left[ \text{erf}\left(\frac{a \cos(\theta)}{\sqrt{2\sqrt{\alpha^2 \cos^2(\theta) + \beta^2 \sin^2(\theta)}}}\right) - \text{erf}\left(\frac{a \cos(\theta) - h_0}{\sqrt{2\sqrt{\alpha^2 \cos^2(\theta) + \beta^2 \sin^2(\theta)}}}\right) \right], \] (D.29)

and simply \( \langle \tau_0(z; 0, h_0, ...) \rangle = \cos(\theta) \). The above expressions are used in evaluating the in-plane stresses of the main text.
E.1. Comparison with linear dispersion relation in a simple case

We will briefly consider the case of vertically-translated interfaces, no-slip and no-penetration boundary conditions, and constant APF throughout the film, with erosion and instantaneous amorphization neglected, which was the focus of an earlier full-spectrum linear stability study [32]. From the equations derived above, we may solve for $\tilde{u}$ and $\tilde{v}$ up to $O(\epsilon^2)$ and substitute both into (6.29), reverse the scalings we have applied, express the $D_{ij}$ terms as their explicit values, and write

$$\eta h_t + \frac{\partial}{\partial x} I_1 + \frac{\partial}{\partial y} I_2 = 0$$

(E.1)

to obtain the evolution equation, where $I_1$ and $I_2$ are the “unscaled” versions of the integrals from (6.29). Carrying out this integration, we have:
\[ I_1 = 3fA\eta\sin(\theta)\cos(\theta)h_x + \gamma h_{yy}h_x + \frac{\gamma}{3} h_{xyy} + \gamma h_xh_{xx} + \frac{\gamma}{3} h_{xxx} \]
\[ + \frac{1}{120}(-40\gamma h_y^2 h_x + 65\gamma h_{yyyy}h_x - 540fA\eta\sin(2\theta)h_x^3 \]
\[ +720fA\eta\cos(2\theta)h_x^3 + 245\gamma h_{yyyy}h_x - 80\gamma h_x^2h_{xy} \]
\[ -285fA\eta h_{xyy} - 435fA\eta\cos(2\theta)h_{xyy} - 160\gamma h_x^2h_{xyy} + 72\gamma h_{yyyyy} \]
\[ -540fA\eta\sin(2\theta)h_{xx} + 1080fA\eta\cos(2\theta)h_xh_{xx} \]
\[ +120\gamma h_x^3h_{xx} + 660\gamma h_xh_{xyy}h_{xx} - 440\gamma h_xh_x^2 + 245\gamma h_xh_{xx} + 305\gamma h_xh_{xyyy} \]
\[ -720fA\eta\cos(2\theta)h_{xxx} - 200\gamma h_x^2h_{xxx} \]
\[ +660\gamma h_xh_{xxx} - 5h_y(24fA\eta\sin(2\theta) + 24\gamma h_x^3 - 83\gamma h_{xyy}) \]
\[ +h_x\{12fA\eta(1 - 3\cos(2\theta)) + 80\gamma h_{xx}\}) - 20h_x^2\{9fA\eta\sin(2\theta) \]
\[ +8\gamma h_{xyy} + h_x(-9fA\eta\{1 + 3\cos(2\theta)\} + 6\gamma h_{yy} \]
\[ +6\gamma h_{xx} + 6\gamma h_{xxx}\} - 5h_y(8\gamma h_{yyyy}h_x + h_{xy}\{-12fA\eta(7 + 9\cos(2\theta)) \]
\[ +80\gamma h_{yy} + 80\gamma h_{xx} \} + \gamma\{-35h_{yyyy} + 24h_xh_{xyy} \]
\[ -35h_{xyy}\}) + 144\gamma h_{xxxyy} + 240\gamma h_xh_{xxxx} + 72\gamma h_{xxxx} \]
We apply the linear stability ansatz and expand in the normal modes simultaneously. Hence
which we seek to reproduce as verification the present lubrication result in a simple case.

Expanding each term about \( Q \)

\[
I_2 = \frac{1}{120}(72\gamma h_{yyyy} - 420fA\eta \sin(2\theta)h_{xy} + 60fA\eta h_x h_{xy} + 900fA\eta \cos(2\theta)h_x h_{xy} \\
- 400\gamma h_{yy} h_x h_{xy} + 245\gamma h_{xy} h_{yy} + 175\gamma h_x h_{yyyy} - 400\gamma h_x h_x h_{xx} \\
- 5h_{yyyy}(72fA\eta - 8\gamma + 72fA\eta \cos(2\theta) - 132\gamma h_{yy} + 24\gamma h_x^2 - 83\gamma h_{xx}) \\
+ 60h_y^4\{6fA\eta \cos^2(\theta) - 2\gamma h_{yy} - 2\gamma h_{xx}\} + 120h_y^3\{9fA\eta \cos^2(\theta) - 2\gamma h_{yy} - 2\gamma h_{xx}\} \\
- 75fA\eta h_{xxxx} + 40\gamma h_{xy} - 645fA\eta \cos(2\theta)h_{xy} + 660\gamma h_{yy} h_{xy} - 160\gamma h_x^2 h_{xy} \\
+ 415\gamma h_x h_{xx} h_{xy} - 20h_y^2\{10\gamma h_{yyy} + 6h_x^2(-3fA\eta \cos^2(\theta) \\
+ \gamma h_{yy} + \gamma h_{xx}) + 8\gamma h_{xyy} + 245\gamma h_{xy} h_{xxx} + 175\gamma h_x h_{xxxx}\} \\
+ 5h_y\{-3fA\eta - 36fA\eta \cos(2\theta) - 88\gamma h_{yy}^2 + 48\gamma h_{yyyy}\} \\
- 72fA\eta \sin(2\theta)h_x + 72fA h_x^2 + 144fA \eta \cos(2\theta) h_x^2 - 16\gamma h_x^2 h_{xx} - 24\gamma h_x h_{xy} + 24fA \eta h_{xx} + 24\gamma h_{xx} - 8\gamma h_{xx}^2 \\
+ 4h_{yy}(27fA \eta + 6\gamma + 27fA \eta \cos(2\theta) - 12\gamma h_x^2 - 20\gamma h_{xx}) \\
+ 61\gamma h_{xyy} - 8\gamma h_x h_{xx} + 12\gamma h_{xxxx} + 72\gamma h_{xxxxy}\}
\]

Substituting the expressions for \( I_1 \) and \( I_2 \) into (E.1), we obtain our evolution equation (fill it in later), which is a very lengthy sixth order nonlinear PDE. Having obtained this equation, we wish to reproduce the long-wave limit of the linear dispersion relation [32] whose full form is

\[
\sigma(k_1, k_2) = -6fA \frac{\cos(2\theta)(h_0 k_1^2) + \cos^2(\theta)(h_0 k_2^2)}{1 + 2Q^2 + \cosh(2Q)} - 3fA \sin(2\theta) i(h_0 k_1) - 3fA \sin(2\theta) i(h_0 k_1) \times \left\{ \frac{2 \cosh(Q) [Q^2 + \sinh^2(Q)]}{1 + 2Q^2 + \cosh(2Q)} - \cosh(Q) \right\} - \frac{\gamma}{2\eta h_0} \frac{Q(\sinh(2Q) - 2Q)}{1 + 2Q^2 + \cosh(2Q)}
\]

Expanding each term about \( Q \approx 0 \) and retaining the first nonzero leads to

\[
\sigma(k_1, k_2) \approx -3fA[\cos(2\theta)(k_1 h_0)^2 + \cos^2(\theta)(k_2 h_0)^2] - \frac{9}{2} fA \sin(2\theta) i(k_1 h_0)Q^2 - \frac{\gamma}{3\eta h_0} Q^4, \quad (E.2)
\]

which we seek to reproduce as verification the present lubrication result in a simple case.

We apply the linear stability ansatz and expand in the normal modes simultaneously. Hence we take

\[
h \to h_0 + h_1 \exp(\sigma t + i(k_1 x + k_2 y))
\]

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and collect the $O(h^1)$ terms. Finally, we obtain growth rate

$$\sigma(k_1, k_2) = -\frac{3}{2} f A(k_2 h_0)^2 + 3 f A(k_1 h_0)^2 (k_2 h_0)^2 + 3 f A(k_2 h_0)^4$$

$$- \frac{\gamma}{3 \eta h_0} (k_1 h_0)^4 + \frac{3}{5 \eta h_0} \gamma (k_1 h_0) - \frac{2}{3 \eta h_0} \gamma (k_1 h_0)^2 (k_2 h_0)^2$$

$$+ \frac{9}{5 \eta h_0} (k_1 h_0)^4 (k_2 h_0)^2 \gamma - \frac{1}{3 \eta h_0} (k_2 h_0)^4 \gamma + \frac{9}{5 \eta h_0} (k_1 h_0)^2 (k_2 h_0)^4 \gamma$$

$$+ \frac{3}{5 \eta h_0} (k_2 h_0)^6 \gamma - 3 f A(k_1 h_0)^2 \cos^2(\theta)$$

$$+ 6 f A(k_1 h_0)^4 \cos^2(\theta) - \frac{3}{2} f A(k_2 h_0)^2 \cos^2(\theta) + 9 f A(k_1 h_0)^2 (h_2 h_0)^2 \cos^2(\theta)$$

$$+ 3 f A(k_2 h_0)^4 \cos^2(\theta) - \frac{9 f A}{2} i(k_1 h_0)^3 \sin(2\theta)$$

$$- \frac{9 f A}{2} i(k_1 h_0)(k_2 h_0)^2 \sin(2\theta) + 3 f A(k_1 h_0)^2 \sin^2(\theta) - 6 f A(k_1 h_0)^4 \sin^2(\theta) + \frac{3}{2} f A(k_2 h_0)^2 \sin^2(\theta)$$

$$- 9 f A(k_1 h_0)^2 (k_2 h_0)^2 \sin^2(\theta) - 3 f A(k_2 h_0)^4 \sin^2(\theta)$$

$$= -3 f A[\cos(2\theta)(k_1 h_0)^2 + \cos^2(\theta)(k_2 h_0)^2] - \frac{9}{2} f A \sin(2\theta)i(k_1 h_0)Q^2 - \frac{\gamma}{3 \eta h_0} Q^4$$

$$+ \frac{3}{5} Q^2 \{ \frac{\gamma}{\eta h_0} Q^4 + 5 f A[Q^2 - 2(k_1 h_0)^2] + 10 f A \cos^2(\theta)Q^2 + \cos(2\theta)[10 f A Q^2 - \frac{15 f A(k_2 h_0)^2}{2}] \}$$

$$= -3 f A[\cos(2\theta)\{(k_1 h_0)^2 + O(Q^4)\} + \cos^2(\theta)\{(k_2 h_0)^2 + O(Q^4)\}]$$

$$- \frac{9}{2} f A \sin(2\theta)i(k_1 h_0)Q^2 - \frac{\gamma}{3 \eta h_0} (Q^4 + O(Q^6))$$

and it is clear that on an effect-per-effect basis, the error term versus (E.2) is always at least two orders higher than leading-order in $Q$; we have grouped the terms to reflect this. Hence the present family of nonlinear evolution equations is consistent with previous linear stability work.

E.2. Details of numerical method

For completeness, we briefly recount the details of the Crank-Nicolson method. We consider

$$H_t = F(H, H_X, H_{XX}, ...) \quad (E.3)$$
and discretize as
\[
\frac{H^{n+1} - H^n}{\Delta t} = \frac{1}{2} \{F^{n+1}(\cdot) + F^n(\cdot)\}
\]  
(E.4)

where we adopt the convention that the superscript denotes time step (in increments of \(\Delta t\)) and we discretize \(H_X, H_{XX}, \ldots\) by the typical second-order central finite differences. We note that the Crank-Nicolson method is an implicit method: the expression for \(H^{n+1}\) involves both itself and the value(s) at the previous time step, \(H^n\). We employ Newton’s method to solve for \(H^{n+1}\) iteratively at each time step; hence we reframe (E.4) as a function \(R\) of \(H^{n+1}\) whose roots are to be computed. Therefore
\[
R(H^{n+1}) = H^n - H^{n+1} + \frac{\Delta t}{2} \{F^{n+1}(\cdot) + F^n(\cdot)\}
\]  
(E.5)

and Newton’s method gives
\[
H^{n+1}_{(p+1)} = H^{n+1}_{(p)} - \left\{ \nabla R(H^{n+1}_{(p)}) \right\}^{-1} R(H^{n+1}_{(p)})
\]  
(E.6)

as the iteration formula for finding the roots, where the subscript denotes the iteration step, and we seed the iteration by
\[
H^{n+1}_{(0)} := H^n,
\]  
(E.7)

which is to say that the initial guess for the next time-step is taken to be the already-known values at the current time-step. When the error between iterations reaches below some tolerance, we accept that iteration as the solution for \(H^{n+1}\).

Ideally, we would use an analytical expression for \(\nabla R(H^{n+1}_{(p)})\); however, the expressions that result from this calculation for the nonlinear equations of Chapter 6 are extremely cumbersome. Instead, we assemble the Jacobian column-by-column, with the \(j^{th}\) column
\[
\left( \nabla R(H^{n+1}_{(p)}) \right)_j = \frac{\frac{1}{12} R(H^{n+1}_{(p)} - 2\epsilon \delta_j) - \frac{2}{3} R(H^{n+1}_{(p)} - \epsilon \delta_j) + \frac{2}{3} R(H^{n+1}_{(p)} + \epsilon \delta_j) - \frac{1}{12} R(H^{n+1}_{(p)} + 2\epsilon \delta_j)}{\epsilon},
\]  
(E.8)

and \(\delta_j\) is simply a column vector of zeros, except for a 1 in its \(j^{th}\) row. This approximation is fourth-order accurate, so that the overall error is dominated by discretization error rather than the approximation of the Jacobian.
Appendix F

Full-spectrum dispersion relation for APF with arbitrary lower interface

An immediate target for future work is in re-working the fitting procedure used in [28] to include (1) our recent developments on the amorphous-crystalline interface, (2) the phase-change term \( V(\theta) \frac{h_0(\theta)}{h_0(\theta)} \left( 1 - \frac{\rho_a}{\rho_c} \right) \) and (3) isotropic swelling. For comparison with GISAXS data, we require full-spectrum dispersion relations. While the work of [48] is presented with \( \tilde{g}_1 \) ready to have a BCA-informed interface relation inserted, this is not the case for [32], where only the “vertical interfaces” case was considered. Here, we obtain the full-spectrum, arbitrary-interfaces dispersion relation for APF. We anticipate that it will be useful in future work, particularly since it appears (due to the Chapter on spatially-resolved IIS) that spatially-resolved APF offers no benefit in seeking agreement between theory and experiment. Although not shown here, we have also verified that its long-wave expansion, when restricted to the \( x \)-axis, agrees with the forms obtained by expanding in the long-wave limit prior to solving for \( \sigma(k; \theta) \).

Generalization of anisotropic plastic flow. Proceeding from the work shown in the Appendix of [32] and taking the crystalline-amorphous interface to be arbitrary \( z = g_1(x, y) \) instead of the vertical-translation assumption \( z = h_1(x, y) \), we may “recycle” almost everything. This is greatly facilitated by the appearance of the lower interface in exactly one location: the no-slip and no-penetration interface conditions, which have been combined into a single expression. We will briefly summarize the linearization, which can be read in full detail in [32], and then provide the (admittedly slight) extension to it.

For the convenience of the reader, we summarize the final stages of the analysis verbatim before deviating from them at a point that we will indicate. Following linearization, we have
conservation of momentum and mass yield, respectively

\[-ik_1 p_1(z) + \eta \{ -(k_1^2 + k_2^2) u_1 + u_1'' \} = 0 \]
\[-ik_2 p_1(z) + \eta \{ -(k_1^2 + k_2^2) v_1 + v_1'' \} = 0 \]
\[p_1'(z) + \eta \{ -(k_1^2 + k_2^2) w_1 + w_1'' \} = 0 \]
\[ik_1 u_1 + ik_2 v_1 + w_1' = 0 \]

whose solutions are, in general,

\[p_1(z) = -2\eta [(RH + ik_1 C + ik_2 E) \cosh(Rz) + (RG + ik_1 D + ik_2 F) \sinh(Rz)]; \]
\[u_1(z) = C \cosh(Rz) + D \sinh(Rz) - \frac{ik_1}{R} [(RG + ik_1 D + ik_2 F) z \cosh(Rz) + (RH + ik_1 + ik_2 E) z \sinh(Rz)]; \]
\[v_1(z) = E \cosh(Rz) + F \sinh(Rz) - \frac{ik_2}{R} [(RG + ik_1 D + ik_2 F) z \cosh(Rz) + (RH + ik_1 + ik_2 E) z \sinh(Rz)]; \]
\[w_1(z) = G \cosh(Rz) + H \sinh(Rz) - [(RG + ik_1 D + ik_2 F) z \sinh(Rz) + (RH + ik_1 + ik_2 E) z \cosh(Rz)].\]

where \( R = \sqrt{k_1^2 + k_2^2} \) and \( C,D,E,F,G,H \) are integration constants. Applying the combined no-slip and no-penetration condition at the lower interface yields

\[v_1(x,y,0) + \frac{\partial v_0}{\partial z}(x,y,0) g_1 = 0, \]

implying

\[C = -3fA \sin(2\theta) g_1(x,y), \quad E = 0, \quad G = 0.\]

The coefficient \( C \) is the only place where we depart from the previous work of [32]; both \( E \) and \( G \) remain zero. The influence of the arbitrary lower interface \( g_1 \) will pass into the pressure and velocity fields, and, from there, into the stress balance at the upper interface and the kinematic condition, at which point it will appear in the dispersion relation. With this modification, we consider the stress balance at the upper interface. That is,

\[T \cdot \hat{n} = -\gamma \kappa \hat{n} \]
at \( z = h_0 + \epsilon h_1(x, y) \). Although we intend to expand in the normal modes later, we will suppress this expansion for \( h_1(x, y) \) and \( g_1(x, y) \) until the end; this delay will prove useful. To facilitate the calculation, we first formally linearize the above equation, leading to

\[
T_0 \cdot \hat{n}_1 + T_1 \cdot \hat{n}_0 = -\gamma \kappa_1 \hat{n}_0, \tag{F.6}
\]

at \( z = h_0 \). Here, we have

\[
\kappa_1 = -(h_{1,yy} + h_{1,xx});
\]

\[
\hat{n}_0 = <0, 0, 1 >;
\]

\[
\hat{n}_1 = <-h_{1,x}, -h_{1,y}, 0 >;
\]

\[
T_0 = -6\eta f A \begin{bmatrix} \cos(2\theta) & 0 & 0 \\ 0 & \cos^2(\theta) & 0 \\ 0 & 0 & 0 \end{bmatrix};
\]

\[
T_1 = \begin{bmatrix} -p_1 + 2\eta u_{1,x} & \eta(u_{1,y} + v_{1,x}) & \eta(u_{1,z} + w_{1,x}) \\ \eta(v_{1,x} + u_{1,y}) & -p_1 + 2\eta v_{1,y} & \eta(v_{1,z} + w_{1,y}) \\ \eta(w_{1,x} + u_{1,z}) & \eta(w_{1,y} + v_{1,z}) & -p_1 + 2\eta w_{1,z} \end{bmatrix}.
\]

We will “recycle” much of the previous work by making a sequence of useful observations: \( T_1 \cdot \hat{n}_0 \) merely selects the third column of \( T_1 \), and within this column, the coefficient \( C \) is never differentiated with respect to \( x \) or \( y \), because \( C \) appears only in \( u_1 \) and \( v_1 \). Then the \( \bar{\alpha}, \bar{\beta}, \bar{\gamma} \) terms, (A13) in [32], are unchanged except for \( h_1 \to g_1 \) in the term involving \( \sin(2\theta) \) on the third column vector. These constants serve the same role in the solution as originally, with coefficients \( D, F \) and \( H \) solved in terms of them. Ultimately, \( D, F \) and \( H \) alter \( w_1(h_0) \), but \( u_0(h_0) \) and \( v_0(h_0) \) are, of course, unchanged. Then we construct a matrix equation for the stress balance at the upper interface as

\[
\begin{bmatrix} R\bar{C} + \frac{k^2}{R}(\bar{C} + 2Q\bar{S}) & \frac{k_1 k_2}{R}(\bar{C} + 2Q\bar{S}) & -2ik_1 Q\bar{C} \\ \frac{k_1 k_2}{R}(\bar{C} + 2Q\bar{S}) & R\bar{C} + \frac{k^2}{R}(\bar{C} + 2Q\bar{S}) & -2ik_2 Q\bar{C} \\ -2ik_1 Q\bar{C} & -2ik_2 Q\bar{C} & 2R(\bar{C} - Q\bar{S}) \end{bmatrix} \begin{bmatrix} D \\ F \\ H \end{bmatrix} = \begin{bmatrix} \bar{\alpha} \\ \bar{\beta} \\ \bar{\gamma} \end{bmatrix}, \tag{F.8}
\]
where $Q = h_0 R$, $C = \cosh(Q)$, $\bar{S} = \sinh(Q)$, and 

$$
\begin{bmatrix}
\bar{\alpha} \\
\bar{\beta} \\
\bar{\gamma}
\end{bmatrix} = -\gamma h_1 
\begin{bmatrix}
0 \\
0 \\
\frac{R^2}{2}
\end{bmatrix} 
+ 6\eta f A h_1 
\begin{bmatrix}
\frac{i k_1 \cos(2\theta)}{2} \\
\frac{i k_2 \cos^2(\theta)}{2} \\
0
\end{bmatrix} 
+ 3\eta f A g_1 \sin(2\theta) 
\begin{bmatrix}
\frac{k_1 k_2}{2} (\bar{S} + 2Q\bar{C}) \\
-k_1 Q \sinh(Q) \\
-2ik_1 \bar{Q} \sinh(Q)
\end{bmatrix} 
+ \frac{k_1}{R} (\bar{S} + 2Q\bar{C}) 
$$

(F.9)

from which we may solve for $D, F$ and $H$. From the kinematic condition at the upper interface, linearized as

$$
h_{1,t} = w_1(h_0) - u_0(h_0) h_{1,x} - v_0(h_0) h_{1,y},
$$

(F.10)

where

$$
\begin{align*}
\bar{u}_0(h_0) &= 3f A \sin(2\theta) h_0; \\
\bar{v}_0(h_0) &= 0, \\
\bar{w}_1(h_0) &= -6f A \bar{h}_1 \left( \frac{\cos(2\theta)(h_0 k_1)^2 + \cos^2(\theta)(h_0 k_2)^2}{1 + 2Q^2 + \cosh(2Q)} \right) \\
&+ 3f A g_1 \sin(2\theta) (i h_0 k_1) \left( \frac{2 \cosh(Q) [Q^2 + \sinh^2(Q)]}{1 + 2Q^2 + \cosh(2Q)} - \cosh(Q) \right) \\
&- \frac{\gamma \bar{h}_1}{2\eta h_0} \frac{Q \sin(2Q) - 2Q}{1 + 2Q^2 + \cosh(2Q)}
\end{align*}
$$

(F.11)

we obtain the dispersion relation, which will change only very subtly. This is quite intuitive when we note that the only term that can possibly change is the $w_1$ term, because the $u_0$ and $v_0$ terms were determined prior to the perturbation of the free interface, where the steady state for $h$ is $h_0$ and the steady state for $g$ is 0. Due to having retained general $g_1(x, y)$, some of the coefficients of terms in the expression for $w_1$ will now have $g_1$ where they originally had $h_1$. In order to isolate dispersion relation $\sigma$, divide through by $\bar{h}_1$, we obtain the generalization of the original result,

$$
\sigma(k_1, k_2) = -6f A \left( \frac{\cos(2\theta)(h_0 k_1)^2 + \cos^2(\theta)(h_0 k_2)^2}{1 + 2Q^2 + \cosh(2Q)} \right) - 3f A \sin(2\theta) (i h_0 k_1) \\
-3f A \sin(2\theta) (i h_0 k_1) \times \frac{g_1}{h_1} \left( \frac{2 \cosh(Q) [Q^2 + \sinh^2(Q)]}{1 + 2Q^2 + \cosh(2Q)} - \cosh(Q) \right) \\
- \frac{\gamma}{2\eta h_0} \frac{Q \sin(2Q) - 2Q}{1 + 2Q^2 + \cosh(2Q)}
$$

(F.12)

where $\frac{g_1}{h_1}$ is the ratio of the expansions of the perturbed lower and upper interfaces in normal modes, as in the main text. We note the agreement with the physical interpretation given
in the original analysis of [32], which describes the third term as associated with the effect of the nonplanar lower interface under the influence of the beam.
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