Spatial-Temporal Stability Analysis of Faceted Growth with Application to Horizontal Ribbon Growth

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Abstract

Spatial-temporal stability analysis has been applied to a solidification model that includes both isotropic and non-isotropic kinetics. In agreement with previous temporal stability analyses, it was shown that the kinetics associated with the propagation of steps across a facet can stabilize solidification processes that would normally be thermally unstable. In cases where the solidification is unstable, it was also shown that pulling the solid with a tangential velocity can cause a transition from “absolute” instability where perturbations cause growth at all locations to “convective” instability where a perturbation grows as it propagates, but at any fixed location disturbances decay away after the perturbation passes. These results were applied to understand instabilities in the floating silicon method (FSM), which is a particular type of horizontal ribbon growth. It was shown that increasing pull-speeds in FSM lead to increasingly unstable thermal growth conditions, but the combination of the kinetics of faceted growth and the tangential pull velocity can stabilize the process.

As the pull speed increases, however, the process becomes increasingly sensitive to perturbation.

Key words: A1 solidification, A1 morphological stability, A2 growth from melt, A2 Single crystal growth, A2 horizontal ribbon growth

1 Introduction

Recently, we have reported experimental and numerical studies [1, 2] of the production of single-crystal silicon sheets using the floating silicon method (FSM). FSM is a form of horizontal ribbon growth where the solid silicon sheet floats on molten silicon before being removed from the furnace. A schematic of the FSM process is shown in Fig. 1. The solid sheet of silicon grows because of heat removal caused by a helium jet, and then floats on the melt surface before being removed from the furnace. One of the interesting findings from our previous studies was that there was a $\{111\}$ facet at the growing edge of the sheet and that solidification kinetics must be considered to predict the

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horizontal position of the growth front as a function of pull speed. Previous studies of horizontal ribbon growth did not consider these effects [3–14]. In [1], it was shown that facet kinetics limit the maximum growth rate attainable by FSM. This limitation manifested itself in the numerical results as a turning point in the response of the growth front position versus the pull speed; beyond a limiting pull speed no steady solutions could be found. Although the growth front position predicted numerically agreed well with the experiment, the numerically predicted pull speed limits were often higher than those attained in the experiment. In the experiment, a transition to dendritic growth was often observed before reaching the predicted pull speed limit.

The goal of this paper is to perform a stability analysis of solidification including solidification kinetics and apply the results to understand the transition to dendritic growth seen in the FSM experiments. The most relevant previous stability analyses are those of Coriell, Murray, and Chernov [15, 16] who analyzed the solidification of binary alloys [15] and pure substances [16] including step propagation kinetics. They showed that for growth on a vicinal plane, step propagation can stabilize growth conditions that would normally be thermally unstable. In this work, we extend their analysis by including the effect of a tangential solid motion on the growth process. To understand this effect, spatial-temporal stability analysis is used [17, 18]. Not to be confused with a spatial-temporal instability such as cellular growth, spatial-temporal stability analysis predicts the system response to a *localized* disturbance. Using the spatial-temporal stability analysis algorithm described in [18], unstable growth conditions are classified as either “absolutely” or “convectively” unstable based on this response. Absolute instability corresponds to the case where the disturbance amplitude at any fixed location grows in time. Convective in-

stability corresponds to growth of a disturbance as it propagates, but decay at any fixed location after the initial disturbance passes. To our knowledge, this is the first time this type of analysis has been applied to solidification. The distinction turns out to be critical for understanding FSM because there is a velocity tangential to the growth direction that causes disturbances to propagate as they grow.

2 Formulation

This is a spatial-temporal stability analysis of the faceted growth of silicon at some misalignment angle from the $[111]$ direction. The basic problem is the same as that analyzed in [15] except that segregation effects are not included; only pure substances are considered and a uniform tangential velocity, u , is imposed on both the solid and liquid. A schematic of the assumed geometry and boundary conditions is shown in Fig. 2. The misalignment angle between the vertical propagation direction and the $[111]$ direction is θ_0 . The wavelength of the perturbation, λ , is used to define a perturbation wavenumber, $k = 2\pi/\lambda$. The perturbation itself is assumed to be small enough in amplitude such that the misalignment angle is bounded away from zero. The height of the domain is given by L , which is typically assumed to be much larger than λ and will be taken to the limit $L \rightarrow \pm\infty$. At the bottom of the domain, liquid is assumed to enter with a velocity v such that the unperturbed solidification front remains stationary even though liquid is being solidified. The total energy flux in and out of the domain is specified as f_s and f_l respectively where the subscripts s and l indicate solid and liquid in all of the following. The domain is assumed to be infinite in the horizontal direction because spatial-temporal stability

analysis relies on the continuous Fourier transform.

Looking ahead to the FSM analysis, the schematic shown in Fig. 2 is meant to represent a localized region in a coordinate system aligned with the solidification front. In the application to FSM, this analysis has some inherent assumptions. Because the domain is assumed infinite in the x -direction, only periodic temperature perturbations in the horizontal direction can be analyzed. In the FSM simulations, the temperature gradients normal to the growth front are generally significantly larger than those tangential to the front so this assumption is reasonable. Another assumption is that the liquid flow is uniform. This is also not a restrictive assumption. In the FSM simulations, a replenishment flow was added such that both the solid and liquid were moving horizontally at a similar velocity. In some cases, Marangoni effects were included which caused non-uniform flows, but this did not significantly change the numerical results. This indicates that non-uniform flow effects are of secondary importance.

3 Governing Equations

The liquid and solid are assumed to be incompressible with equal density, ρ , and specific heat, c . In this case, mass conservation ensures that the velocity of the solid and liquid are everywhere v in the vertical direction and u in the horizontal direction. The equations governing the temperature, T , in the solid and liquid are then given by

$$\frac{\partial \rho c T}{\partial t} + \frac{\partial}{\partial x} \left(\rho c u T - k_i \frac{\partial T}{\partial x} \right) + \frac{\partial}{\partial y} \left(\rho c v T - k_i \frac{\partial T}{\partial y} \right) = 0 \quad (1)$$

where k_i is the thermal conductivity and $i = s, l$. Note that k is the wavenumber, while k_s and k_l are thermal conductivities.

The boundary conditions at the bottom and top of the domain are given by

$$\left(\rho c v T - k_i \frac{\partial T}{\partial y} \right) \Big|_{y=L_i} = f_i \quad (2)$$

where for convenience we define $L_l = -L$ and $L_s = L$.

At the interface between the solid and liquid, conservation of energy gives

$$\llbracket -k_i \nabla T \cdot \vec{n} \rrbracket = \rho L_f v_g \quad (3)$$

where double brackets indicate a jump in a quantity across the interface. The normal to the interface, $\vec{n}$, is taken to point from the liquid towards the solid and jumps are defined as the solid quantity minus the liquid quantity. The right side of the equation is the energy released by solidification. L_f is the heat of fusion of the material, and v_g is the solidification velocity. For the configuration given in Fig. 2, v_g is given by $(u\vec{i} + (v - h_t)\vec{j}) \cdot \vec{n}$ where $h(x)$ is the height of the interface relative to $y = 0$, and h_t is the vertical velocity of the interface. In the following, subscripts of t and x denote differentiation with respect to that variable. The temperature at the interface is assumed to be continuous such that

$$\llbracket T \rrbracket = 0. \quad (4)$$

Lastly, it is assumed that the interface propagates according to the kinetic solidification model of Weinstein and Brandon [19]. This model includes mechanisms for nucleation of atoms onto a facet, step propagation along a facet, and roughened growth. The model constants are all taken from [19]. The model relates the supercooling to the growth velocity as

$$T_e - T = K v_g \quad (5)$$

where all variables are evaluated on the interface, T_e is the equilibrium melting

temperature, and K is the inverse of the more typically used kinetic coefficient β which relates the growth velocity to the amount of supercooling. This less standard form was chosen because it allows kinetic effects to be eliminated by setting $K = 0$ rather than setting β to infinity.

The kinetic constant K is evaluated based on the rates of the individual 2D nucleation, step propagation, and roughened growth mechanisms. The kinetic coefficients of these mechanisms are combined using the following expression

$$K = (K_r^p + K_{min}^p)^{1/p} \quad (6)$$

where K_r is the kinetic constant associated with roughened growth given by 79.4 Ks/m ($\beta_r = 0.0126 \text{ m/(sK)}$). $p \in [1, \infty)$ is a modeling constant which creates a smooth transition between K_{min} (discussed next) and K_r . When $p = 1$ the sum of the K_r and K_{min} is used whereas when $p = \infty$ the maximum is used. The FSM simulations used a value of $p = 2$, but for the analysis it was simpler to change the value to 1. The choice of p only makes a difference when the magnitudes of K_{min} and K_r are similar and does not have any significant effect on the conclusions drawn below.

K_{min} is given by

$$K_{min} = \min \left(K_{2Dn}, \frac{K_{st}}{|\sin(\theta)|} \right). \quad (7)$$

This expression chooses the process that requires less supercooling to define K_{min} between 2D nucleation and step propagation. The value used for K_{st} was 1.59 Ks/m ($\beta_{st} = 0.63 \text{ m/(sK)}$) and θ is the local misalignment angle. K_{2Dn} models the 2D nucleation effect and is given by

$$K_{2Dn} = B^{-1} \exp \left(\frac{A}{\Delta T} \right). \quad (8)$$

where $A = 140 \text{ K}$, and $B = 1.5 \times 10^{10} \text{ m/(sK)}$.

For the stability analysis only the roughened growth and step propagation mechanisms affected the results. The mathematical reason for this is that in all of the conditions analyzed $K_{st}/|\sin(\theta)|$ was smaller than K_{2Dn} . The physical reason is that in most faceted growth processes, the nucleation of atoms on a [111] surface is typically a localized phenomenon (this is true for the FSM simulations discussed later in this paper) and does not occur over a large enough region to make planar stability analysis relevant [20]. Nucleation is needed to create the steps which then propagate across the facet. Because the analysis is on an infinite domain, however, a nucleation point is not necessary. A cartoon of the atomistic layer growth explaining this is shown in Fig. 3. In the cartoon, there are only three steps and each step propagates with velocity of v_{st} . Because the domain is assumed infinite, there is no need to include a step nucleation point on the right side of the domain; there are an infinite number of steps coming from $x = \infty$. The step propagation leads to a net downward growth of the solid with an apparent misalignment angle of θ_0 , which is the situation analyzed. For large misalignment angles, roughened growth dominates step propagation so both effects are included.

Given that $p = 1$ in the analysis and there is no nucleation point for the steps, the kinetic relation can be simplified to

$$T_e - T = \left(\frac{K_{st}}{|\sin(\theta)|} + K_r \right) v_g \quad (9)$$

Except for the inclusion of the roughened growth term, this model is essentially equivalent to that analyzed in [15].

The surface energy effect on the equilibrium melting temperature is given by

$$T_e - T_m = -\sigma\kappa \quad (10)$$

where $\sigma = T_m \Gamma$, Γ is the capillary length, T_m is the melting temperature, and κ is the mean curvature [21].

4 Analysis

The first step of spatial-temporal stability analysis is to derive the dispersion relation that governs the standard linear stability problem. As this has been done previously, only a brief summary of this step is given. In all of the following, variables are non-dimensionalized using $\bar{\alpha}/v$ and $\bar{\alpha}/v^2$ for length and time scales respectively, where $\bar{\alpha}$ is the average thermal diffusivity, $(\alpha_s + \alpha_l)/2$ with $\alpha_i = k_i/(\rho c)$. Temperatures and heat fluxes are nondimensionalized by L_f/c and $\rho L_f v$ respectively.

To start, the temperature in the solid and liquid and the interface position are expanded as

$$T_i = T_{i,0}(y) + \epsilon T_{i,1}(x, y, t) \quad (11)$$

and

$$h = \epsilon h_1(x, t) \quad (12)$$

which results in a leading order solution where the interface is perfectly horizontal. Assuming the interface is propagating vertically downward with a growth velocity of v , the leading order interface temperature, $T_{y=0}$, is determined by eq. 9 as

$$T_{y=0} = T_m - K_e v \quad (13)$$

where K_e is the effective kinetic coefficient given by $(K_{st}/\sin(\theta_0) + K_r)$.

In eq. 1 only the derivatives in the vertical direction matter for the leading

order solution, so this ordinary differential equation can be solved to obtain

$$T_{i,0} = T_{y=0} + q_i (1 - e^{y/\alpha_i}) \quad (14)$$

where q_i is the nondimensional diffusive flux at the interface (in dimensional terms it is given by $(f_i - \rho c T_{y=0})$). Note that the temperature exponentially grows as y goes to infinity. One can verify that in spite of this growth, the convective and diffusive energy flux balance to give a constant vertical heat flux as required by eq. 1. Also note that based on the specified non-dimensionalization, α_i is now the ratio of the dimensional thermal diffusivity to the mean thermal diffusivity.

Conservation of energy at the interface gives a relation between the heat fluxes in and out of the domain and the growth rate of the solid. As heat fluxes are non-dimensionalized by the latent heat release, in dimensionless terms this becomes $q_s - q_l = 1$.

At the next order of the solution, we assume the solution has the form

$$T_{i,1} = T_{i,1}(y)e^{ikx+st} \quad (15)$$

and

$$h_1(x, t) = h_1 e^{ikx+st}. \quad (16)$$

This is actually equivalent to taking the Fourier and Laplace transforms which is the formal procedure used for spatial-temporal stability analysis.

Linearizing the governing equations and boundary conditions at the next or-

der, one can finally obtain the following dispersion relation

$$0 = \left[\frac{q_s}{\alpha_s} - \frac{q_l}{\alpha_l} + \tilde{s} - \left(-\frac{K_{st} \cot(\theta_0)}{\sin(\theta_0)} ik + K_e \tilde{s} + k^2 \sigma + \frac{q_s}{\alpha_s} \right) \frac{1 + \lambda_{s,-}}{2} + \left(-\frac{K_{st} \cot(\theta_0)}{\sin(\theta_0)} ik + K_e \tilde{s} + k^2 \sigma + \frac{q_l}{\alpha_l} \right) \frac{1 + \lambda_{l,+}}{2} \right]. \quad (17)$$

where $\tilde{s} = s + iku$. s always appears in this grouping because of the rigid translation caused by the tangential velocity. $\lambda_{i,\pm}$ is given by

$$\lambda_{i,\pm} = \pm \sqrt{1 + 4\alpha_i(\tilde{s} + \alpha_i k^2)} \quad (18)$$

where the plus or minus sign is chosen such that $\lambda_{i,+}$ has a positive real component and $\lambda_{i,-}$ has a negative real component. For there to be a nonzero solution, the dispersion relation, which is denoted $D(\tilde{s}, k)$, must be equal to zero, thus for a given wavenumber, k , eq. 17 determines $\tilde{s}$. The real part of $\tilde{s}$ determines the exponential growth rate of a periodic disturbance. This dispersion relation is nearly identical to eq. 26 of [15] except that the above includes the roughened growth term and the translation effect, but does not include segregation effects.

5 Temporal Stability

Before examining the spatial-temporal stability, it is first necessary to determine under what conditions the problem is unstable in the classical sense (temporal stability analysis). Because s always appears in the group $s + iku$, translation does not change the real component of s and thus does not change the results analyzed in [15] and [16]. However in [15] the focus was on the effect of segregation on the stability behavior and in [16], although pure substances

were studied, it was assumed that all the heat was removed through the liquid ($q_s = 0$). For this reason the temporal stability results are reviewed before examining the spatial-temporal results.

Using a large k expansion, $|k| \gg 1$, and assuming that $|\tilde{s}| \ll k^2$ (to be verified later) gives

$$\lambda_{i,\pm} = \pm \sqrt{1 + 4\alpha_i(\tilde{s} + \alpha_i k^2)} \approx \pm 2\alpha_i |k|. \quad (19)$$

The dispersion relation (eq. 17) can then be simplified to obtain a leading order expansion for the growth rate,

$$\tilde{s} \approx - \frac{\left[2 \left(-\frac{K_{st} \cot(\theta_0)}{\sin(\theta_0)} i k + k^2 \sigma \right) + (q_s + q_l) \right] |k|}{1 + 2K_e |k|}. \quad (20)$$

where we have used the fact that $\alpha_s + \alpha_l = 2$ because of the non-dimensionalization.

Ignoring the K_e term in the denominator to get an upper bound for $|\tilde{s}|$, it can be seen that $|\tilde{s}| \ll k^2$ if $|k| \ll 1/\sigma$. For silicon, this corresponds to wavelengths larger than 3.4 nm. Higher wavenumbers approach the molecular scale and thus are not physically relevant. Thus, the above approximation is valid for all physical conditions of interest.

For non-faceted growth, the above agrees with classical results that if $q_s + q_l < 0$ there may be instability. As $q_s - q_l = 1$, this is equivalent to $q_s < 1/2$ i.e. if less than half of the solidification flux is removed through the solid there will be instability and if kinetic effects are ignored, the magnitude of the instability will increase linearly with k until at large values of k , the instability will be damped by the capillary effect. The isotropic kinetics of roughened growth, which are included in the K_e term, alter the growth rates but do not change the stability behavior.

Fig. 4 shows curves of $\text{real}(\tilde{s})$ versus k for the conditions given as Case 1 in

Table 1 and various values of q_s . Case 1 corresponds to a misalignment angle where roughened growth dominates the kinetics. The numerical solutions to the dispersion relation are shown as solid lines and the approximation given by eq. 20 are shown as dashed lines. For this case, the approximate solution is accurate and it is impossible to distinguish between the two results. As can be seen in the figure, when $q_s < 0.5$ the problem is thermally unstable for wavenumbers less than $k = 1000$. For larger wavenumbers, the capillary effect stabilizes the growth.

Case 2 in Table 1 corresponds to faceted growth with a small misalignment angle. Fig. 5 shows the imaginary part of $\tilde{s}$ as predicted by a numerical solution to the dispersion relation and by eq. 20. The same values of q_s were chosen as for Fig 4, but in this case the solution is insensitive to q_s so the individual curves were not labeled. This insensitivity to q_s was observed in [15] as well except for the case of solidification of a binary alloy. The non-zero imaginary part implies that there will be propagation as part of the growth process which was also observed and discussed in [15].

The real component for Case 2 is shown in Fig. 6 again for the same values of q_s as the previous figures. A higher order approximate solution (given in the appendix) is shown as well because the leading order solution does not predict the real part of the growth rate for these conditions. In spite of the fact that several thermally unstable conditions are plotted ($q_s = 0.05, .25, .45$), the growth is stable for all wavenumbers. This is consistent with the analysis of [16] which showed that with $q_s = 0$, the growth of a pure substance was stable at small misalignment angles. The stabilization is caused by step propagation. With $\sigma = 0$, the curve is basically the same except that as $k \rightarrow \infty$ the growth rate approaches zero rather than $-\infty$. From the curve, it is apparent

that there is a wavenumber at which these kinetics have their strongest influence (the local minimum in the curve). Using the approximate solution given in the appendix, it was determined that when $kK_{st}/\sin(\theta_0)$ is $O(1)$, which corresponds to $k = O(128)$, the decay rate transitions from linearly decreasing with k to turn back towards zero. In Fig. 6, the most negative growth rate occurs at $k \approx 270$, which is in reasonable agreement with this estimate. Inserting the material constants for silicon in the order of magnitude estimate gives a prediction for the wavelength of $200 \text{ nm}/\sin(\theta_0)$, which for this case is $100 \text{ }\mu\text{m}$.

6 Spatial-Temporal Instability Analysis

In this section, we examine whether the tangential velocity, u , can change the instability from an absolute instability to a convective instability. As mentioned in the introduction, once a system has been classified as temporally unstable (i.e., periodic disturbances grow exponentially in time), further analysis may be performed to determine if the system is convectively or absolutely unstable with respect to a localized disturbance. Convective instability corresponds to a disturbance that convects away from the location of the initial disturbance as it grows; at the initial location of the disturbance the effects of the perturbation decay. Absolute instability corresponds to exponential growth of the disturbance as it propagates *and* growth at the location of initial disturbance as well [17]. This distinction is important, as a steady process may still be established in the convectively unstable case while this is not possible for an absolutely unstable system.

To determine whether the system is convectively or absolutely unstable, the

spatial-temporal stability analysis algorithm described in [18] is used. To summarize, the prediction of absolute or convective instability is determined by the sign of the absolute growth rate, s_0 , defined as the exponential growth rate of a spatially localized disturbance measured at a fixed location x (i.e. $h(x=0) \sim e^{s_0 t}$). If $\text{real}(s_0) > 0$, the response grows for fixed x and is thus absolutely unstable. If $\text{real}(s_0) < 0$, the response decays for fixed x and is thus convectively unstable. The quantity s_0 is obtained directly from the dispersion relation $D(s+iku, k) = 0$ by first finding a complex wave number k_0 that satisfies $ds/dk=0$. This point is called a saddle point and arises in the process of approximating the long-time behavior of the inverse Fourier transform using the method of steepest descent [18, 22]. Once the saddle point location is known, the absolute growth rate then satisfies $D(s_0+ik_0u, k_0) = 0$. The growth rate can actually be determined along any constant velocity path $x = ct$, but to determine whether a system is absolutely or convectively unstable only the behavior along the path $x = 0$ is needed. The subscript 0 is used to denote that these results are for the $x = 0$ path. See [17, 18] for further details.

As this type of analysis has never been applied to solidification, a simplified case that includes only thermal and capillary effects is examined first. The short-wave dispersion relation (eq. 20) reduces to

$$s \approx -|k| \left[2k^2\sigma + q_s + q_l \right] - iuk. \quad (21)$$

Following the method described above, we determine values of k such that $ds/dk=0$, namely

$$k_0 = \pm \frac{\sqrt{3(iu - q_l - q_s)}}{3\sqrt{2\sigma}}. \quad (22)$$

Substituting $k = k_0$ from above into eq. 21 leads to a rather lengthy expression for s_0 . If the problem is temporally unstable, i.e. $q_s < 1/2$, the real part of s_0

is positive if

$$|u| < \sqrt{3} |q_l + q_s| \quad (\text{absolutely unstable}) \quad (23a)$$

and negative if

$$|u| > \sqrt{3} |q_l + q_s| \quad (\text{convectively unstable}). \quad (23b)$$

The absolute growth rate $\text{real}(s_0)$ is plotted versus u in Fig. 7 for this simplified case. Note that for $u = 0$ i.e. a non-moving solid, the response is always absolutely unstable if $q_s < 1/2$. As one increases the magnitude of u away from zero in Fig. 7, eventually $\text{real}(s_0) < 0$, indicating a transition to convective instability. For smaller values of q_s , this transition from absolute to convective instability occurs at larger values of $|u|$. As a reminder u is nondimensionalized by the growth velocity, so the dimensional velocities for which this occurs are on the order of the growth rate. Thus, one can stabilize a thermally unstable growth process by tangentially pulling the growing solid. In any real configuration, there must be some upstream point where the growth is nucleated for this to be possible. This is exactly the situation in horizontal ribbon growth, which is discussed in the next section.

Although closed-form instability criteria such as eq. 23 are not easily obtained for the full dispersion relation (eq. 17), the behavior qualitatively agrees with the simplified criteria described above. For illustration, a growth versus u plot is shown in Fig. 8 for the parameters of Case 1. In this case, the curve is not symmetric with respect to the tangential velocity u . The reason is that the step propagation along the surface induces a directionality to the growth. It can be shown that tangential velocity that causes the maximum in $\text{real}(s_0)$ is

of the form

$$u_{max} = \frac{K_{st} \cot(\theta_0)}{\sin(\theta_0)} f(\sigma, \alpha_s, \alpha_l, q_s, q_l, K_e), \quad (24)$$

which is only nonzero when $K_{st} \neq 0$.

For a positive misalignment angle, which is the configuration analyzed, the steps propagate in the negative direction (shown in Fig. 3). A small tangential velocity in the opposite direction of the step propagation ($u > 0$) slows the steps down and, as shown by the figure, increases the absolute growth rate. A small tangential velocity in the direction of the steps ($u < 0$) increases the speed of step propagation and decreases the absolute growth rate.

7 Application to Horizontal Ribbon Growth

In [1], we presented numerical simulations of the floating silicon method (FSM). These simulations showed that as the pull speed of the process increased, the temperatures in the liquid decreased and the temperatures in the solid increased, indicating increased heat being removed through the liquid. In this section, the stability analysis is used to understand the implications of this for FSM.

The numerical solidification model is an arbitrary-Lagrangian-Eulerian *hp*-finite element method [23, 24]. The domain for the calculation is shown in Fig. 9, which also shows an initial element mesh used for the calculations. The full specification of the problem and the numerical method are given in [1]. To summarize, the governing equations are the same as used in the stability analysis: In the solid, the governing equation is the energy equation, which includes a convective term because of the translation of the solid with

the horizontal pull speed, denoted u_s . In the liquid, the energy equation is supplemented by the Navier-Stokes equations, which are used to solve for the flow field and pressure. Constant properties were used in the calculations that approximate silicon, with $\rho = 2530 \text{ kg/m}^3$, $c = 1000 \text{ J/(kgK)}$, $k_s = 22 \text{ W/(mK)}$, $k_l = 64 \text{ W/(mK)}$, and $\mu = 7 \times 10^{-4} \text{ kg/(ms)}$ [25] where μ is the liquid viscosity. The equations governing the motion of the interface are also the same as in the stability analysis, namely energy conservation across the interface and the kinetic equation to determine the interface temperature. For the energy conservation (eq. 3), the latent heat, L_f , was taken as $1.8 \times 10^6 \text{ J/kg}$ [25]. The same constants were used for the kinetic equation except, as mentioned earlier, $p = 2$ and the surface energy effect was not included ($\sigma = 0$).

The boundary conditions for the flow were applied as follows. On the left side of the domain shown in Fig. 9, an inflow condition was used given by

$$u = u_s \left(1 - (y/d)^2\right) \quad (25)$$

where u is the horizontal flow velocity. This profile was used to model the replenishment flow. However, the mass flux associated with this profile is slightly larger than that needed for replenishment so an outflow boundary condition was used for the liquid on the right side of the domain. The top surface of the liquid was treated as a slip boundary condition and at the bottom a no slip condition was applied. The flow generated in this way does not have sharp gradients which is consistent with the assumptions of the analysis. More realistic boundary conditions are discussed in [1], but these do not have a significant effect on the behavior of the system.

The thermal boundary conditions are described in detail in [1]. To summarize,

there was a Gaussian-like heat removal profile specified on the top surface, which modeled the effect of a helium jet impinging on the silicon surface. The heat removal profile was centered around $x = 0$, had a half-width on the order of 0.5 mm , and a peak heat flux of nearly 400 W/cm^2 . On the bottom of the domain, there was a broader heat flux into the domain with a magnitude of around 25 W/cm^2 . The inlet temperature was fixed at a few degrees above melting temperature and an adiabatic condition was enforced at the outlet.

7.1 Results

Fig. 10 shows temperature contours and the interface position calculated at pull speeds of 1.2, 1.6, and 2.2 mm/s . The triple junction stabilizes slightly upstream of $x = 0$, which is the center of the helium jet. Most of the growth occurs around this point because it is the peak of the heat removal flux. The kinetics were evaluated assuming the sheet was oriented with the $[100]$ direction normal to the top surface. At the leading edge of the sheet there is a $\{111\}$ facet. This can be seen in the blow-up region at each pull speed; starting at the triple junction there is a nearly flat facet at an angle of 55° clockwise from the horizontal. At some depth the facet transitions away from a 55° slope and the solidification front smoothly curves towards horizontal. In the following, this location is called the “elbow.” At the triple junction, where the interface is the most super-cooled, the kinetic expression is determined by the 2D nucleation expression. Between the triple junction and the elbow, the kinetic expression is determined by the step propagation expression, and beyond the elbow, the roughened growth constant determines K .

In [1], we showed that the model predictions of the location of the triple

junction, x_{tj} , as a function of pull speed agree well with that seen in the experiment. Fig. 11 shows this behavior. As the pull speed is increased, the triple junction moves downstream and eventually crosses the peak of the heat removal profile (shown as a dashed line at $x_{tj} = 0$). With further increases in pull speed, there is a turning point beyond which no steady solutions exist. All of the solutions found along this curve are stable steady solutions. In the experiment, however, we were never able to pull at a speed for which the leading edge was near the center of the jet (5 mm/s for the case shown in the figure); before reaching this speed, there was typically a transition to dendritic growth. In the numerical simulations, it was observed that with increased pull speeds, the amount of supercooling increased. For example, for the three pull speeds shown in Fig. 10, the minimum liquid temperature, which occurred just upstream of the triple junction, was 1672, 1665, and 1660 K respectively. Conversely, the minimum temperature in the solid increased with pull speed with values of 1571, 1601, and 1627 K . These observations led to the conjecture that there was some sort of thermally induced instability occurring.

To verify this, the instability analysis is applied using the conditions along the interface as inputs. Fig. 12a and 12b shows the misalignment angle and the solid flux fraction along the interface for the three cases shown in Fig. 10. The horizontal axis corresponds to a 0.22 cm region around $x = 0$. As the pull speed increases the triple junction moves to the right, thus the lowest pull speed in Fig. 12 corresponds to the curve that starts farthest left. In all of the cases, along the facet, the misalignment angles are small until reaching the elbow. The elbow occurs where $K_{st}/|\sin(\theta)| = K_r$, which is at an angle of 1.15° . Subsequently, the misalignment smoothly approaches 55° , which corresponds to a horizontal interface. For the low pull speed case, the solid heat flux

fraction is greater than 0.5 everywhere except near the triple junction on the facet. With increasing pull speed, the solid heat flux fraction decreases along the facet. For the highest pull speed, the entire facet and the elbow region have a solid heat flux fraction below 0.5 indicating the possibility of thermal instability. In all cases, there is a sharp change in the slope of q_s exactly at the elbow.

To better understand the stability of the numerical solutions, the conditions along the interface were evaluated for their temporal and spatial-temporal stability. Essentially, each position on the interface was assumed to be locally planar and uniform so that the stability results could be applied. The information used for the stability analysis from each location was the solidification velocity, the tangential velocity, the liquid and solid heat fluxes, and the misalignment angle.

Fig. 12c shows the maximum temporal growth rate over all wavenumber disturbances, $\max_k(\text{real}(s))$, (solid curves) as well as the absolute growth rate, $\text{real}(s_0)$, (dashed curves) calculated for the conditions at each location on the interface. If the location was stable, then the maximum growth rate was zero because, as shown in Figs. 4 and 6 as well as by eq. 20, for stable cases the maximum growth rate is $s = 0$ at $k = 0$. The low pull speed case was stable everywhere along the interface. The higher pull speed cases were temporally unstable in a narrow region around the elbow where the kinetics transitioned from the step propagation mechanism to the roughened growth mechanism.

Fig. 12c also shows that the absolute growth rates, calculated only at unstable locations, are negative, which is consistent with the fact that the numerical simulations predicted stable steady solutions. Thus, the instability is convec-

tive, and a disturbance that propagates through the unstable region will grow, but after it passes, the solution will return to its steady configuration. In terms of the FSM experiments, if the amplitude of a disturbance grows large enough, it could cause some irreversible change to the solidification front (possibly a local change in crystal structure) which would be accompanied by a transition to dendritic growth. Qualitatively, this agrees with experimental observations in that there did not seem to be a repeatable speed at which FSM transitioned to dendritic growth. Because of the convective instability, the transition will be dependent on disturbances to the system.

One caveat with this analysis is that the width of the unstable region is on the order of $30\ \mu m$, which is narrow enough to make the locally planar assumption inaccurate; the most unstable wavelength for the conditions at the elbow is $80\ \mu m$, which is larger than the width of the region. Furthermore, the transition between the step propagation kinetics and the roughened growth kinetics (eq. 6) is not well-validated experimentally, which introduces further uncertainty. What can be safely concluded is that the elbow is the most sensitive location to perturbation and the sensitivity increases with increasing pull speed.

Finally, to show what effect the pull-speed has on stability, Fig. 13 shows the maximum temporal growth rate over the interface as a function of pull speed. As the pull speed increases, the width and magnitude of the temporal instability region grows. This implies that although the solution itself is not absolutely unstable, it becomes increasingly susceptible to perturbation.

8 Conclusions

Spatial-temporal stability analysis has been applied to a solidification model that includes both isotropic and non-isotropic kinetics. For the classical solidification problem that includes only thermal and capillary effects, it was shown that pulling the solid with a tangential velocity can cause a transition from absolute instability to convective instability. A simple criterion was derived for this transition that showed that it occurs for pull speeds that have the same magnitude as the growth velocity with the precise value depending on the solid heat flux fraction. For unstable growth with step propagation effects, disturbances propagate in the direction of the step motion as they grow. Pulling the solid tangentially in the direction of the step propagation is more effective at stabilizing the growth than pulling in the opposite direction.

These results were applied to understand possible instabilities in the floating silicon method. It was shown that as the pull-speed in FSM increases, the heat flux fraction decreases with more heat being removed through the liquid and less through the solid. This leads to conditions that normally would be thermally unstable at the faceted edge of the growth, but because of the kinetics remain stable. As the pull speed increases further, a small region at the transition between the faceted and roughened growth regions becomes convectively unstable. This convective instability was used to explain the transition to dendritic growth that has been seen in FSM experiments.

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10 Appendix

When step kinetics dominates (i.e. on a facet), eq. 20 accurately determines the imaginary part of $\tilde{s}$, but higher order terms are needed to predict the real part. The higher order correction is derived by expanding $\tilde{s} = \tilde{s}_0 + \tilde{s}_1$ in the dispersion relation and using the leading order solution for $\tilde{s}_0$ given by eq. 20. This gives the following correction

$$\tilde{s}_1 = -\frac{(\alpha_s K_0 + q_s) \left(\sqrt{1 + \frac{\tilde{s}_0}{\alpha_s k^2}} - 1 \right) + (\alpha_l K_0 + q_l) \left(\sqrt{1 + \frac{\tilde{s}_0}{\alpha_l k^2}} - 1 \right)}{1 + 2K_e k}, \quad (26)$$

where

$$K_0 = -\frac{K_{sp} \cot(\theta_0)}{\sin(\theta_0)} i k + K_e \tilde{s}_0 + k^2 \sigma. \quad (27)$$

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Fig. 1. Schematic of the experimental configuration.

Fig. 2. Geometry and boundary conditions for the linear stability analysis of faceted growth.

Fig. 3. Cartoon showing the atomistic layers producing downward propagation of a facet with a misalignment angle.

Fig. 4. Real part of $\tilde{s}$ for Case 1 (exponential growth rate). Solid curves are numerical solutions to dispersion relation (eq. 17). Dashed lines are approximate solutions from eq. 20 and are indistinguishable from the numerical solutions.

Fig. 5. Imaginary part of $\tilde{s}$ for Case 2 (oscillation rate). Solid curves are numerical solutions to dispersion relation (eq. 17). Dashed lines are approximate solutions given by eq. 20.

Fig. 6. Real part of $\tilde{s}$ for Case 2 (exponential growth rate). Solid curves are numerical solutions to dispersion relation (eq. 17). Dashed lines are approximate solutions given by the sum of eqs. 20 and 26.

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Fig. 8. Absolute growth rate s_0 for Case 1.

Fig. 9. Schematic showing the physical domain, spatial coordinates, and the initial mesh for the solidification simulations.

Fig. 10. Temperature contours at three different pull speeds: 1.2, 1.6, and 2.2 mm/s . Temperature contours correspond to 5 K intervals from 1665 to 1710 K with two additional contours at $T_m \pm 2.5 K$ where T_m is 1685 K . The two contours shown in the liquid in the blow-up region are $T_m - 2.5 K$ and $T_m - 5 K$.

Fig. 11. Triple junction position as a function of pull. Dashed line at $x_{tj} = 0$ indicates the peak of the heat removal profile.

(a) Misalignment angle

(b) Fraction of heat flux removed through the solid

(c) Maximum temporal (solid) and absolute (dashed) growth rates.

Fig. 12. Interface conditions as a function of position for pull speeds of 1.2, 1.6, and 2.2 mm/s

Fig. 13. Temporal growth rate as a function of pull speed.

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Table 1
Conditions used for stability analyses (based on silicon)

Parameter	Dimensional	Case 1	Case 2
v	4 mm/s	1	1
α_s	$8.7 \times 10^{-6} \text{ m}^2/\text{s}$	0.52	0.52
α_l	$2.5 \times 10^{-5} \text{ m}^2/\text{s}$	1.48	1.48
α_s/v	8.7 mm	1	1
θ_0	-	10°	0.1°
K_{st}	1.59 Ks/m	3.5×10^{-6}	3.5×10^{-6}
$K_{st}/\sin(\theta_0)$	-	2.0×10^{-5}	2.0×10^{-3}
$K_{st} \cot(\theta_0)/\sin(\theta_0)$	-	1.2×10^{-4}	1.15
K_r	79.4 Ks/m	1.8×10^{-4}	1.8×10^{-4}
σ	$9.8 \times 10^{-7} \text{ Km}$	2.5×10^{-7}	2.5×10^{-7}