Weierstrass’ variational theory for analysing meniscus stability in ribbon growth processes

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Abstract

We use the method of free energy minimization based on the first law of thermodynamics to derive static meniscus shapes for crystal ribbon growth systems. To account for the possibility of multivalued curves as solutions to the minimization problem, we choose a parametric representation of the meniscus geometry. Using Weierstrass’ form of the Euler-Lagrange equation we derive analytical solutions that provide explicit knowledge on the behaviour of the meniscus shapes. Young’s contact angle and Gibbs pinning conditions are also analyzed and are shown to be a consequence of the energy minimization problem with variable end-points. For a given ribbon growth configuration, we find that there can exist multiple static menisci that satisfy the boundary conditions. The stability of these solutions is analyzed using second order variations and are found to exhibit saddle node bifurcations. We show that the arc length is a natural representation of a meniscus geometry and provides the complete solution space, not accessible through the classical variational formulation. We provide a range of operating conditions for hydro-statically feasible menisci and illustrate the transition from a stable to spill-over configuration using a simple proof of concept experiment.
1 Introduction

*These investigations, which have found their confirmation in striking agreement with careful experiments, are among the most beautiful enrichment’s of natural science that we owe to the great mathematician.*

—Carl Fredrich Gauss on Laplace’s theory of capillary action, which was later refined by him into its modern variational form.

The Young-Laplace equation was developed by Thomas Young [1], who provided a qualitative theory for surface tension, and Pierre-Simon Laplace [2], who mathematically formalized the relationship described by Young. This theory was later refined by Carl Fredrich Gauss [3] using Bernoulli’s principle of virtual work. Using the fundamental principles of dynamics, he derived the Young-Laplace equation and Young’s contact angle condition from a single variational framework. He argued that the energy of a mechanical system in equilibrium is unvaried under arbitrary virtual displacements consistent with the constraints. This spirit of variational analysis is still used in practice to describe the meniscus shape in interface problems.

The existence of a static meniscus plays a critical role in capillary-shaped ribbon growth systems such as the Dendritic web growth (WEB), Edge-defined Film Growth (EFG), Low Angle Silicon Sheet (LASS) growth and the Horizontal Ribbon Growth (HRG) process [4]. Fig. 1 describes the schematic of a HRG process which will serve as an example to illustrate the application of our theory. A bath of molten substrate is cooled from the top to form a thin ribbon of single crystal which is continuously extracted. A narrow Helium cooling jet is used to provide intense cooling for solidification and keeps the starting point of the ribbon almost fixed for all feasible pull speeds [5]. A seeding process takes place at the outlet while the melt is being continuously replenished at the other end. Thin sheets of single crystal can be pulled at relatively high speeds due to enhanced heat transfer with the surroundings [6]. This provides an advantage over the present crystal growing methods, like the Czochralski process, where the sheets are prepared by slicing a single crystal boule followed by tedious, time consuming grinding and lapping operations which result in a large percentage of the original crystal being wasted [7]. The weight of the ribbon is supported by the melt, which forms a meniscus between the ribbon and the edge of the crucible, thereby reducing the mechanical stresses on the crystal.

![Figure 1: Schematic for a horizontal crystal ribbon growth process. The formation of a meniscus at the end of the crucible is essential for steady state operation](image)

Several well known ribbon growth techniques can be characterized by the angle ($\beta$)
at which the ribbon is pulled from the melt as illustrated in Fig. 2. For the case when \( \beta = 90^\circ \), the ribbon is pulled perpendicular to the surface of the melt and relates to the family of vertical ribbon growth techniques like the Edge-defined Film Growth (EFG) and the Dendritic web (WEB) growth process. In the EFG process, the role of the crucible in the previous example is substituted by a melt-wettable die which provides a pinning boundary for the meniscus. The dye determines the shape of the meniscus and thus the cross section of the growing crystal ribbon [8, 9]. This makes it important to understand the meniscus geometry in order to study its effect on crystal shape and quality.

\[
\begin{align*}
\beta &= 90^\circ \\
\beta &\approx 5^\circ \\
\beta &= 0^\circ
\end{align*}
\]

Figure 2: Characterization of the ribbon growth family based on pull angle (\( \beta \))

Another family of ribbon growth methods, characterized by their low pull angles, are the horizontal ribbon growth techniques like the Low Angle Silicon Sheet (LASS) process and the Horizontal Ribbon Growth (HRG) process [10, 11]. These methods have the advantage of having a large solid-liquid interfacial area making it easier for the latent heat to dissipate and leading to higher production speeds. However, two technical issues appear while running experiments: the ribbon freezing onto the crucible (down-growth) and the melt spilling-over the crucible [6, 12, 13]. These two issues are directly related to the formation of a short or unstable meniscus between the ribbon and the crucible edge [14, 15, 16]. The main objective of this paper is to analyze these instabilities of the meniscus for the horizontal ribbon growth processes, while at the same time keeping the analysis general enough to be extended to any other problem of physical or engineering importance.

The meniscus profiles for HRG were first investigated by Rhodes et al. [17] around the same time as Kudo [12] performed his first HRG experiments with Silicon. They developed a mathematical model based on hydrostatics, to describe the shape of the meniscus that must be formed between the ribbon and the crucible edge. They found that the hydrostatically feasible configurations require the meniscus to be “taller” than the melt height, and that the ribbon be pulled at a slight angle, which coincided with Kudo’s experimental operation. In 2012, Carl Bleil and researchers at the University of Minnesota [14, 15, 18] constructed a thermal-capillary model describing the interaction between fluid flow and heat transfer in a HRG system. In their results they captured the critical nonlinearities in the system, such as the existence of multiple menisci for a given pulling speed. The problems of melt spilling over and freezing to the crucible were assessed by doing a sensitivity analysis on the length of the meniscus as a function of melt height and pulling angle [15]. Multiplicity of menisci with respect to pull speeds have also been observed in the case of EFG process [19, 20]. This multiplicity is manifested as two states of the ribbon thickness for the same pull speed but with different failure limits [21].

Classical variational analysis have proven useful in the study of meniscus stability for capillary based processes. For the Czochralski process, Mika and Uelhoff [22] used free energy minimization along with the concepts of variational calculus to numerically determine instability conditions for the meniscus. Mazuruk and Volz [23, 24] addressed the static stability problem for the Bridgman process using numerical simulations to calculate the
sign of the second order variations for the governing free energy formulation. Using the observation that the contact angle between the crystal and the melt should converge towards a constant value—11° for the case of Silicon as investigated by Mazuruk et al. [25]—Surek [26] developed a theory for shape stability in capillary shaped crystal growth systems based on deviations from the growth angle. Dynamic and static stability was numerically addressed by Tarachenko [27, 28] using Lyapunov based techniques and variational principles.

Outside the area of crystal growth, variational principles have been used to prove the existence and stability menisci shapes for varying capillary geometries [29]. A similar approach has been used by Pitts [30] and Vogel [31] to study the shape of liquid pendant drops and identify regions of stability before the drop breaks. Soligno et al. [32] implemented a numerical method to minimize thermodynamic potential function and calculate the interface shape of liquids for various wall geometries. Lawal and Brown [33, 34] used polar co-ordinates in their variational formulation to obtain multiple critical solutions for their drop geometries on an inclined surface. They observed that for a fixed Bond number, axisymmetric sessile shapes on horizontal surfaces lose stability at a drop volume that corresponds to a point of bifurcation into a family of asymmetric shapes. As we shall see in Section 6, the ribbon growth configuration also admits a point of bifurcation into stable and unstable family of menisci.

Very recently Oliveros et al. [16] used the classical variational approach to find existence and stability conditions for menisci in a HRG process. The analysis showed that stationary menisci arising as solutions to the classical Euler-Lagrange equation were stable as long as the solution satisfied the existence conditions. Due to the well known complexity and non-linearity of the ribbon growth systems, we were left with the question of whether or not the system had any unstable configurations that can’t be captured by the traditional variational tools. In Weierstrass’ variational theory this limitation is overcome by formulating the geometry of the meniscus in parametric form. This approach allows for the possibility to find stationary curves described by multi-valued functions (see Fig. 3). The theory, originally developed by Weierstrass in his university lectures, is comprehensively described in the book by Oskar Bolza [35]. We refer the reader to Oliveros [36] for further details on the development of the theory.

It is important to note that heat transfer and fluid flow also play a critical role in capillary shaped crystal growth processes [37, 38]. However here we decouple these phenomena and focus only on the static stability of the meniscus in absence of heat transfer. In doing so, we are able to provide deeper explanations to some complex phenomena like multiplicity of menisci as observed in EFG and HRG processes [15, 19] and the existence of destabilizing multi-valued menisci in crystal growth systems [39, 40], among others, using a more fundamental, first principles approach.

2 Problem Statement

For a general (three-phase) system described in Fig. 3 consisting of fluid and gas separated by an interface \( x = x(y) \) and a rigid fixed ribbon, the free energy (\( \Delta U \)) in question is given as [16]:

\[
\Delta U = \int_0^H -\Delta P(x)dy + \gamma \sqrt{1 + x^2} dy = \int_0^H F(x, y, x') dy. \tag{1}
\]

This is commonly known as the classical free energy formulation. In this expression, the right-hand side is divided into two terms. The first term is the potential energy due to hydro-static pressure (\( \Delta P \)). The second term is the free surface energy of the interface due to surface tension (\( \gamma \)). \( H \) is the maximum height of the meniscus and corresponds to the total height of integration. The Euler-Lagrange equation for this variational problem gives
us the Young-Laplace equation:

\[ \Delta P + \gamma \frac{x''}{(1 + x'^2)^{3/2}} = 0. \]  

(2)

Analytic solutions have been developed for ribbon growth configurations in 2 and 3 dimensions using Legendre elliptic functions [16, 41]. However, in some cases the solution to geometric problems that use the classical variational formula cannot be described by functions of the form \( x = x(y) \) in a Cartesian coordinate system. For example, the multi-valued meniscus of a non-wetting sessile drop on an incline plane cannot be described using single-valued functions [34, 42]. A similar problem exists in describing the full spectrum of minimum energy curves for ribbon growth systems. The stationary curves arising from free energy minimization are often multi-valued and therefore require a less restricting and more “natural” representation. By choosing an appropriate parametric variable, we see the emergence of a natural representation of the meniscus shape which allows us to find the complete solution space. These interface curves are also shown to share similarities with the family of Euler’s elastic curves.

3 Free energy reformulation

\[ \Delta U = \int_{s_0}^{s_1} -\Delta P \times (xy')ds + \gamma \sqrt{x'^2 + y'^2}ds = \int_{0}^{s_1} g(x, y, x', y')ds. \]  

(3)

where \( s_0 \) is the total length of integration. The first term accounts for the hydro-static energy of the system. Due to the presence of gravity, the pressure in the liquid is given as a function of height, \( \Delta P = \rho g (h - y) \), where \( \rho \) is the density of the liquid. The second term is the surface energy of the interface due to interfacial tension \( \gamma \).

In performing this transformation, the value of \( \Delta U \) must remain invariant for any type of parametric form chosen for \( x \) and \( y \). Weierstrass showed that the necessary and sufficient
condition for the invariance of $\Delta U$ is that the functional $G$ be homogeneous and of degree one in the variables $x'$ and $y'$ [35, p. 118], i.e.
\[
g(x, y, kx', ky') = kg(x, y, x', y'), \tag{4}
\]
where the prime represents differentiation with respect to $s$. From this homogeneity condition, there follow several relationships between the partial derivatives of $G$, which are useful in constructing the expressions for the first and second variation of $\Delta U$.

Non-dimensionalizing length scales with respect to the capillary constant, $\lambda_c = \sqrt{\gamma/(\rho g)}$ yields,
\[
U(X, Y) = \int_0^{S_t} (Y - H)XY'dS + \sqrt{X' + Y'}dS \\
= \int_0^{S_t} G(X, Y, X', Y')dS, \tag{5}
\]
where,
\[
\Delta U = \gamma \lambda_c U, \quad x = \lambda_c X, \quad y = \lambda_c Y, \quad s_t = \lambda_c S_t \quad \text{and} \quad h = \lambda_c H \tag{6}
\]
The homogeneity condition is not affected by non-dimensionalization.

4 Stationary curves via the first variation

Our objective is to find conditions the stationary curves $X(S), Y(S)$ that set the first variation of $U$ to zero. Let $\epsilon \xi(S)$ and $\epsilon \eta(S)$ be small perturbations to these curves with $\epsilon$ as small as desired. The end points on the curve are kept fixed i.e. $\xi$ and $\eta$ are zero at the end points. The energy of this neighbouring curve is given by
\[
U(X + \epsilon \xi, Y + \epsilon \eta) = \int_0^{S_t} G(X + \epsilon \xi, Y + \epsilon \eta, X' + \epsilon \xi', Y' + \epsilon \eta')dS. \tag{7}
\]

Applying Taylor’s formula to the integrand, we obtain
\[
U(X + \epsilon \xi, Y + \epsilon \eta) = U(X, Y) + \epsilon \delta U + \frac{\epsilon^2}{2} \delta^2 U + O(\epsilon^3) \tag{8}
\]
\[
\delta U = \int_0^{S_t} \left( \xi G_X + \xi' G_{X'} + \eta G_Y + \eta' G_{Y'} \right) dS. \tag{9}
\]
We call $\epsilon \delta U$ the first variation of the energy functional $U$. For $X(S), Y(S)$ to be a critical point of $U$, we infer that $\delta U = 0$ i.e.
\[
\int_0^{S_t} \left( \xi G_X + \xi' G_{X'} + \eta G_Y + \eta' G_{Y'} \right) dS = 0, \tag{10}
\]
for otherwise, we could increase or decrease the value of $U$ by choosing $\epsilon$ to be of the same or a different sign of the integral in Eq. (10), respectively.

Using integration by parts and assuming continuous derivatives of the functions involved, we arrive at the Euler-Lagrange equations:
\[
G_X + \frac{dG_X'}{dS} = 0, \quad G_Y + \frac{dG_Y'}{dS} = 0 \tag{11}
\]

Due to the homogeneity condition (4), these two equations (11) are not independent of each other, as we proceed to show. Differentiating Eq. (4) with respect to $K$, and putting $K = 1$, yields
\[
X'G_X + Y'G_Y = G. \tag{12}
\]
Differentiating this expression with respect to $X'$ and then to $Y'$, we obtain

$$\frac{1}{Y'^2}G_{X'X'} = -\frac{1}{X'^2}G_{X'Y'} = \frac{1}{Y'^2}G_{Y'Y'} = G_1,$$  \hspace{1cm} (13)

where $G_1$ is the common value among these expressions. Differentiating Eq. (4) partially with respect to $X$ and $Y$ we get

$$G_X = X'G_{X'X'} + Y'G_{X'Y'}, \quad G_Y = X'G_{Y'X} + Y'G_{Y'Y}.$$  \hspace{1cm} (14)

Using Eqs. (13) and (14) in the Euler-Lagrange equation yields:

$$G_X - \frac{d}{dS}G_{X'} = Y'T, \quad G_Y - \frac{d}{dS}G_{Y'} = -X'T,$$  \hspace{1cm} (15)

where

$$T = G_{XY'} - G_{YX'} - G_1(Y''X' - X''Y').$$  \hspace{1cm} (16)

Assuming that $X'$ and $Y'$ don’t vanish simultaneously in the interval $[0, S_t]$, the two expressions (11) are equivalent to the following differential equation:

$$G_{XY'} - G_{YX'} - G_1(Y''X' - X''Y') = 0.$$  \hspace{1cm} (17)

This equation is the Weierstrass’ form of the Euler-Lagrange equation. In order to solve this equation, we need to define the parameter $S$ and its relationship with $X$ and $Y$. The choice of the parameter must be such that both functions come out as single-valued functions of $S$.

In our case we have that

$$G_1 = \frac{1}{(X'^2 + Y'^2)^{3/2}},$$  \hspace{1cm} (18)

$$G_{XY'} = Y' - H,$$  \hspace{1cm} (19)

$$G_{YX'} = 0.$$  \hspace{1cm} (20)

So the Euler-Lagrange equation becomes:

$$H - Y = \frac{X''Y' - X'Y''}{(X'^2 + Y'^2)^{3/2}}.$$  \hspace{1cm} (21)

Eq. (21) is the Young-Laplace equation in parametric form. The term to the right hand side of the equation is also known as the curvature.

### 4.1 Analytic form and family of solutions

The differential equation (21) together with the initial condition determines the critical curve, but not the function $X(S)$ and $Y(S)$. In order to do find these functions we must add a second equation or differential relation between $S, X, Y$. This additional relation should be such that $X$ and $Y$ come out as single valued functions of $S$. In order to find analytic solutions to the parametric Young-Laplace equation, we make the transformation

$$X'(S) = \cos \Omega(S), \quad Y'(S) = \sin \Omega(S),$$  \hspace{1cm} (22)

where $\Omega$ is the tangential angle to the meniscus. These substitutions define the independent variable $S$ to be the arc length of the meniscus and turn the Young-Laplace equation into

$$\Omega'(S) = H - Y.$$  \hspace{1cm} (23)

This transformation splits the Young-Laplace equation into a system of 3 ODEs. We set the initial conditions of the meniscus to have general contact angle conditions

$$X(0) = 0, \quad Y(0) = 0, \quad \Omega(0) = \theta.$$  \hspace{1cm} (24)
To find an analytic solution to the system of ODE’s we differentiate Eq. (23) and substitute Eq. (22) to find

\[ \Omega''(S) = -Bo \sin(\Omega(S)). \]  

We observe that the dynamics of the tangent angle \( \Omega \) are similar to the dynamics of a pendulum or an elastic rod [43]. Multiplying Eq. (25) with \( \Omega'(S) \) and integrating, we get,

\[ \frac{1}{2} \Omega^2 - \cos \Omega = A. \]  

In the case of a simple pendulum, the integration constant \( A \) is defined as the energy of the system. Using the initial conditions \( \Omega(0) = \theta \) and \( \Omega'(0) = H \), we evaluate the integration constant as,

\[ A = \frac{H^2}{2} - \cos \theta. \]  

Using the trigonometric identity \( \cos \Omega = 1 - 2 \sin^2 \Omega/2 \), we arrive at

\[ \Omega'(S) = 2 \sqrt{\frac{A + 1}{2} - \sin^2 \Omega/2}. \]  

the solution to this differential equation can be explicitly written down in terms of Legendre elliptic and Jacobi amplitude functions,

\[ \Omega(S) = 2 \text{am}\left(\sqrt{\frac{1 + A}{2} S + F\left(\frac{\theta}{2} \big| \frac{2}{1 + A} \right)} \bigg| \frac{2}{1 + A}\right). \]  

where \( F(u|m) \) and is the incomplete elliptic integrals of the first kind and \( \text{am}(u|m) \) is the Jacobi amplitude function. \( Y(S) \) can be calculated directly using Eq. (23) and the identity \( \text{am}(u,k) = \int_0^u \text{dn}(u',k)du' \),

\[ Y(S) = H - \sqrt{2(1 + A)} \text{dn}\left(\sqrt{\frac{1 + A}{2} S + F\left(\frac{\theta}{2} \big| \frac{2}{1 + A} \right)} \bigg| \frac{2}{1 + A}\right). \]  

where \( \text{dn}(u|m) \) is the Jacobi delta amplitude function. Using the result from Eq. (30) and substituting Eq. (26) into the expression for \( X'(S) \) in Eq. (22) we obtain

\[ X(S) = \sqrt{2(1 + A)} E\left(\text{am}\left(\sqrt{\frac{1 + A}{2} S + F\left(\frac{\theta}{2} \big| \frac{2}{1 + A} \right)} \bigg| \frac{2}{1 + A}\right) \bigg| \frac{2}{1 + A}\right) - AS. \]  

\( E(u|m) \) is the incomplete elliptic integral of the second kind.

Now that we have an an analytic expression for the meniscus geometry, we can plot the interface for different values of energy \( A \). For the purpose of this illustration, we consider the non-dimensional melt height \( H \) to be 1 while the pinning angle \( \theta \) is being varied in Fig. 4.

Since Eq. (25) is equivalent to the dynamics of an elastic rod under compression [44]; the family of curves, that describe the shape of a meniscus, are also solutions to Euler’s elastica problem [45]. This analogy was previously known to Laplace [46, p. 379] and Maxwell [47, p. 265]. Plotting for \( A > 1 \) might require certain inversion transformations. These along with other identities mentioned in this section can be found in Abramowitz and Stegun [48].

5 Stability analysis via the second variation

In this section we consider the stability of the variations to the critical curves when the end-points are considered fixed. Using Taylor series representation, the second variation in parameter representation is expressed as follows:

\[ \delta^2 U_0 = \int_0^{S_t} \delta^2 G \, dS, \]  

(32)
Figure 4: Family of solutions for the parametric Young-Laplace equation

\[
\delta^2 G = G_{XX} \xi^2 + 2G_{XY} \xi \eta + G_{YY} \eta^2 + 2G_{XX}' \xi' \xi'' + 2G_{YY}' \eta' \eta'' + 2G_{XY}' \xi' \eta'' + 2G_{YX}' \eta' \xi'' + G_{XX}'' \xi''^2 + 2G_{XY}'' \xi' \eta'' + G_{YY}'' \eta''^2. \tag{33}
\]

Recall that in order for the curve described by \( Y(S) \) and \( X(S) \) to be a minimum -and therefore stable-, its second variation should be positive; so the value of the integral above must be always positive in the range of integration. Using a lengthy factorization, Weierstrass transformed the second variation into the classical quadratic functional

\[
\delta^2 U_0 = \int_{S_0}^{S_f} \left[ G_1 \left( \frac{d\omega}{dS} \right)^2 + G_2 \omega^2 \right] dS. \tag{34}
\]

In the above integral we have that

\[
\omega = Y' \xi - X' \eta, \tag{35}
\]

and \( G_2 \) satisfies the following relationships:

\[
G_2 = \frac{L_2}{Y'^2} = \frac{M_1}{X'Y'} = \frac{N_1}{X''}, \tag{36}
\]

with

\[
L_2 = G_{XX} - Y'' \ G_1 - \frac{dL_1}{dS}, \tag{37}
\]

\[
M_2 = G_{XY} + X'' Y'' \ G_1 - \frac{dM_1}{dS}, \tag{38}
\]

\[
N_2 = G_{YY} - X''^2 \ G_1 - \frac{dN_1}{dS}, \tag{39}
\]

\[
L_1 = G_{XX}' - Y'' \ G_1, \tag{40}
\]

\[
M_1 = G_{XY}' + X'' Y'' \ G_1 = G_{YX}' + Y' X'' \ G_1, \tag{41}
\]

\[
N_1 = G_{YY}' - X'' \ G_1. \tag{42}
\]

The form of the integral allowed Weierstrass to apply the classical results of the calculus of variations. Namely, Legendre’s necessary condition and Jacobi’s test. Legendre’s necessary condition for a minimum requires that

\[
G_1 \geq 0, \tag{43}
\]
along the stationary curve described by $X(S)$ and $Y(S)$.

Jacobi’s test requires that the solution to the differential equation,

$$G_2 u - \frac{d}{dS} \left(G_1 \frac{du}{dS}\right) = 0,$$  \hspace{1cm} (44)

must not have conjugate points in the integration interval, i.e:

$$u(S) \neq 0 \quad 0 < S < S_t.$$  \hspace{1cm} (45)

The strength of this theory lies in the fact that it is possible to find an extended solution space to the original meniscus problem and a more general criterion for static stability, which is not possible to accomplish with the usual Cartesian representation of a function, such as $X = X(Y)$.

In order to have the Jacobi test satisfied we require that the solution to the differential equation

$$G_2 u - (G_1 u')' = \frac{Y'''}{Y'} u - u'' = 0,$$  \hspace{1cm} (46)

must not have conjugate points in the interval of integration, i.e.

$$u(S) \neq 0 \text{ for } 0 < S < S_t.$$  \hspace{1cm} (47)

Eq. (46) is equivalent to:

$$(Y'u' - Y''u)' = 0.$$  \hspace{1cm} (48)

Thus

$$Y'u' - Y''u = K_1.$$  \hspace{1cm} (49)

Dividing the expression above by $Y'^2$ we get:

$$\frac{Y'u' - Y''u}{Y'^2} = \left(\frac{u}{Y'}\right)' = \frac{K_1}{Y'^2}.$$  \hspace{1cm} (50)

So the condition for stability becomes,

$$u(S) = K_1 Y'(S) \int_0^S \frac{dS}{Y'^2(S)^2} \neq 0 \text{ for } 0 < S < S_t.$$  \hspace{1cm} (51)

The integral in Eq. (51) is always positive as long as $Y'(S) \neq 0$ between 0 and $S_t$ (given $S_t > 0$), otherwise the integral does not converge. The term $K_1 Y'(S)$ does not change sign as long as $Y'(S)$ does not change sign in the $(0, S_t)$ interval. Therefore the issue of stability reduces to finding the range of values for which $Y'(S)$ crosses zero in the $(0, S_t)$ interval. This is easier to analyze recalling the fact that $Y'(S) = \sin \Omega(S)$, where $\Omega(S)$ is the tangential angle of the meniscus with respect to the horizontal axis. If $\sin \Omega(S)$ is always positive or always negative in the integration interval, the function $u(S)$ will be a well defined function with no conjugate points. Thus we simplify our stability criterion to the following expression:

$$\sin(\Omega(S)) > 0 \lor \sin(\Omega(S)) < 0 \forall \ S \in (0, S_t),$$  \hspace{1cm} (52)

6 Results

We apply the theory developed in the previous sections to study the properties of a meniscus in a silicon ribbon growth process while keeping in mind that they can be applied to buckling problems or a range of other problems of physical and engineering importance. To characterize Fig. 1 in more detail, the edge of the crucible is considered to be rectangular ($\phi = 90^\circ$). The growth angle ($\sigma$) for silicon was taken to be $11^\circ$ from investigations by
Figure 5: (left) Stationary meniscus shapes obtained using the analytical solution in parametric form. Curves correspond to a value of $\beta = 10^\circ$ and $h = 5.35 \, mm$. (right) The sine of the tangent angle $\Omega(s)$ for different pinning angles. The curves crossing zero correspond to the unstable modes.

| Parameter                             | Symbol | Value       |
|---------------------------------------|--------|-------------|
| Density of liquid silicon             | $\rho$ | $2570 \, [kg \, m^{-3}]$ |
| Acceleration of gravity               | $g$    | $9.8 \, [m \, s^{-2}]$   |
| Surface tension of silicon           | $\gamma$ | $0.72 \, [J \, m^{-2}]$  |
| Silicon growth angle                  | $\sigma$ | $11^\circ$  |
| Melt-graphite wetting angle           | $\theta_e$ | $30^\circ$  |

Table 1: Material properties and parameters used in the illustrative example.

Mazuruk et al. [25], Swartz et al. [49], Surek and Chalmers [50] and Champion et al. [51]. One end of the meniscus is considered to remain pinned at the edge of the crucible and the other end to intersect the ribbon at a fixed growth angle of $\sigma = 11^\circ$. These boundary conditions are shown to be a consequence of a variational formulation with moving boundaries as derived in the Appendix. The pull angle ($\beta$) and the height of the melt ($h$) are degrees of freedom. It is of interest to find stable operating regimes for the meniscus over the parameter space of $\beta$ and $h$. The required material properties are summarized in Table 1 and are used to dimensionalize the equations and the results.

The plot on the left in Fig. 5 describes the various stationary meniscus shapes for a representative pulling angle of $\beta = 5^\circ$ and melt height $h = 5.35 \, mm$ ($H = 1$). We use the analytic expressions for $x(s)$ and $y(s)$, with different $\theta$ values to plot the interface curves and stop when the interface reaches the angle of $\sigma + \beta$. In order to show the concept of static stability, we focus on the results obtained from Jacobi’s test (Legendre’s condition for a minimum is always satisfied for all meniscus shapes). The plot on the right in Fig. 5 shows the sine of the tangential angle as a function of the arc length. As we mentioned before, the sine of the tangential angle must not vanish between 0 and $s_t$. From the figures we show that menisci in which the contact angles are greater than zero are statically stable, whereas the curves for values of contact angle lower than zero cross the horizontal axis. The family of stable and unstable curves converge in the limit $\theta \to 0$.

Let $x^*$ and $y^*$ be the parametric co-ordinates describing the equation for a ribbon. We assume the shape of the ribbon to be a straight line starting from $l_c = -5.35 \, cm$ ($L_c = -10$) and represented by

$$L(x^*, y^*) = (y^* - h) - \tan \beta (x^* - l_c) = 0.$$  \hspace{1cm} (53)
The desired solution is then given by any curve described in figure 5, whose end point lies on this line. This can be formulated as a boundary value problem:

\[
\begin{align*}
    x(0) &= 0 \\
    y(0) &= 0 \\
    \Omega(s_t) &= \beta + \sigma \\
    L(x(s_t), y(s_t)) &= 0
\end{align*}
\]

(54)

We use a Newton-Raphson solver to find curves that satisfy (54). Two curves, one stable and one unstable, are found and illustrated in Fig. 6 along with a diagram of the system (to scale) to better visualize the concept of hydro-static stability.

Figure 6: Hydro-statically stationary configuration for a melt level of 5.35 mm and a pulling angle of 5°. The solid curve corresponds to a statically stable configuration and the dashed curve corresponds to an unstable configuration.

Figure 7: Saddle node bifurcations in the meniscus length \( S_t \) and pull angle \( \beta \) solution space. Inset: A zoomed up diagram of the solution space for negative pull angles.

6.1 Effect of pull angle

The behaviour of the meniscus shapes is influenced by the pull angle (\( \beta \)) through the boundary conditions described in (54). To get a better description of the meniscus multiplicity
observed above, we vary the pulling angle to evaluate the feasibility of stationary menisci. Fig. 7 provides a description of the solution space for a melt height of $h = 5.35\, \text{mm}$ as a function of the pulling angle. The choice of meniscus length as the $Y$-axis was motivated from literature on elasticity and bifurcation theory, where the geometry of curves described by Eq. (25) has been extensively studied [44].

Representative meniscus shapes are drawn along the solution curves to describe their geometry for a few choice of pull angles. The dashed curves describe the family of unstable solutions, characterized by a point of zero slope where the Jacobi condition is not met. In the neighbourhood of this point, it is possible to perturb the curve such that the second order variation is negative and the solution is not a minimum. Vice versa, the solid curves describe the statically stable solutions which minimize the thermodynamic energy of the system. The pinning condition at the crucible edges due to Gibbs has not been considered here and is commented on separately in Appendix A.

We observe that it is not always possible to find a feasible solution for any given value of pull angle. This operational limit has also been realized in the thermal-capillary simulations performed by Daggolu et al. [15], however their analysis was limited to the narrow stability region on the left. Two saddle node bifurcations are observed in our analysis that divide the feasible solution space into two disjoint regions. The feasible region on the left has a smaller range of pull angles available for stable operation. The crucible limit shown inset is the limit at which the meniscus length goes to zero. Decreasing the pulling angle to this limit would cause the bottom part of the ribbon to get closer to the crucible edge and result in ribbon freezing onto the crucible. On the other hand, increasing the pull angle beyond the bifurcation point results in the meniscus becoming unstable and cause the melt to spill-over from the crucible.

Given the narrow range of operation for negative pull angles, it would be desirable to operate the ribbon growth process at positive pull angles, beyond $4.5^\circ$ for the case of $H = 1$, as there is no upper limit to how high the pulling angles can be. The feasible region on the right illustrates the variety of meniscus shapes that can be achieved for positive pull angles. This compares to the method of low-angle silicon sheet (LASS) growth process where is the ribbon is extracted from the melt at a slight positive angle with the horizontal [10].

### 6.2 Effect of melt height

A successful design for a ribbon growth process requires understanding the effect of melt height ($H$) on the stability of the meniscus. It is therefore useful to study how the melt height influences the landscape of the solution space described in Section 6.1.

Fig. 8 succinctly illustrates the range of feasible pull angles as a the melt height is varied. For example, the region in the shaded section at $H = 1$ can be thought of as a projection of the feasible pull angles (solid curves) in Fig. 7 onto the X-axis. Therefore, the entire shaded region in Fig. 8 describes the existence of a stable meniscus at every point over the parameter space of $H$ and $\beta$.

Representative meniscus shapes have been drawn for some chosen values of $H$ and $\beta$. At some places, a plus symbol has been used to denote the point where the meniscus belong. For $H < 2$ ($h < 10.7\, \text{mm}$) we see that it is possible to find a stable meniscus for pull angles as large as $90^\circ$. At this point the arrangement corresponds to vertical ribbon growth techniques like WEB, EFG. What is interesting to note is that as the pull angle increases, the meniscus becomes longer and the meniscus-ribbon triple point goes further away from the crucible edge. This observation is the guiding principle behind low-angle silicon sheet (LASS) growth process and circumvents the problem of ribbon freeze-over by moving the triple phase contact point on the ribbon away from the crucible edge.

As the melt height increases, we see that above $H = 0.2$, the feasible solution space splits. The portion in between the regions is the melt spill-over region. In this region it is not possible to form a stable meniscus to support the melt from spilling over the crucible. Since the solution space for positive pull angles is much larger than the negative pull angles,
the scale for the negative pull angles has been increased to meaningfully show the feasible solution space. Notice also that there is an upper limit to the height of the melt that the meniscus can accommodate. Beyond this height, a meniscus can no longer exist and the melt spills over from the crucible edge.

The feasibility region shown in Fig. 8 does not consider the Gibbs inequality condition that arises at the crucible edge. Gibbs’ inequality provides a range of pinning angles ($\theta$) at the crucible edge for which the meniscus remains stable. Since the Gibbs limit is a material property and also depends on the geometry of the crucible edge, we provide contours for some chosen pinning angles to find a subset of the feasible (shaded) region that satisfies Gibbs’ inequality.

![Figure 8: Stable meniscus region over the parameter space of melt height and pull angle](image)

7 Experimental Design

A miniature proof-of-concept experiment is used to study and illustrate the mechanism of melt spill over in a HRG configuration when the pull angle is varied. A polyethylene ribbon ($\rho_{pe} = 0.93 \text{g/cm}^3 < \rho_{water}$) rests completely on top of the water contained in a plastic bath such that the inclination with respect to the top surface of the water can be varied.

From Section 6.2, we observed a range of infeasible pull angles around the horizontal position ($\beta = 0$) when $H$ was greater than 0.2. To test this hypothesis, we induce spill-over by slowly decreasing the angle of inclination with the water surface while photographing the changes in the shape of the water meniscus. Fig. 9 displays a sequence of photographs showing the bulking of the meniscus as the pulling angle decreases. The top left-most photograph shows the shape of a meniscus in which the ribbon is inclined at a positive angle (a stable configuration). This configuration would make it least likely for the ribbon to freeze on top of the crucible edge. The right-most bottom photograph is the shape of a meniscus prior to spilling over the crucible (an unstable configuration) as the ribbon becomes horizontal. Despite the difference in materials, we see that the stability analysis from our theory agree qualitatively with the experimental observations.
8 Conclusions

This paper provides a parametric formulation and a solution to the generalized static stability problem for the meniscus in a ribbon growth process. Due to the geometric nature of the meniscus problem, we observe that the method of parametric representation is not only preferable but also one which furnishes a complete solution. Using Weierstrass' variational theory, we found analytic expressions describing the shape of the meniscus and compare it with the family of Euler's elastic curves. This similarity can be used to exchange concepts from elasticity theory in order to study stability and bifurcations of menisci shapes in liquids and vice versa.

The stability of the meniscus are evaluated using Legendre and Jacobi test conditions. A range of stable operating conditions are provided over the parameter space of melt height and pull angle. Two bifurcation points are observed which divide the solution space into two regions. The infeasibility zone between the two solution spaces, which include the horizontal position of the ribbon, didn’t have a stable meniscus solution to support the ribbon. Growing a ribbon in this region leads to melt spilling over from the meniscus until the melt height decreases to the stable region. This argument is supported by doing a simple proof of concept experiment, in which the phenomena of spill-over is created using a polyethylene ribbon resting on a bath of water. Given the vast range of stable positive pull angles, we conclude that it is appropriate to incline the ribbon above a certain threshold angle to ensure stability of ribbon growth as the horizontal configuration was statically unstable.

A Young’s contact angle and Gibbs pinning condition

The parametric Young-Laplace equation (21) derived in Section 4 relies on contact angle conditions at the boundary in order to find stationary curves that describe the meniscus shape. By perturbing the end points of the meniscus, we show that the contact angle and the Gibbs pinning conditions follow as a consequence of the free energy minimization of the system.

Consider the free energy formulation for the meniscus $x(s)$, $y(s)$ as defined in Section 3 and add the surface energy of the solid boundaries in contact with the air and the liquid. For simplicity, we briefly consider the case where only the end point at the origin is varied while the end point at $s_t$ is considered fixed. In this case,

$$
\Delta U = \int_0^{s_t} -\Delta P \times (xy') ds + \gamma \sqrt{x'^2 + y'^2} ds + A_1 \gamma_1 + A_2 \gamma_2,
$$

(55)
where $A_1$ and $A_2$ are the areas of the solid crucible in contact with the melt and the air respectively. $\gamma_1$, $\gamma_2$ are the interfacial energies of the melt and the air boundaries with the crucible.

As before, we introduce small perturbations of $\epsilon \xi(s)$ and $\epsilon \eta(s)$ into $x(s)$ and $y(s)$. These perturbations are fixed at $s_t$ such that $\xi(s_t) = \eta(s_t) = 0$. Using the definition in Eq. (3), the first variation in energy is given by

$$\delta U = \int_{s_t}^{s} \left( \xi g_x + \xi' g_{x'} + \eta g_y + \eta' g_{y'} \right) ds + \delta r (\gamma_2 - \gamma_1),$$

where $|\delta r| = \sqrt{\xi(0)^2 + \eta(0)^2}$. Integrating by parts, we arrive at the following form of the first variation,

$$\delta U = \int_{s_t}^{s} \left[ \xi \left( g_x - \frac{d}{ds} g_{x'} \right) + \eta \left( g_y - \frac{d}{ds} g_{y'} \right) \right] ds - \xi g_x' \bigg|_0 - \eta g_y' \bigg|_0 + \delta r (\gamma_2 - \gamma_1).$$

Setting the integrand to zero gives us the Euler-Lagrange equation for optimality. Substituting the expressions for $g_{x'}$ and $g_{y'}$ give us

$$\delta U = -\gamma \frac{\xi x' + \eta y'}{\sqrt{x'^2 + y'^2}} \bigg|_0 + \delta r (\gamma_2 - \gamma_1).$$

Figure 10: For the case of variable end points, the perturbation at the origin is considered to be $\delta r$. The aim is to find conditions on the meniscus shape such that $\delta r = 0$ is a minima.

The first term can be interpreted as a dot product between $\delta r = [\xi(0), \eta(0)]$ and $\vec{t} = [x'(0), y'(0)]$, which can be written in terms of the cosine of the angle between them.

$$\delta U = \begin{cases} \gamma \delta r \left( -\cos(\theta) + \frac{2\gamma_2 - \gamma_1}{\gamma} \right) & \delta r > 0 \\ \gamma \delta r \left( \cos(2\pi - \phi - \theta) + \frac{2\gamma_2 - \gamma_1}{\gamma} \right) & \delta r < 0 \end{cases}$$

We see that the first variation is minimized and becomes zero at $\delta r = 0$ when

$$\theta_e \leq \theta \leq \pi - \phi + \theta_e,$$

$$\theta_e = \arccos \left( \frac{\gamma_2 - \gamma_1}{\gamma} \right).$$
Figure 11: The shaded region displays the range of pinning angles for the inner and the outer edges. The figure shows how the starting position of the meniscus would change as the contact angle is varied.

This range of $\theta$ values is known as the Gibbs pinning condition and is illustrated by the shaded region in Fig. 10. A corollary to the Gibbs pinning condition is that if we set $\phi = \pi$ we arrive at the Young-Dupre contact angle condition.

When $\theta \leq \theta_e$ the meniscus recedes horizontally along the crucible boundary until it gets pinned to the inner corner of the crucible. This can provide an extended range of pinning angles as illustrated by the shaded region in Fig. 11. The overall range for the pinning conditions can be derived using a similar analysis.

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