

1 **MOVING BOUNDARY PROBLEMS FOR QUASI-STEADY**
2 **CONDUCTION LIMITED MELTING***

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5 **Abstract.** The problem of melting a crystal dendrite is modelled as a quasi-steady Stefan
6 problem. By employing the Baiocchi transform, asymptotic results are derived in the limit that
7 the crystal melts completely, extending previous results that hold for a special class of initial and
8 boundary conditions. These new results, together with predictions for whether the crystal pinches off
9 and breaks into two, are supported by numerical calculations using the level set method. The effects of
10 surface tension are subsequently considered, leading to a canonical problem for near-complete-melting
11 which is studied in linear stability terms and then solved numerically. Our study is motivated in
12 part by experiments undertaken as part of the Isothermal Dendritic Growth Experiment, in which
13 dendritic crystals of pivalic acid were melted in a microgravity environment: these crystals were
14 found to be prolate spheroidal in shape, with an aspect ratio initially increasing with time then
15 rather abruptly decreasing to unity. By including a kinetic undercooling-type boundary condition in
16 addition to surface tension, our model suggests the aspect ratio of a melting crystal can reproduce
17 the same non-monotonic behaviour as that which was observed experimentally.

18 **Key words.** conduction-limited melting, melting in microgravity, moving-boundary problem,
19 surface tension, extinction, formal asymptotics, level set method.

20 **AMS subject classifications.** 35R37, 80A22, 65M99

21 **1. Introduction.** While there is a variety of simple models to approximate the
22 shape of a melting particle [33, 38], the traditional approach from a mathematical
23 perspective is to employ a Stefan problem, which involves the linear heat equation
24 subject to appropriate boundary conditions on the solid-melt interface. These moving
25 boundary problems are well studied via rigorous analysis, asymptotic techniques, some
26 exact solutions and numerical computation. Almost all of the analytical progress has
27 been made for one-dimensional problems or those with radial symmetry [23, 39, 47,
28 48, 55], although there have been successful studies in which the symmetry is broken
29 [37, 44, 46, 56]. We continue this direction in the present study, focusing on the
30 melting of an axially symmetric dendritic crystal. We employ both analytical and
31 numerical techniques to study the shape of the evolving crystal, focussing on the very
32 final stages of melting.

33 A key aspect of a traditional Stefan problem is that the effects of convection
34 are ignored. An excellent example of a relevant physical application involves certain
35 experiments undertaken on the space shuttle Columbia, as part of the so-called Iso-
36 thermal Dendritic Growth Experiment (IDGE) [21, 22, 43], in which convection is
37 not an issue. The conduction-limited melting that was studied in those experiments
38 provides a physical motivation for the kind of theoretical Stefan problems considered
39 here. A brief summary of these experiments is as follows. A pure liquid melt, pivalic
40 acid, is held at a temperature $u^* > u_m^*$, where $u_m^* \approx 35.9$ °C is the equilibrium melting
41 temperature. The temperature is then reduced to slightly supercool the melt so that
42 $u \lesssim u_m$ throughout. The growth of dendrites is initiated by activating a thermoelec-
43 tric cooler to chill a small isolated volume of the melt, leading to a dendritic mushy

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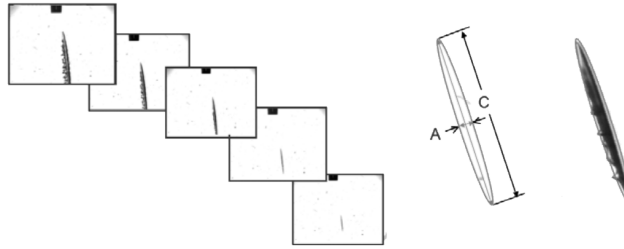


Fig. 1: Left: A sequence of video frames of melting ellipsoidal PVA crystal collected as part of the Isothermic Dendritic Growth experiment. Right: Digital analysis of the middle frame on the left. The major, C , and minor, A , axis were computed using automated edge detection software to approximate the aspect ratio as a function of time. The black tip of the glass injector at the top of each frame has a diameter of 1 mm. Reproduced from Glicksman et al. [22] with permission from Springer Nature.

44 zone. Finally, the temperature is raised to remelt the crystals, returning the system
 45 to a stable melt phase.

46 We are particularly interested in the final component of the IDGE. After sufficient
 47 melting of the mushy zone had occurred, the remaining fragments consisted of isolated
 48 crystallites that resulted from partially melting dendritic side branches. Typically
 49 these were roughly prolate spheroidal in shape (see Figure 1). For the final minute
 50 of melting of a particular crystal, video data (filmed at 30 frames per second) was
 51 analysed to determine the aspect ratio at each time. For the examples presented
 52 by Glicksman and co-workers [22, 43], the aspect ratio of the needle-shaped crystals
 53 increased with time from about 7 at $t_e - t = 60$ s to 17 at $t_e - t = 10$ s, where
 54 t_e is the final melting time (also referred to as the extinction time). After about
 55 $t_e - t = 10$ s, the aspect ratio rapidly decreased, and appeared to approach unity as
 56 $t \rightarrow t_e^-$, meaning that the crystals were spherical just before extinction.

57 In order to make analytical progress, Glicksman et al. [21] model the process
 58 with a one-phase quasi-steady problem, which results by ignoring heat conduction
 59 within the crystals and assuming an infinite Stefan number. Here, the Stefan number
 60 is defined by

$$61 \quad (1.1) \quad \beta = \frac{L}{c(u_\infty^* - u_m^*)},$$

62 where c is the specific heat, L the latent heat of fusion per mass and $u_\infty^* - u_m^*$
 63 is the temperature difference between the melt away from the crystal and the melting
 64 temperature. In reality, for this particular experiment the parameter values were
 65 $L/c \approx 10.99$ K, $u_\infty^* - u_m^* \approx 1.8$ K, so $\beta \approx 6.1$, which is not reasonably large. Glicksman
 66 et al. [21] derive an exact solution to the infinite-Stefan-number problem in an infinite
 67 domain in prolate spheroidal coordinates, which applies under the further assumption
 68 that the aspect ratio of the dendrite remains constant. This solution is a special
 69 case of that presented earlier by Ham [25] and Howison [28] (which holds for the
 70 more general shape of an ellipsoid with constant aspect ratios), for example, and that
 71 derived using the Baiocchi transform by McCue et al. [45] (again, for an ellipsoid).
 72 The solution was used by Glicksman et al. [21] to approximate the time-dependence
 73 of the melting process, with quite good agreement with experimental results.

74 Glicksman and co-workers [21, 22, 43] did not provide an explanation for the ob-
 75 served increase in aspect ratio during the first 50 s of melting; however, the subsequent
 76 decrease in aspect ratio (during the final 10 s of melting) was accounted for by noting
 77 that by this stage of the melting process the crystals had become small enough for
 78 surface tension effects to begin to dominate [22, 43]. As a consequence, the needle
 79 tips with high curvature melted more quickly than the remainder of the crystals, in
 80 accordance with the Gibbs-Thomson law

$$81 \quad (1.2) \quad u^* = u_m^*(1 - \gamma\kappa^*) \quad \text{on} \quad \partial\Omega^*,$$

82 which states that the actual melting temperature on a curved surface is not constant,
 83 but instead depends weakly on the mean curvature κ^* (defined to be positive for a
 84 sphere) via the surface tension coefficient γ (defined to be $\gamma = 2\sigma^*/\rho_s L$, where σ^*
 85 measures surface energy effects with dimensions Nm^{-1} or Jm^{-2} and ρ_s is the density
 86 of the solid phase) [3]. Here $\partial\Omega^*$ denotes the solid-melt interface. For the IDGE
 87 experiments, the surface tension coefficient is roughly $\gamma \sim 10^{-10}$ m.

88 In this article, we are motivated by these issues to undertake a theoretical study
 89 of the one-phase quasi-steady Stefan problem. The mathematical problem is re-
 90 formulated in Section 2 with a Baiocchi transform for the special zero-surface-tension
 91 case. In Section 3, we go on to provide a near extinction analysis for a general shaped
 92 initial crystal, including numerical results for cases in which crystals ultimately melt
 93 to a single point or pinch off and break into two separate pieces. The role of surface
 94 tension is then explored in Section 4, while in Section 5 we consider an additional
 95 effect on the moving boundary, kinetic undercooling. We show that kinetic under-
 96 cooling acts as a de-stabilising term, and is effectively in competition with surface
 97 tension. When these two terms are considered simultaneously, we find that the aspect
 98 ratio of a prolate spheroid can initially increase before decreasing suddenly to unity in
 99 the extinction limit, which is the same behaviour as observed in the IDGE. We close
 100 in Section 6 with a summary of the key results and a brief discussion of how our work
 101 relates to the experiments described by Glicksman and co-workers [21, 22, 43]. An
 102 important point to note is that the quasi-steady assumption used in this article leads
 103 to a moving boundary problem that also describes bubble contraction in a porous
 104 medium [12, 28, 45]. Thus our study also describes the effect that surface tension has
 105 on the shape of a bubble in the limit that it contracts to a point. This connection is
 106 revisited in Section 6.

107 2. Quasi-steady formulation with zero surface tension.

108 **2.1. Governing equations.** Consider a solid substance (the crystal dendrite),
 109 initially at melting temperature u_m^* occupying the region $\Omega^*(0)$, surrounded by the
 110 same substance in liquid form in $\mathbb{R}^3 \setminus \Omega^*$. In the far field, a higher temperature u_∞^*
 111 is applied, and thus melting proceeds until the crystal melts completely at the extinction
 112 time t_e^* .

113 Setting k to be the thermal diffusivity, we scale variables using

$$114 \quad (2.1) \quad t = \frac{k}{\ell^2 \beta} t^*, \quad \mathbf{x} = \frac{1}{\ell} \mathbf{x}^*, \quad u = \frac{u^* - u_m^*}{u_\infty^* - u_m^*},$$

115 where ℓ is a characteristic length scale of the initial crystal shape, and β is the Stefan

116 number (1.1). The resulting one-phase Stefan problem for melting the crystal is

$$117 \quad (2.2a) \quad \text{in } \mathbb{R}^3 \setminus \Omega(t) : \quad \frac{1}{\beta} \frac{\partial u}{\partial t} = \nabla^2 u,$$

$$118 \quad (2.2b) \quad \text{on } \partial\Omega : \quad u = 0,$$

$$119 \quad (2.2c) \quad \text{on } \partial\Omega : \quad V_n = -\frac{\partial u}{\partial n},$$

$$120 \quad (2.2d) \quad \text{as } r \rightarrow \infty : \quad u \rightarrow 1,$$

122 where V_n represents the normal velocity of the solid-melt interface $\partial\Omega$, defined to be
123 negative for a shrinking surface.

124 For what follows we shall take the quasi-steady limit $\beta = \infty$, which is an appro-
125 priate approximation for experiments in which the latent heat is large or the specific
126 heat is small. As a result, the parabolic equation (2.2a) becomes Laplace's equation

$$127 \quad (2.2e) \quad \text{in } \mathbb{R}^3 \setminus \Omega(t) : \quad \nabla^2 u = 0,$$

129 and thus we do not require an initial condition for u .

130 As mentioned in the Introduction, the governing equations (2.2e) with (2.2b)-
131 (2.2d) are also relevant for the problem of a bubble that is forced to contract in a
132 saturated medium, where the fluid flow is governed by Darcy's law [12, 28, 45], as
133 well as the two-dimensional analogue for Hele-Shaw flow [15, 14, 42]. These equations
134 also arise in other moving boundary problems, for example the small Péclet num-
135 ber limit of advection-diffusion-limited dissolution/melting models [6, 27, 32, 53, 57],
136 for which it is also of interest to track the moving boundary and predict its shape
137 and location (the collapse point [53]) close to the extinction time; other closely re-
138 lated advection-diffusion-like moving boundary problems in potential flow have similar
139 governing equations in the small Péclet number limit [4, 7].

140 **2.2. Baiocchi transform.** We use the Baiocchi transform defined by

$$141 \quad (2.3a) \quad \text{in } \mathbb{R}^3 \setminus \Omega(0) : \quad w = \int_0^t u(\mathbf{x}, t') dt'$$

$$142 \quad (2.3b) \quad \text{in } \Omega(0) \setminus \Omega(t) : \quad w = \int_{\omega(\mathbf{x})}^t u(\mathbf{x}, t') dt',$$

144 where we are using the notation $t = \omega(\mathbf{x})$ to denote the solid-melt interface $\partial\Omega$. The
145 Baiocchi transform is widely used in the analysis of moving boundary problems with
146 boundary conditions of the form (2.2b)-(2.2c), for example [8, 13, 31, 36, 40, 45]. Note
147 that while here we restrict ourselves to (2.2e), the approach is also applicable to (2.2a)
148 [44, 46].

149 Transforming the governing equations (2.2e) with (2.2b)-(2.2d), we derive the
150 nonlinear moving boundary problem for w :

$$151 \quad (2.4a) \quad \text{in } \mathbb{R}^3 \setminus \Omega(0) : \quad \nabla^2 w = 0,$$

$$152 \quad (2.4b) \quad \text{in } \Omega(0) \setminus \Omega(t) : \quad \nabla^2 w = 1,$$

$$153 \quad (2.4c) \quad \text{on } \partial\Omega : \quad w = 0,$$

$$154 \quad (2.4d) \quad \text{on } \partial\Omega : \quad \frac{\partial w}{\partial n} = 0,$$

$$155 \quad (2.4e) \quad \text{as } r \rightarrow \infty : \quad w \rightarrow t.$$

157 Once a solution for the Baiocchi variable w is determined, the temperature u can be
 158 recovered via $u = \partial w / \partial t$. We note that an advantage of the Baiocchi transform is
 159 that it transforms the inhomogeneous boundary condition (2.2c) into a homogeneous
 160 boundary condition. Another is that time appears as a parameter in (2.4a)-(2.4e),
 161 so that the problem can be solved at any time without knowledge of the solution at
 162 previous times.

163 **2.3. Exact solution for prolate spheroid.** For the case in which the initial
 164 crystal shape $\partial\Omega(0)$ is an ellipsoid, (2.4a)-(2.4e) can be solved in ellipsoidal coordi-
 165 nates exactly, as done as part of the analysis by McCue et al. [45]. The solution for
 166 the interface $\partial\Omega(t)$ remains ellipsoidal with constant aspect ratios for all time un-
 167 til extinction. An equivalent solution without the Baiocchi transform is provided in
 168 Howison [28].

169 We present here a summary of this exact solution in the special case for which
 170 the initial crystal shape $\partial\Omega(0)$ is the prolate spheroid

$$171 \quad (2.5) \quad x^2 + y^2 + \frac{z^2}{z_0(0)^2} = 1,$$

172 with initial aspect ratio $\mathcal{A}(0) = z_0(0)$. (This special case, together with the case in
 173 which the crystal is initially an oblate spheroid, is also recorded by McCue et al. [45].)
 174 The exact solution is that $\partial\Omega(t)$ retains its prolate spheroidal shape as

$$175 \quad (2.6) \quad \frac{x^2 + y^2}{\rho_0(t)^2} + \frac{z^2}{z_0(t)^2} = 1,$$

176 where $z_0(t) > 0$ and $\rho_0(t) > 0$ measure the major and minor axes of the dendrite,
 177 respectively, with constant aspect ratio $\mathcal{A}(t) = z_0(t)/\rho_0(t) = z_0(0)$ (here the length
 178 scale ℓ is chosen so that $\rho_0(0) = 1$). The full solution has the time-dependence

$$179 \quad (2.7) \quad \frac{z_0(t)}{z_0(0)} = \rho_0(t) = \sqrt{1 - \frac{t}{t_e}},$$

180 where

$$181 \quad (2.8) \quad t_e = \frac{z_0(0)}{4\sqrt{z_0(0)^2 - 1}} \ln \left(\frac{z_0(0) + \sqrt{z_0(0)^2 - 1}}{z_0(0) - \sqrt{z_0(0)^2 - 1}} \right).$$

182 The result (2.8) is also derived in Glicksman et al. [21]. Although, as mentioned
 183 above, the aspect ratio of the melting crystals in the Isothermal Dendritic Growth
 184 Experiment was not constant, these authors make a rough guess for the average value
 185 of the aspect ratio over the first 50 seconds of melting, and then compare (2.7) with
 186 experimental results. Their agreement is quite good, reflecting the square root of time
 187 dependence near extinction.

188 **3. Analysis of zero-surface-tension problem.** McCue et al. [45] were con-
 189 cerned primarily with analysing the near extinction behaviour for a variation of (2.4a)-
 190 (2.4e) in which $\Omega(0)$ coincides with an outer boundary (i.e., a finite-domain problem
 191 in which the crystal initially occupies the entire domain). Here we provide equiva-
 192 lent results for the full infinite-domain problem (2.4a)-(2.4e) and apply the level set
 193 method to support these findings.

194 **3.1. Extinction time and extinction points.** For a given initial crystal shape
 195 $\Omega(0)$, we wish to determine how long it takes to melt (the extinction time t_e) and the
 196 point at which the crystal eventually vanishes as $t \rightarrow t_e^-$ (the extinction point \mathbf{x}_e). The
 197 convenient framework for this analysis is via the Baiocchi transform. As mentioned
 198 above, time appears as a parameter in (2.4a)-(2.4e), meaning we can skip to the
 199 extinction time to compute $w_e(\mathbf{x}) = w(\mathbf{x}, t_e)$. It is convenient to set $w_e = W(\mathbf{x}) + t_e$,
 200 so W satisfies the linear problem

$$201 \quad (3.1a) \quad \text{in } \mathbb{R}^3 \setminus \Omega(0) : \quad \nabla^2 W = 0,$$

$$202 \quad (3.1b) \quad \text{in } \Omega(0) : \quad \nabla^2 W = 1,$$

$$203 \quad (3.1c) \quad \text{as } r \rightarrow \infty : \quad W \rightarrow 0.$$

205 The extinction point \mathbf{x}_e is then the local minimum of W , and the extinction time is
 206 recovered via $t_e = -W(\mathbf{x}_e)$. As noted by Entov & Etingof [15], (3.1a)-(3.1c) defines
 207 the dimensionless gravity potential of $\Omega(0)$, thus

$$208 \quad (3.2) \quad W = -\frac{1}{4\pi} \iiint_{\Omega(0)} \frac{1}{|\mathbf{x} - \mathbf{x}'|} dV',$$

209 which provides an interesting connection between our problem and gravity potential
 210 generated by a uniform body.

211 Whilst in practice it is not feasible to compute W analytically for a general initial
 212 crystal shape $\Omega(0)$, such a calculation can be performed numerically. Indeed, we
 213 provide a number of simple examples in Subsection 3.4 in which we compute W for
 214 both convex and non-convex initial shapes. We include in those examples cases for
 215 which W has two local minima. In such instances, if the two local minima are equal,
 216 then the crystal must pinch off into two, with the local minima corresponding to the
 217 extinction points for each of the two satellite crystals. We also provide an example
 218 of the more complicated case in which there are two local minima that are not equal;
 219 here, the use of W can only predict the final extinction for the largest of the two
 220 satellite crystals.

221 **3.2. Near-extinction analysis.** For the case of an axially symmetric initial
 222 crystal with the z axis pointing down the centreline, we can translate the coordinate
 223 system so that the extinction point \mathbf{x}_e lies on the origin. Since $w_e = 0$ at $\mathbf{x} = \mathbf{x}_e$
 224 and \mathbf{x}_e is a local minimum of w_e , a simple Taylor series for this axially symmetric
 225 geometry implies that $w_e \sim a(x^2 + y^2) + bz^2$ as $r \rightarrow 0$. Further, as a consequence of
 226 (3.1b), we then have

$$227 \quad (3.3) \quad w_e \sim a(x^2 + y^2) + \left(\frac{1}{2} - 2a\right) z^2 \quad \text{as } r \rightarrow 0,$$

228 where $1/6 < a < 1/4$. As we shall see, the parameter a is effectively all the melting
 229 crystal “remembers” from its initial condition; it is this single parameter that controls
 230 the aspect ratio of the crystal at extinction. Note that the higher order terms in (3.3)
 231 are not required in the following analysis (they would be for the special case $a = 1/4$,
 232 which represents the borderline between the type of extinction considered in this
 233 section and when a bubble breaks up into two, as treated in Subsection 3.4).

234 In the limit $t \rightarrow t_e^-$, the inner region is for $r = \mathcal{O}(T)$, where $T(t)$ is a length scale
 235 defined so that the volume of the melting crystal is fixed to be $4\pi T^3/3$. We write

236 $w \sim T^2 \Phi(\mathbf{X})$ as $t \rightarrow t_e^-$, where $\mathbf{X} = \mathbf{x}/T$, so that

237 (3.4a) $\quad \text{in } \mathbb{R}^3 \setminus \Omega_0(0) : \quad \frac{\partial^2 \Phi}{\partial X^2} + \frac{\partial^2 \Phi}{\partial Y^2} + \frac{\partial^2 \Phi}{\partial Z^2} = 1,$

238 (3.4b) $\quad \text{on } \partial\Omega_0 : \quad \Phi = 0, \quad \frac{\partial \Phi}{\partial N} = 0,$
 239

240 where Ω_0 denotes the crystal which has volume $4\pi/3$ in these self-similar coordinates,
 241 and N denotes a normal direction. In order to match with (3.3) we require that

242 (3.4c) $\quad \Phi \sim a(X^2 + Y^2) + \left(\frac{1}{2} - 2a\right) Z^2 - d + \frac{1}{3R},$

243 as $R \rightarrow \infty$, where d is a constant found as part of the solution to (3.4a)-(3.4c). We
 244 see from (3.4c) that a matching condition for the outer region is

245 (3.5) $\quad w \sim a(x^2 + y^2) + \left(\frac{1}{2} - 2a\right) z^2 - dT^2 + \frac{T^3}{3r} \quad \text{as } r \rightarrow 0.$

246 The solution to (3.4a)-(3.4c) in prolate spheroidal coordinates is provided in [Ap-](#)
 247 [pendix A](#). According to this solution the dendrite boundary $\partial\Omega_0$ is described by

248 (3.6) $\quad \frac{X^2 + Y^2}{q_0^2 - 1} + \frac{Z^2}{q_0^2} = \frac{1}{q_0^{2/3}(q_0^2 - 1)^{2/3}},$

249 where q_0 is a parameter that is related to the special constant a by

250 (3.7) $\quad a = \frac{1}{4}q_0^2 - \frac{1}{8}q_0(q_0^2 - 1) \ln \left(\frac{q_0 + 1}{q_0 - 1} \right).$

251 Further, the constant d in (3.4c) is related implicitly to a by

252 (3.8) $\quad d = \frac{1}{4}q_0^{1/3}(q_0^2 - 1)^{1/3} \ln \left(\frac{q_0 + 1}{q_0 - 1} \right).$

253 Note that the prolate spheroid approaches a perfect sphere in the limit $a \rightarrow 1/6^+$, in
 254 which case $d \rightarrow 1/2^+$.

255 The outer region is for $r = \mathcal{O}(1)$, for which

256 (3.9) $\quad w \sim w_e - (t - t_e) + \frac{T^2}{3r} \quad \text{as } t \rightarrow t_e^-.$

257 Matching with the inner gives the time-dependence

258 (3.10) $\quad t = t_e - dT^2 + \mathcal{O}(T^5) \quad \text{as } T \rightarrow 0,$

259 or, in other words,

260 (3.11) $\quad T \sim \frac{1}{\sqrt{d}}(t_e - t)^{1/2} \quad \text{as } t \rightarrow t_e^-.$

261 Thus we see that, regardless of the shape of the initial crystal, the square root of time
 262 scaling determined experimentally in Glicksman et al. [21] is as expected.

263 In summary, the zero-surface-tension model predicts that, provided there is no
 264 pinch-off, an axially symmetric dendrite will melt to a spheroid in the extinction limit.
 265 While this spheroid could be prolate or oblate, we concentrate here on the prolate
 266 case, as this is the one observed in the IDGE [21, 22, 43]. The aspect ratio of the
 267 prolate spheroid at extinction is given by

$$268 \quad (3.12) \quad \mathcal{A}(t_e) = \frac{q_0}{\sqrt{q_0^2 - 1}},$$

269 which provides an implicit dependence of \mathcal{A} on the constant a via (3.7). Here a is
 270 the only parameter that is required to characterise the initial dendrite shape (it is
 271 found by solving (3.2) and expanding w_e about \mathbf{x}_e). The time-dependence of the
 272 melting is given by (3.11), where the volume of the dendrite shrinks like $4\pi T^3/3$ (in
 273 other words, T provides a natural length scale for the melting dendrite). Again, this
 274 time-dependence is related to the initial dendrite shape via the parameter a (since d
 275 is given by a through (3.8) and (3.7)).

276 In the special case in which the dendrite is initially the prolate spheroid (2.5), then
 277 it retains its aspect ratio. This is, of course, the exact solution listed in Subsection 2.3.

278 Finally, for sufficiently symmetric crystals we have $a = 1/6$ which gives $d = 1/2$.
 279 Here $\Phi = R^2/2 - 1/2 + 1/3R$ and the dendrite becomes spherical in the limit with
 280 $T \sim \sqrt{2}(t_e - t)^{1/2}$. The special case of an initially spherical dendrite remains spherical.

281 At this point it is worth mentioning that for large Stefan numbers, $\beta \gg 1$, the
 282 scaling (3.11) eventually ceases to hold for the full classical Stefan problem with (2.2a)
 283 instead of (2.2e) [46]. However, this discrepancy would not be observed on the scale
 284 of the IDGE experiments.

285 **3.3. Null quadrature domains.** It is worth relating some of the above ar-
 286 guments to well-known and long-established results [12, 18, 28]. First, by applying
 287 Green’s theorem it can be shown that

$$288 \quad (3.13) \quad \frac{d}{dt} \iiint_{\mathbb{R}^3 \setminus \Omega(t)} \Phi(\mathbf{x}) \, dV = 0,$$

289 where Φ is a suitable harmonic function and $\Omega(t)$ is the shape of a melting crystal
 290 from the infinite-domain problem (2.4a)-(2.4e) (Howison [28]). Noting that these
 291 quasi-steady problems with zero surface tension are time-reversible, we can seek so-
 292 called ‘ancient’ solutions for which the entire domain $\mathbb{R}^3 \setminus \Omega(t)$ vanishes in the limit
 293 $t \rightarrow -\infty$. From (3.13) it follows that for these ancient solutions $\mathbb{R}^3 \setminus \Omega(t)$ must
 294 be a null quadrature domain. The only suitable such domain is the exterior of an
 295 ellipsoid (see Karp [34] for a discussion on null quadrature domains). For any other
 296 initial crystal shape, the backwards problem with t decreasing leads to some kind
 297 of finite-time blow-up or perhaps a scenario in which part of the crystal boundary
 298 expands infinitely leaving behind ‘fjords’ or ‘tongues’ (these scenarios are much better
 299 understood in the two-dimensional Hele-Shaw problem; see also Howison [29, 30] for
 300 explicit examples of each case).

301 As discussed in Section 3, for a melting crystal (of general initial shape) the
 302 generic limiting behaviour is that it becomes ellipsoidal in shape as $t \rightarrow t_e^-$. This
 303 result can also be derived using an alternative approach, as suggested more recently
 304 by King & McCue [36], who treated the two-dimensional Hele-Shaw case. First, we
 305 see that for the integral in (3.13) to converge we could choose $\Phi = r^\ell Y_\ell^m$, where Y_ℓ^m
 306 are spherical harmonics and ℓ is an integer such that $\ell \leq -4$. Rescaling lengths such

307 that $\bar{r} = r/T$, we have from (3.13) that

$$308 \quad (3.14) \quad \iiint_{\mathbb{R}^3 \setminus \bar{\Omega}(t)} \Phi(\bar{\mathbf{x}}) d\bar{V} = \mathcal{O}(T^{-\ell-3}) \quad \text{as } T \rightarrow 0 \quad \text{for } \ell \leq -4.$$

309 Thus, the left-hand side vanishes as $T \rightarrow 0$, or $t \rightarrow t_e^-$, meaning that the exterior of
 310 the crystal approaches a null quadrature domain in the limit, and thus the crystal
 311 itself approaches an ellipsoid in shape.

312 **3.4. Numerical examples.** We present some numerical examples that demon-
 313 strate the key features discussed above. To solve (3.1a)-(3.1c) numerically, we formu-
 314 late a level set function, $\phi(\mathbf{x})$, such that $\phi > 0$ for $\mathbf{x} \in \Omega(0)$ and $\phi < 0$ for $\mathbf{x} \in \mathbb{R}^3 \setminus \Omega(0)$.
 315 Thus we can reformulate (3.1a) and (3.1b) as

$$316 \quad (3.15) \quad \nabla^2 W = H(\phi),$$

317 where H is the Heaviside function. We note that $H(\phi)$ is discontinuous at $\mathbf{x} \in \partial\Omega(0)$,
 318 so for numerical purposes we implement a smoothed Heaviside function

$$319 \quad (3.16) \quad \hat{H}(\phi) = \begin{cases} 0 & \text{if } \phi < -\delta, \\ \frac{1}{2} \left(1 + \frac{\phi}{\delta} + \frac{1}{\pi} \sin \frac{\pi\phi}{\delta} \right) & \text{if } |\phi| \leq \delta, \\ 1 & \text{if } \phi > \delta, \end{cases}$$

320 where $\delta = 1.5\Delta x$. For this purpose, it is convenient to work in spherical polar
 321 coordinates (r, θ, φ) and represent the axially symmetric moving boundary $\partial\Omega$ by
 322 $r = s(\theta, t)$. Thus, (3.15) becomes

$$323 \quad (3.17) \quad \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\partial W}{\partial r} \right) + \frac{1}{r^2 \sin \theta} \frac{\partial}{\partial \theta} \left(\sin \theta \frac{\partial W}{\partial \theta} \right) = \hat{H}(\phi).$$

325 The spatial derivatives in (3.17) are approximated using central finite differencing,
 326 with homogeneous Neumann boundary conditions applied at $r = 0$, $\theta = 0$, and $\theta = \pi$.
 327 The far-field boundary condition (3.1c) is incorporated using a Dirichlet-to-Neumann
 328 map described in Appendix B.2.2.

329 **3.4.1. Symmetric initial condition.** We consider a selection of initial condi-
 330 tions to illustrate a few different qualitative behaviours. Again, using spherical polar
 331 coordinates (r, θ, φ) with $\partial\Omega$ denoted by $r = s(\theta, t)$, the first is the prolate spheroid

$$332 \quad (3.18) \quad s(\theta, 0) = \frac{r_0}{\sqrt{r_0^2 \cos^2 \theta + \sin^2 \theta}},$$

333 where r_0 describes the initial aspect ratio. The second initial condition is a peanut-
 334 shaped interface described by

$$335 \quad (3.19) \quad s(\theta, 0) = r_0 + (1 - r_0) \cos^2 \theta,$$

336 where r_0 can be interpreted as a measure of the depth of the pinch in the middle of
 337 the peanut. Following Garzon et al. [19], the third initial condition considered is a
 338 dumbbell shaped interface of the form $s(\theta, 0) = (\rho^*(\theta)^2 + z^*(\theta)^2)^{1/2}$, where

$$339 \quad (3.20a) \quad z^*(\theta) = 1 + r_0 \sin^2(\theta/2),$$

$$340 \quad (3.20b) \quad \rho^*(\theta) = g(\theta) + 2g(\pi - \theta),$$

342 with

$$343 \quad (3.20c) \quad g(\theta) = \sqrt{r_0 k(\theta)} \left(e^{-(r_0^2 k(\theta)^2)/2} - e^{-r_0^2/2} \right),$$

$$344 \quad (3.20d) \quad k(\theta) = \cos^2(\theta/2),$$

346 for $0 \leq \theta \leq \pi/2$; for $\pi < \theta \leq 2\pi$ this initial condition is made symmetric by reflecting
347 about $\theta = \pi/2$.

348 In [Figure 2](#), we illustrate some numerical results by choosing parameter values
349 from these three initial conditions. For the prolate spheroid [\(3.18\)](#) we provide results
350 for $r_0 = 0.8$, noting that this initial condition is obviously convex. For the peanut
351 shaped surface [\(3.19\)](#), we choose $r_0 = 0.5$, which is not convex but is instead mean
352 convex. Finally, for the dumbbell shape [\(3.20a\)](#)-[\(3.20d\)](#), we choose $r_0 = 4.75$, which
353 again corresponds to a nonconvex shape which is still mean convex, but this time with
354 a particularly thin neck region. In all of these case, we show in [Figure 2](#) the initial
355 shape, the numerical solution to [\(2.2b\)](#)-[\(2.2e\)](#) shortly before the extinction time, and
356 the corresponding solution to the Baiocchi transform problem [\(3.1c\)](#) and [\(3.17\)](#).

357 For both of the first two examples in [Figure 2](#), namely [\(3.18\)](#) with $r_0 = 0.8$
358 and [\(3.19\)](#) with $r_0 = 0.5$, the solution to [\(2.2b\)](#)-[\(2.2e\)](#) contracts to a single point
359 at extinction. By observing the third column of [Figure 2](#), we see this is consistent
360 with the solution of [\(3.1c\)](#) and [\(3.17\)](#), which shows $|W|$ having one local maximum at
361 the origin, predicting one point at extinction. This comparison highlights that convex
362 shapes and some nonconvex shapes will contract to a single point. The extinction time
363 predicted by the Baiocchi transform is computed by evaluating $|W|$ at \mathbf{x}_e (which, for
364 this problem is the origin) giving the values $t_e = 0.370$ and $t_e = 0.233$ for [\(3.18\)](#)
365 with $r_0 = 0.8$ and [\(3.19\)](#) with $r_0 = 0.5$, respectively. Comparing this to the extinction
366 times computed from the numerical solution to [\(2.2b\)](#)-[\(2.2e\)](#), we find there is less than
367 0.1% relative difference, suggesting excellent agreement.

368 The equation [\(3.19\)](#) with $r_0 = 0.5$ provides a good test for the prediction [\(3.12\)](#).
369 For this purpose we take the solution to the Baiocchi transform problem [\(3.1\)](#), which
370 in this case predicts that $q_0 = 1.100$ and $a = 0.215$. As such, our prediction for the
371 aspect ratio at extinction is $\mathcal{A} = 2.395$. The time-dependent behaviour of the aspect
372 ratio for our numerical solution to the full problem (using the level set method) is
373 presented in [Figure 3](#). This figure demonstrates how well these two results agree with
374 other.

375 For initial condition [\(3.20a\)](#)-[\(3.20d\)](#) with $r_0 = 4.75$, [Figure 2](#) shows different qual-
376 itative behaviour. Here, we see that solutions to [\(2.2b\)](#)-[\(2.2e\)](#) will undergo pinch-off
377 and ultimately the two satellite crystals will contract to separate points of extinction.
378 Again, this is consistent with the solution to [\(3.1c\)](#) and [\(3.17\)](#) as the third column of
379 [Figure 2](#) indicates that $|W|$ has two local maxima. By approximating the locations
380 of these maxima and the values of $|W|$ at these points, we find the Baiocchi trans-
381 forms predicts that the interface will contract to extinction points at $z_e = \pm 0.577$
382 at time $t = 0.100$. Comparing these results with the extinction locations and times ap-
383 proximated from the numerical solution to [\(2.2b\)](#)-[\(2.2e\)](#), we find a relative difference
384 less than 0.2%. This example shows, for symmetric initial conditions, how well the
385 Baiocchi transform approach can be used to predict whether pinch-off will occur, as
386 well as the extinction points and time.

387 In summary, these numerical results indicate that for a given initial interface,
388 $\partial\Omega(0)$, each of the aspect ratio at extinction, the extinction time and location of the
389 extinction point for an interface evolving according to [\(2.2b\)](#)-[\(2.2e\)](#) can be predicted
390 from the solution to [\(3.1a\)](#)-[\(3.1c\)](#). Further, the indication is that this is true both

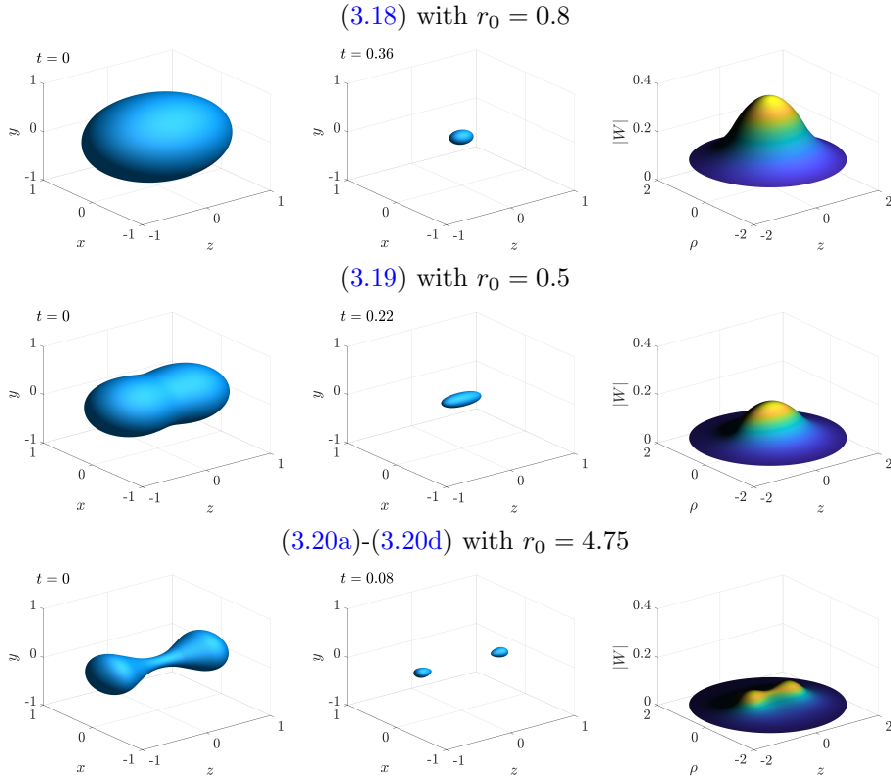


Fig. 2: Numerical solution to (2.2b)-(2.2e) with initial conditions of the form (3.18), (3.19), and (3.20a)-(3.20d), and the corresponding numerical solution to (3.1a)-(3.1c). Numerical solutions to (2.2b)-(2.2e) are computed using the level set based method described in Appendix B, while the numerical solution to (3.1c) and (3.17) is found using the procedure described in Subsection 3.4. Solutions are computed on the domain $0 \leq \theta \leq \pi$ and $0 \leq r \leq 2$ using 628×400 equally spaced nodes.

391 for interfaces that contract to a single point of extinction, or undergo pinch-off and
 392 contract to multiple points of extinction, at least for symmetric initial conditions.
 393 Finally, these results illustrate the capacity of the level set based numerical scheme,
 394 presented in Appendix B, to accurately describe the dynamics of the interface once a
 395 change in topology has occurred.

396 **3.4.2. Asymmetric initial condition.** The numerical solutions of (2.2b)-(2.2e)
 397 presented in Subsection 3.4.1 indicate that when $\partial\Omega(t)$ is sufficiently non-convex then
 398 the interface will undergo a change in topology. As initial conditions considered
 399 in Subsection 3.4.1 are symmetric along the major axis (about $\theta = \pi/2$), the two
 400 interfaces which form after pinch-off will have the same extinction time. We now
 401 investigate a class of asymmetric initial conditions that undergo pinch-off into two
 402 surfaces of differing volumes. We expect the smaller of the two volumes to contract
 403 to a point first, followed by the larger, thus giving two distinct extinction times.

404 We again consider an initial condition of the form of (3.20a)-(3.20d), but this
 405 time for $0 \leq \theta \leq \pi$. In Figure 4, we plot the time evolution of the numerical so-

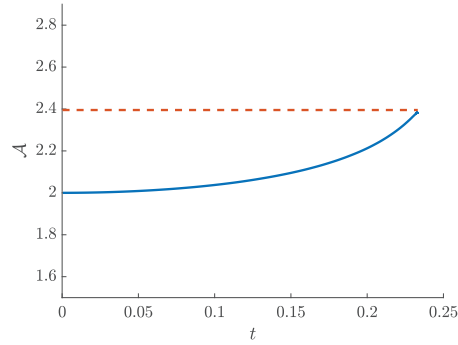


Fig. 3: The evolution of the aspect ratio for the example initial condition (3.19) with $r_0 = 0.5$ is presented as a solid (blue) curve. The (red) dashed curve is the predicted aspect ratio at extinction, given by (3.12). The agreement is quite good.

406 lution to (2.2b)-(2.2e) and the corresponding numerical solution to (3.1a)-(3.1c) for
 407 the representative case $r_0 = 5.1$. We observe that the full time-dependent solution to
 408 (2.2b)-(2.2e) undergoes a change in topology at approximately $t = 0.076$, with crystal
 409 domain $\Omega(t)$ pinching off into two. The smaller of the two satellite crystals contracts
 410 to a point at $z_e = 0.564$ when $t = 0.086$, followed by the remaining larger satellite
 411 crystal which contracts to a point at $z_e = -0.773$ when $t = 0.127$. The corresponding
 412 numerical solution to the Baiocchi transform problem (3.17), Figure 4 indicates that
 413 $|W|$ has two local maxima, located at $z_e = 0.506$ and $z_e = -0.767$, with $|W|$ equal to
 414 0.092 and 0.127 at these points, respectively. Thus we see that the predicted values of
 415 the extinction points and times agree well for the larger of the two satellite crystals (as
 416 it should) but not at all for the smaller crystal. That our approach can only provide
 417 information about the extinction time and point for the largest satellite crystal is a
 418 minor limitation to the Baiocchi transform framework.

419 **4. Effects of surface tension.** An inevitable consequence of melting a small
 420 crystal is that eventually the curvature will become large enough so that surface
 421 tension effects become important. For what follows, instead of (2.2b) we use the
 422 dimensionless version of the Gibbs-Thomson law (1.2), which is

$$423 \quad (4.1) \quad \text{on } \partial\Omega : \quad u = -\sigma\kappa,$$

424 where $\sigma = \gamma u_m^* / \ell(u_\infty^* - u_m^*)$ is the dimensionless surface tension coefficient, and κ is
 425 the dimensionless signed mean curvature.

426 **4.1. Linear stability analysis for near spherical crystal.** It proves use-
 427 ful to outline the linear stability analysis for interfaces evolving according to (2.2c)-
 428 (2.2e) and (4.1) with a near-spherical initial condition. In spherical polar coordinates
 429 (r, θ, φ) , we represent the axially symmetric moving boundary $\partial\Omega$ by $r = s(\theta, t)$, so

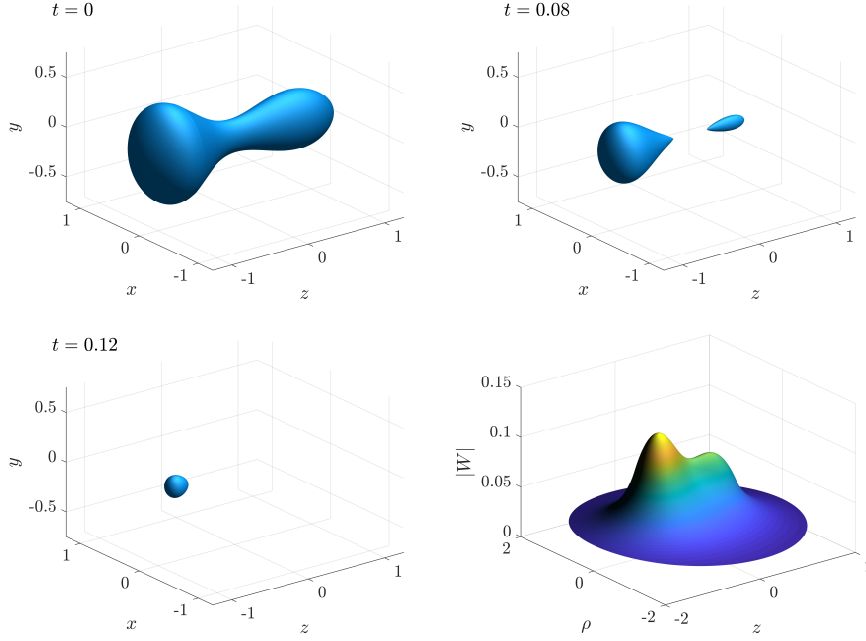


Fig. 4: Time evolution of the numerical solution to (2.2b)-(2.2e) (computed using the level set based method described in Appendix B), and corresponding numerical solution to (3.1a)-(3.1c) (found using the procedure described in Subsection 3.4). The initial condition is (3.20a)-(3.20d) with $r_0 = 5.1$. Solutions are computed on the domain $0 \leq \theta \leq \pi$ and $0 \leq r \leq 2$ using 628×400 equally spaced nodes.

430 that our problem is

$$431 \quad (4.2a) \quad \text{in } r > s : \quad 0 = \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\partial u}{\partial r} \right) + \frac{1}{r^2 \sin \theta} \frac{\partial}{\partial \theta} \left(\sin \theta \frac{\partial u}{\partial \theta} \right),$$

$$432 \quad (4.2b) \quad \text{on } r = s : \quad s_t = u_r - \frac{1}{s^2} u_\theta s_\theta,$$

$$433 \quad (4.2c) \quad \text{on } r = s : \quad u = \sigma \frac{3ss_\theta^2 - \cot \theta s_\theta^3 - s^2(s_{\theta\theta} + s_\theta \cot \theta) + 2s^3}{s(s^2 + s_\theta^2)^{3/2}},$$

$$434 \quad (4.2d) \quad \text{as } r \rightarrow \infty : \quad u \sim 1,$$

436 We seek a perturbed spherical solution to (4.2a)-(4.2d) of the form

$$437 \quad (4.3a) \quad u(r, \theta, \varphi, t) = u_0(r, t) + \varepsilon u_1(r, \theta, t) + \mathcal{O}(\varepsilon^2),$$

$$438 \quad (4.3b) \quad s(\theta, t) = s_0(t) + \varepsilon s_1(\theta, t) + \mathcal{O}(\varepsilon^2),$$

440 where $\varepsilon \ll 1$. The leading order solution is

$$441 \quad (4.4) \quad u_0 = 1 + \frac{2\sigma - s_0}{r}, \quad s_0 = \frac{8\sigma^2 \ln |(r_0 - 2\sigma)/(s_0 - 2\sigma)| + 2t + r_0(4\sigma + r_0)}{4\sigma + s_0}.$$

442 where $s_0(0) = r_0$. For the $\mathcal{O}(\varepsilon)$ system,

443 (4.5a) in $r > s_0$: $0 = \frac{\partial u_1}{\partial r} \left(r^2 \frac{\partial u_1}{\partial r} \right) + \frac{1}{\sin \theta} \frac{\partial}{\partial \theta} \left(\sin \theta \frac{\partial u_1}{\partial \theta} \right),$

444 (4.5b) on $r = s_0$: $\frac{\partial s_1}{\partial t} = \frac{\partial u_1}{\partial r} + s_1 \frac{\partial^2 u_0}{\partial r^2},$

445 (4.5c) on $r = s_0$: $u_1 + s_1 \frac{\partial u_0}{\partial r} = -\sigma \frac{2s_1 + \cot \theta \partial_\theta s_1 + \partial_\theta^2 s_1}{s_0^2},$

446 (4.5d) as $r \rightarrow \infty$: $u_1 \sim 0,$

448 the solutions are of the form

449 (4.6) $u_1(r, \theta, t) = \sum_{n=2}^{\infty} A_n r^{-n} P_n(\cos \theta), \quad s_1(\theta, t) = \sum_{n=2}^{\infty} \gamma_n(t) P_n(\cos \theta)$

450 where A_n is a sequence of unknown coefficients, P_n is the n th Legendre polynomial,
451 and γ_n is the n th mode of perturbation to the sphere. We are able to eliminate A_n
452 to obtain

453 (4.7) $\frac{1}{\gamma_n} \frac{d\gamma_n}{ds_0} = \frac{(n-1)((n^2 + 3n + 4)\sigma + s_0)}{s_0(s_0 + 2\sigma)}.$

454 Since $(1/\gamma_n)d\gamma_n/ds_0 \rightarrow 0$ in the limit that $s_0 \rightarrow 0$ for $n \geq 2$, we see that each mode of
455 perturbation is stable, and a perturbed sphere will evolve to a sphere in the extinction
456 limit, as expected.

457 The special case in which the initial condition is the prolate spheroid with major
458 and minor axes $r_0 + \varepsilon$ and r_0 , respectively, then

459
$$s(\theta, 0) = \frac{r_0(r_0 + \varepsilon)}{\sqrt{(r_0 \cos \theta)^2 + ((r_0 + \varepsilon) \sin \theta)^2}},$$

460 (4.8)
$$= r_0 + \varepsilon \left(\frac{1}{2} + \frac{2}{3} P_2(\cos \theta) \right) + \mathcal{O}(\varepsilon^2).$$

461

462 That is, $\gamma_2(0) = 2/3$ and $\gamma_n(0) = 0$ for $n \geq 3$. This initial condition has an aspect
463 ratio of $1 + \varepsilon/r_0 + \mathcal{O}(\varepsilon^2)$. The exact solution for γ_2 is

464 (4.9a)
$$\gamma_2 = \frac{2s_0^7}{3r_0^7} \left(\frac{r_0 + 2\sigma}{s_0 + 2\sigma} \right)^6,$$

465

466 and the aspect ratio for this particular initial condition therefore becomes

467 (4.10)
$$\mathcal{A}(s_0) = 1 + \varepsilon \frac{3\gamma_2}{2s_0} + \mathcal{O}(\varepsilon^2).$$

468 Note that when $\sigma = 0$, then $3\gamma_2/2s_0 = 1/r_0$, resulting in the aspect ratio remaining
469 constant, which is consistent with the known exact solution of [Subsection 2.3](#). Oth-
470 erwise, for $\sigma > 0$, the aspect ratio decreases monotonically to unity, as shown later in
471 [Figure 5](#).

472 **4.2. Long thin needle problem.** We consider here the limit of a long thin
 473 melting dendrite. Suppose the axially-symmetric shape of the dendrite is given by
 474 $\rho = S(z, t)$ where $\rho^2 = x^2 + y^2$. Suppose also that $S_0(z) = S(z, 0)$, $\rho_0(t) = S(0, t)$,
 475 $S(z_0(t), t) = 0$, where $\alpha = z_0(0)/\rho_0(0) \ll 1$ such that the initial aspect ratio, $\mathcal{A}(0) =$
 476 $1/\alpha$, is large.

477 The inner region is for $r = \mathcal{O}(\rho_0(t))$. Here the melting is almost two-dimensional
 478 with $\partial u/\partial z \ll 1$ and $\partial S/\partial z \ll 1$ so that, to leading order,

$$479 \quad (4.11a) \quad \text{in } \rho > S(z, t) : \quad \frac{\partial^2 u}{\partial \rho^2} + \frac{1}{\rho} \frac{\partial u}{\partial \rho} = 0,$$

$$480 \quad (4.11b) \quad \text{on } \rho = S(z, t) : \quad u = -\frac{\sigma}{\rho},$$

$$481 \quad (4.11c) \quad \text{on } \rho = S(z, t) : \quad \frac{\partial S}{\partial t} = -\frac{u}{\rho}.$$

483 The solution to (4.11a)-(4.11c) is

$$484 \quad (4.12) \quad u = -S \frac{\partial S}{\partial t} \ln(\rho/S),$$

485 where the form for S is determined by the missing far-field condition, which is found
 486 by considering the outer region.

487 In this outer region, which is for $r = \mathcal{O}(z_0(t))$, the dendrite appears as a slit. We
 488 scale $\tilde{\rho} = \rho/(\alpha\rho_0(t))$, $\tilde{t}/\ln \alpha$ and rewrite the inner solution (4.12) to be

$$489 \quad (4.13) \quad u = -S \frac{\partial S}{\partial \tilde{t}} - \frac{\sigma}{S} - S \frac{\partial S}{\partial \tilde{t}} \frac{\ln(\rho_0 \tilde{\rho}/S)}{\ln \alpha}.$$

490 The leading order solution in the outer region is $u = 1$, thus, after matching with the
 491 leading order term in (4.13) as $\alpha \rightarrow \infty$, we find

$$492 \quad (4.14) \quad \frac{t}{\ln \alpha} = -\frac{1}{2}(S^2 - S_0^2) + \sigma(S - S_0) - \sigma \ln \left(\frac{S + \sigma}{S_0 + \sigma} \right).$$

493 For the zero surface tension case $\sigma = 0$, we can solve (4.14) explicitly to give

$$494 \quad (4.15) \quad S(z, t) = \left(S_0^2 - \frac{2t}{\ln \alpha} \right)^{1/2},$$

495 again providing square root time dependence.

496 Of particular interest is the special case in which the initial dendrite is the prolate
 497 spheroid (2.5). Here $\rho_0 = \alpha$ and $z_0(0) = 1$, so initially the dendrite has the aspect
 498 ratio $\mathcal{A}(0) = 1/\alpha$. From (4.14) we find the interface is given implicitly by

$$499 \quad (4.16) \quad 1 - \frac{2t}{\ln \alpha} = S^2 + \frac{z^2}{\alpha^2} + 2\sigma \left[\left(1 - \frac{z^2}{\alpha^2} \right)^{1/2} - s + \ln \left(\frac{S + \sigma}{(1 - z^2/\alpha^2)^{1/2} + \sigma} \right) \right].$$

500 Note that the small parameter in this limit is $1/\ln \alpha$, which suggests the analysis here
 501 is valid only for extremely large aspect ratios.

502 **4.3. Numerical results for canonical problem.** For the melting prolate
 503 spheroidal crystal considered in [Subsection 2.2](#), whose surface is (2.6), we find the
 504 mean curvature is largest near the tip, given by

$$505 \quad (4.17) \quad \kappa = \frac{z_0(t)}{\rho_0(t)^2} = \frac{t_e^{1/2} z_0(0)}{(t_e - t)^{1/2}}.$$

506 Thus the right hand side of (4.11b) becomes $\mathcal{O}(1)$ when $t_e - t = \mathcal{O}(\sigma^2)$, suggesting
 507 we rescale according to

$$508 \quad (4.18) \quad t_e - t = \sigma^2 \hat{t}, \quad \mathbf{x} = \sigma \hat{\mathbf{x}}, \quad u = \hat{u},$$

509 and treat the following problem

$$510 \quad (4.19a) \quad \text{in } \mathbb{R}^3 \setminus \hat{\Omega}(\hat{t}) : \quad \hat{\nabla}^2 \hat{u} = 0,$$

$$511 \quad (4.19b) \quad \text{on } \partial \hat{\Omega} : \quad \hat{u} = -\hat{\kappa},$$

$$512 \quad (4.19c) \quad \text{on } \partial \hat{\Omega} : \quad \hat{v}_n = -\frac{\partial \hat{u}}{\partial \hat{n}},$$

$$513 \quad (4.19d) \quad \text{as } \hat{r} \rightarrow \infty : \quad \hat{u} \rightarrow 1,$$

514 when $\hat{t} = \mathcal{O}(1)$, $|\hat{\mathbf{x}}| = \mathcal{O}(1)$, where hats denote scaled quantities. For the case in which
 516 the initial crystal, $\hat{\Omega}(0)$, is a prolate spheroidal in shape, this is a canonical problem
 517 for melting a solid. This one parameter in the problem is the initial aspect ratio.

518 Using the numerical scheme described in [Appendix B](#), we solve (4.19a)-(4.19d)
 519 for \hat{u} and $\hat{\Omega}$. We first consider a near spherical prolate spheroid initial condition
 520 such that the initial aspect ratio is close to unity. [Figure 5](#) compares the aspect ratio
 521 of the numerical solution to (4.19a)-(4.19d) with $\alpha = 0.85$ with the aspect ratio
 522 as predicted by linear stability analysis given by (4.10). This figure shows excellent
 523 agreement between the numerical solution and linear stability analysis, confirming
 524 that the numerical scheme presented in [Appendix B](#) is able to describe the behaviour
 525 of the interface as the aspect ratio decreases to unity. Further, we numerically solve
 526 (4.19a)-(4.19d) with $\alpha = 1/6$, and plot the time evolution of the solution and cor-
 527 responding aspect ratio in [Figure 6](#). As expected, this figure shows that the aspect
 528 ratio decays to unity in the limit that $t \rightarrow t_e^-$.

529 **5. Kinetic undercooling.** In this section, we very briefly consider the effects
 530 of extending the dynamic boundary condition (4.1) to include a kinetic undercooling-
 531 type term:

$$532 \quad (5.1) \quad \text{on } \partial \Omega : \quad u = cv_n - \sigma \kappa,$$

533 where v_n is the normal velocity of $\partial \Omega$ and c is the kinetic coefficient. An argument
 534 for this extended boundary condition is that (4.1) can be derived under equilibrium
 535 conditions, while (5.1) is a corrected version that takes into account nonequilibrium
 536 kinetic effects [24, 41]. Physically, a nonzero kinetic coefficient $c > 0$ penalises high
 537 interface speeds, which is important near extinction since our interface speed scales
 538 like $(t_e - t)^{-1/2}$. A wide variety of studies of Stefan problems have considered kinetic
 539 undercooling [2, 3, 10, 11, 16, 17, 35]. The other important previous study is Dallaston
 540 & McCue [9], where the two-dimensional analogue of the quasi-steady problem (2.2e),
 541 (5.1), (2.2c)-(2.2d) is treated in some detail.

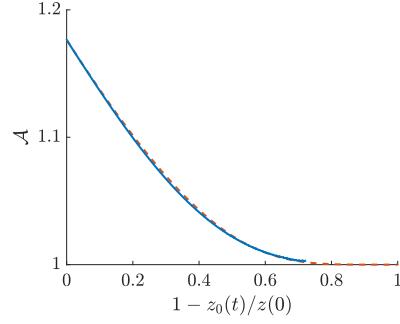


Fig. 5: Comparison of the aspect ratio of the numerical solution to (4.19a)-(4.19d) (blue) with that predicted by linear stability analysis given by (4.10) (dashed red). Initial aspect ratio of the interface is $\mathcal{A}(0) = 20/17$. Numerical solution is computed on the domain $0 \leq \theta \leq \pi$ and $0 \leq r \leq 1.5$ with 314×150 equally spaced nodes.

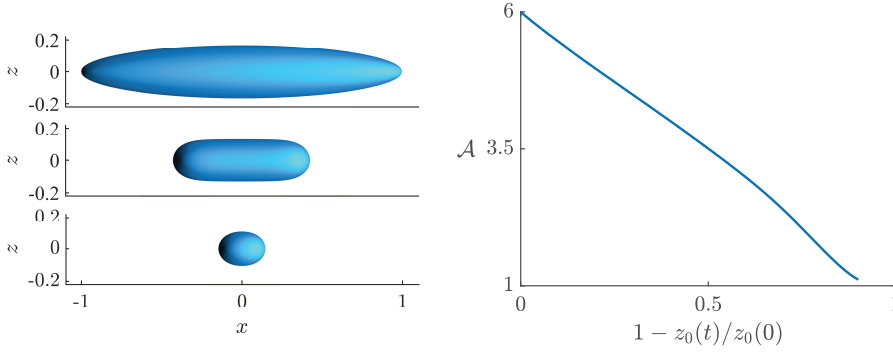


Fig. 6: Left: Numerical solution to (4.19a)-(4.19d) at $t = 0, 0.0033,$ and 0.0052 computed using the scheme presented in Appendix B. Initial condition is of the form (3.18) with $r_0 = 1/6$. Computations are performed on the domain $0 \leq \theta \leq \pi$ and $0 \leq r \leq 1.7$ with 624×340 equally spaced nodes. Right: The corresponding aspect ratio as a function of time.

542 Following the linear stability analysis outlined in Subsection 4.1 using (5.1) with
 543 $c > 0$, we find the second mode of perturbation satisfies

$$544 \quad (5.2) \quad \gamma_2 = \frac{2s_0^2}{3r_0^2} \left(\frac{3c + s_0}{2c + r_0} \right)^{\frac{3c-10\sigma}{3c-2\sigma}} \left(\frac{r_0 + 2\sigma}{s_0 + 2\sigma} \right)^{\frac{6(c-2\sigma)}{3c-2\sigma}},$$

545 from which we see that

$$546 \quad (5.3) \quad \lim_{s_0 \rightarrow 0^+} \frac{\gamma_2}{s_0} = 0,$$

547 suggesting that an initially prolate spheroidal crystal will tend to a sphere in the
 548 extinction limit. This conclusion is that same as before in Subsection 4.1 when $c = 0$.
 549 On the other hand, a significant difference in qualitative behaviour is that the aspect
 550 ratio with $c > 0$ may first increase and then decrease (to unity), which is a feature

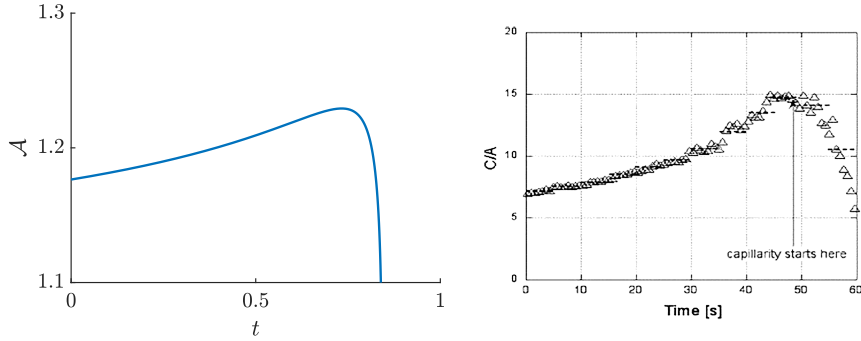


Fig. 7: Left: The aspect ratio of a near spherical prolate spheroid as predicted by linear stability analysis from (4.10) with $\sigma = 0.075$ and $c = 1$. Right: The aspect ratio of a melting PVA crystal [22], reproduced with permission from Springer Nature.

551 not observed when $c = 0$. The turning point can be calculated via

$$552 \quad (5.4) \quad \frac{d}{ds_0} \left(\frac{\gamma_2}{s_0} \right) = 0 \quad \Rightarrow \quad s_0 = \frac{2\sigma c}{c - 4\sigma}.$$

553 Given s_0 is defined on the domain $0 \leq s_0 \leq r_0$, the aspect ratio will monotonically
554 decrease to unity if

$$555 \quad (5.5) \quad \frac{2\sigma c}{c - 4\sigma} < 0, \quad \text{or} \quad r_0 > \frac{2\sigma c}{c - 4\sigma};$$

556 otherwise, the aspect ratio will be non-monotone.

557 Our work is motivated in part by a series of experiments performed as part of
558 the IDGE [21, 22, 43]. In these experiments, it was observed that the aspect ratio of
559 melting crystals increased for a period of time before decreasing to unity at extinction.
560 In the context of the results presented in this section, Figure 7 illustrates the aspect
561 ratio of a (near-spherical) prolate spheroid predicted by linear stability analysis and
562 the aspect ratio of the melting PVA crystals [22]. This figure shows that when both
563 the effects of surface tension and kinetic undercooling are considered, the solution to
564 (2.2c)-(2.2e) and (5.1) is qualitatively similar to the experimental results (while of
565 course the scale is different).

566 **6. Discussion.** In this paper, we have studied a quasi-steady one phase Stefan
567 problem for melting an axially symmetric crystal. In Section 3 we treat a zero-surface-
568 tension model and use analytical tools to show that axially symmetric crystals will
569 tend to prolate spheroids in the limit that they melt completely, namely $t \rightarrow t_e^-$, with
570 an aspect ratio that depends on the initial condition. The point to which the crystals
571 ultimately shrink, together with the melting time, is predicted by this analysis and
572 confirmed using a novel numerical scheme based on the level set method (presented
573 in Appendix B). An advantage of this scheme is that we are also able to present
574 numerical results for crystals that undergo pinch-off and contract to multiple points
575 of extinction.

576 We consider the effects of surface tension by the Gibbs-Thomson law (1.2) in
577 Section 4. By performing linear stability analysis on the spherical solution, we show
578 that surface tension acts to smooth out perturbations to the interface, suggesting it

579 becomes spherical in the extinction limit. A numerical study of canonical problem
 580 confirms this prediction. These results are as expected and also indicated by the ex-
 581 perimental results summarised by Glicksman and co-workers [21, 22, 43]. However,
 582 the one feature of the IDGE not described by the model with surface tension is the
 583 non-monotonic behaviour of the aspect ratio, where the aspect ratio first increases as
 584 the crystal becomes very long and thin, and then very quickly decreases to unity as
 585 surface tension ultimately acts to produce a perfect sphere in the extinction limit. In
 586 order to mimic this non-monotonic behaviour, we have included the effects of kinetic
 587 undercooling in the model in Section 5, which shows that the competition between
 588 kinetic effects and surface tension does indeed produce the qualitative behaviour ob-
 589 served.

590 A key assumption in our paper is that the Stefan number in (2.2a) is taken to
 591 be large, namely $\beta \gg 1$, so that (2.2a) reduces to (2.2e) and our moving boundary
 592 is therefore quasi-steady. There are two issues related to this assumption that we
 593 wish to mention. First, our problem for melting a crystal is the same as that for a
 594 bubble contracting in a porous medium where the flow is governed by Darcy’s law
 595 [12, 28, 45], although in that context the far-field (Dirichlet-type) boundary condition
 596 (2.2d) should probably be replaced with a flux condition that dictates how quickly
 597 the bubble volume is decreasing (in two dimensions the equations describe bubble
 598 contraction in a Hele-Shaw cell [15, 14, 42]). For the case in which a bubble pinches
 599 off to produce two shrinking bubbles, the problem formulation would also need to
 600 consider two points of extraction that coincide with the eventual extinction points.
 601 The second issue is that, strictly speaking, for the extremely late stages of melting,
 602 our quasi-steady model with (2.2e) is no longer applicable in the large Stefan number
 603 limit, and instead (2.2a) must be retained. The mathematical details of such an
 604 exponentially short final-melting stage have been recorded in a number of previous
 605 studies [1, 26, 44, 46, 55].

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609 **Appendix A. Prolate spheroids with constant aspect ratio.**

610 To solve the inner problem (3.4a)-(3.4c) we employ prolate spheroidal coordinates
 611 (ξ, η, ϕ) defined by

612 (A.1a) $X = k \sinh \xi \sin \eta \cos \phi$

613 (A.1b) $Y = k \sinh \xi \sin \eta \sin \phi$

614 (A.1c) $Z = k \cosh \xi \cos \eta,$

616 where $\xi \geq 0$, $0 \leq \eta \leq \pi$, $0 \leq \phi < 2\pi$, and k is a constant to be determined below.

617 The crystal boundary $\partial\Omega_0$ is described by $\xi = \xi_0$ or, equivalently,

618 (A.2)
$$\frac{X^2 + Y^2}{\sinh^2 \xi_0} + \frac{Z^2}{\cosh^2 \xi_0} = k^2.$$

619 Motivated by the relationship

620 (A.3)
$$a(X^2 + Y^2) + \left(\frac{1}{2} - 2a\right) Z^2 = \frac{1}{2} k^2 \left[\left(\frac{1}{2} - a\right) \cosh^2 \xi - a \right] + \frac{1}{2} k^2 \left[\left(\frac{1}{2} - 3a\right) \cosh^2 \xi + a \right] \cos 2\eta,$$

621 we look for a solution of the form

622 (A.4)
$$\Phi = f_1(q) + f_2(q) \cos 2\eta,$$

623 where $q = \cosh \xi$ and $q_0 = \cosh \xi_0$ and obtain a coupled system of two second order
 624 (Legendre-type) differential equations for f_1 and f_2 . These (and the constant k) are
 625 solved subject to the four conditions $f_1 = f_1' = f_2 = f_2' = 0$ on $q = q_0$, and the
 626 far-field condition (3.4c) to give

(A.5)

$$627 \quad f_1 = \frac{1}{2}k^2 \left[\left(\frac{1}{2} - a \right) q^2 - a \right] - d + \frac{1}{8}k^2 q_0 (q_0^2 - 1) \left[q - \frac{1}{2}(q^2 - 3) \ln \left(\frac{q+1}{q-1} \right) \right],$$

(A.6)

$$628 \quad f_2 = \frac{1}{2}k^2 \left[\left(\frac{1}{2} - 3a \right) q^2 + a \right] - d + \frac{1}{8}k^2 q_0 (q_0^2 - 1) \left[3q - \frac{1}{2}(3q^2 - 1) \ln \left(\frac{q+1}{q-1} \right) \right],$$

629 where

$$630 \quad (A.7) \quad k = q_0^{-1/3} (q_0^2 - 1)^{-1/3},$$

631 and d is given by (3.8). The important relationship between q_0 and the special con-
 632 stant a is given by (3.7).

634 **Appendix B. Numerical solution - A level set approach.**

635 To find the numerical solution of (2.2b)-(2.2e), we implement a level set based
 636 approach. The level set method (LSM), first proposed by Osher and Sethian [52], is a
 637 tool used to study a wide range of moving boundary problems. We refer the reader to
 638 Osher & Fedkiw [51] and Sethian [54] for comprehensive overviews of implementation
 639 strategies and applications. The LSM utilises an Eulerian approach by representing
 640 an n -dimensional interface, $\partial\Omega(t)$, as the zero level set of a $n+1$ -dimensional surface,
 641 $\phi(\mathbf{x}, t)$, such that

$$642 \quad (B.1) \quad \partial\Omega(t) = \{\mathbf{x} | \phi(\mathbf{x}, t) = 0\}.$$

643 By representing the interface implicitly, the LSM can be used to describe complex
 644 behaviour such as the changes in topology observed in Figure 2, while operating on a
 645 simple regular two-dimensional grid.

646 The evolution of the level set function ϕ is described by the level set equation

$$647 \quad (B.2) \quad \frac{\partial\phi}{\partial t} + F|\nabla\phi| = 0,$$

648 where F is a continuous function defined on all of the computational domain, satisfying
 649 $F = V_n$ on $\mathbf{x} = \partial\Omega(t)$. In the context of (2.2b)-(2.2e), by noting that the outward
 650 normal of ϕ is $\mathbf{n} = \nabla\phi/|\nabla\phi|$, a suitable expression for F on and outside the interface
 651 is

$$652 \quad (B.3) \quad F = \frac{\nabla u \cdot \nabla\phi}{|\nabla\phi|} \quad \mathbf{x} \in \mathbb{R}^3 \setminus \Omega(t).$$

653 This leaves the matter of defining a suitable extension of F to inside the inter-
 654 face. Among several possibilities in the level set literature, we opt for a biharmonic
 655 extension as proposed by Moroney et al. [49], and compute F inside the interface to
 656 satisfy

$$657 \quad (B.4) \quad \nabla^4 F = 0 \quad \mathbf{x} \in \Omega(t),$$

658 together with the boundary conditions that F and $\partial F/\partial n$ are continuous across $\partial\Omega(t)$.
 659 This method of extension shares the main property of the LSM itself, in not requiring
 660 the location of the interface to be calculated explicitly. To solve (B.4), we formulate
 661 the biharmonic stencil over the entire domain, which is then modified so that values
 662 of F outside the interface, whose location is determined from the sign of ϕ , are not
 663 overwritten. The resulting linear system is solved using LU decomposition. This
 664 extension is a variant of a two-dimensional thin plate spline interpolant defined on
 665 the level set grid.

666 **B.1. General algorithm.** The algorithm used to solve (2.2b)-(2.2e) numeri-
 667 cally is outlined as follows:

668 *Step 1* For a given initial condition $s(\theta, 0)$, construct a level set function $\phi(r, \theta, 0)$
 669 such that $\phi < 0$ inside the interface and $\phi > 0$ outside the interface. This
 670 function is then converted to a signed distance function using the method of
 671 crossing times as described by Osher & Fedkiw [51].

672 *Step 2* Compute the temperature, u , on the domain $r \geq s(\theta, t)$ using the procedure
 673 described in Appendix B.2.

674 *Step 3* Compute F according to (B.3), where the derivatives are evaluated using
 675 central finite differences. F is extended over the entire computational domain
 676 by solving (B.4) at nodes where $\phi < 0$, with boundary data from step 3.

677 *Step 4* Update ϕ by advancing the level set equation given by (B.2), where the time
 678 step is $\Delta t = 0.25 \times \Delta x / \max |F|$. We discretise the spatial derivatives in (B.2)
 679 using a ENO2 scheme for the spatial derivatives and integrate in time using
 680 second order Runge-Kutta where $\Delta t = 0.25 \times \Delta r / \max |F|$.

681 *Step 5* Reinitialise ϕ every 5 time-steps to a signed distance function by solving the
 682 reinitialisation equation

$$683 \quad (B.5) \quad \partial_\tau \phi + S(\phi)(|\nabla \phi| - 1) = 0,$$

684 where

$$685 \quad (B.6) \quad S(\phi) = \frac{\phi}{\sqrt{\phi^2 + \Delta r^2}}.$$

686 We use 5 pseudo-timesteps with $\Delta \tau = 0.2 \Delta r$.

687 *Step 6* Repeat steps 2-5 until the desired simulation time is attained.

688 **B.2. Solving for temperature.** Evaluating the speed function F in the level
 689 set equation (B.2) requires first calculating the temperature u . This is achieved by
 690 using a modified finite difference stencil for Laplace's equation in the region outside
 691 the interface. For nodes away from the interface, a standard 5-point stencil is used
 692 such that the discrete equation is

$$693 \quad (B.7) \quad 0 = \frac{u_{i-1,j} - 2u_{i,j} + u_{i+1,j}}{\Delta r^2} + \frac{2}{r_{i,j}} \frac{u_{i+1,j} - u_{i-1,j}}{2\Delta r} \\ + \frac{1}{r_{i,j}^2} \frac{u_{i,j-1} - 2u_{i,j} + u_{i,j+1}}{\Delta \theta^2} + \frac{\cot \theta}{r_{i,j}^2} \frac{u_{i,j+1} - u_{i,j-1}}{2\Delta \theta}.$$

694 For the singularity at $\theta = 0$, noting that $\partial u/\partial \theta = 0$ and using L'Hôpital's rule then

$$695 \quad (B.8) \quad \lim_{\theta \rightarrow 0^+} \cot \theta \frac{\partial u}{\partial \theta} = \frac{\partial^2 u}{\partial \theta^2}.$$

696

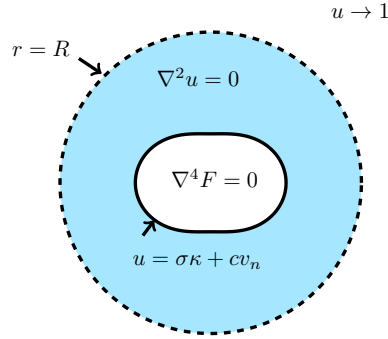


Fig. 8: Schematic of how the speed function, F , is computed for each time step. Blue region denotes where temperature, u , is solved for using finite differences. This finite difference stencil must be adjusted to incorporate the dynamic boundary condition (Appendix B.2.1). To incorporate the far-field boundary condition, we impose an artificial boundary at $r = R$ and implement a Dirichlet to Neumann mapping (Appendix B.2.2). F is computed outside the interface using (B.3), and is extended to be defined over the entire computational domain by solving the biharmonic equation.

697 The same procedure is applied at $\theta = \pi$. Difficulties arise when attempting to in-
 698 corporate the dynamic condition (2.2b) on the interface and the far-field boundary
 699 condition (2.2d). We detail the methodology used to overcome each of these difficul-
 700 ties in Appendices B.2.1 and B.2.2, respectively. A schematic of the problem is given
 701 in Figure 8, which illustrates the different equations to be solved in each part of the
 702 computational domain.

703 We note that since the governing equation for temperature satisfies Laplace's
 704 equation, an alternative approach for computing the temperature u is the boundary
 705 integral method, which can be coupled with the level set method to solve problems
 706 where changes in topology occur [19]. However, an advantage of using a finite differ-
 707 ence stencil is that it can easily be adapted to problems where the boundary integral
 708 method is not applicable. For example, we have used a similar method to the one
 709 presented in this section to study non-standard Hele-Shaw flow where pressure is not
 710 harmonic and for which the boundary integral method is much less suitable [50].

711 **B.2.1. Incorporating the dynamic boundary condition.** Special consider-
 712 ation must be taken when solving for nodes adjacent to the interface as we can no
 713 longer use the second order central differencing scheme (B.7). Instead we follow the
 714 work of Chen et al. [5] and approximate the spatial derivatives by fitting a quadratic
 715 polynomial from values on and near the interface and differentiating this polynomial
 716 twice. Supposing the interface is located between two nodes $(i-1, j)$ and (i, j) , the
 717 quadratic is fitted using the three points (r_b, u_b) , $(r_{i,j}, u_{i,j})$, and $(r_{i+1,j}, u_{i+1,j})$. Here
 718 r_b denotes the location of the interface and u_b is the temperature at the interface.
 719 The value of r_b is found by noting that ϕ is a signed distance function and so the
 720 distance between r_b and $r_{i,j}$, denoted h , can be calculated by

721 (B.9)
$$h = \Delta r \left| \frac{\phi_{i,j}}{\phi_{i,j} - \phi_{i-1,j}} \right|.$$

722 Thus (B.7) becomes

(B.10)

$$723 \quad \frac{\partial^2 u}{\partial r^2} + \frac{2}{r} \frac{\partial u}{\partial r} \rightarrow \left(\frac{2}{h(h + \Delta r)} - \frac{2}{r_{i,j}} \frac{\Delta r}{h(\Delta r + h)} \right) u_b + \left(\frac{2}{r_{i,j}} \frac{\Delta r - h}{h\Delta r} - \frac{2}{h\Delta r} \right) u_{i,j} \\ + \left(\frac{2}{\Delta r(h + \Delta r)} + \frac{2}{r_{i,j}} \frac{h}{\Delta r(h + \Delta r)} \right) u_{i+1,j}.$$

724 The same procedure is applied if the interface is between r_i and r_{i+1} , or in the
725 azimuthal direction.

726 The value of u_b is determined by the dynamic condition (5.1), where in the case
727 of surface tension the mean curvature term

$$728 \quad \kappa = \nabla \cdot \left(\frac{\nabla \phi}{|\nabla \phi|} \right)$$

729 is approximated using central finite differences, while the normal velocity from the
730 previous time step is used for the kinetic undercooling term.

731 **B.2.2. The far-field condition.** Special consideration must also be given when
732 considering the boundary condition at $r \rightarrow \infty$. One method for simulating this far-
733 field condition is to make the computational domain much larger than the radius
734 of the interface and then impose $u = 1$ on the outer boundary. However, this is
735 computationally expensive as very large domains must be used to form an accurate
736 solution. Instead, we simulate the far-field condition using the Dirichlet-to-Neumann
737 (DtN) method [20]. This method is implemented by introducing a spherical artificial
738 boundary, R , which is larger than the radius of the interface, i.e. $R > s(\theta, t)$. Outside
739 of this boundary

$$740 \quad \text{(B.11a)} \quad \text{in } r > R : \quad \nabla^2 u = 0,$$

$$741 \quad \text{(B.11b)} \quad \text{on } r = R : \quad u = f(\theta),$$

$$742 \quad \text{(B.11c)} \quad \text{as } r \rightarrow \infty : \quad u \sim 1,$$

744 holds, where $f(\theta)$ is an unknown function. This problem can be solved exactly via
745 separation of variables giving

$$746 \quad \text{(B.12)} \quad u(r, \theta, t) = 1 + (c_0 - 1) \frac{R}{r} + \sum_{n=1}^{\infty} c_n \left(\frac{R}{r} \right)^{n+1} P_n(\cos \theta),$$

747 where

$$748 \quad \text{(B.13)} \quad c_n = \frac{2n+1}{2} \int_0^\pi f(\theta) P_n(\cos \theta) \sin \theta d\theta$$

749 and P_n denotes the n th Legendre polynomial. Matching this outer solution with the
750 inner numerical solution on the artificial boundary R provides the necessary Neumann
751 boundary conditions for the numerical scheme. By taking the derivative of (B.12)
752 with respect to r at $r = R$ and evaluating (B.13) using the trapezoidal rule, the finite
753 difference stencil for the radial derivatives is updated with

$$754 \quad \text{(B.14)} \quad \frac{u_{i-1,j} - 2u_{i,j} + u_{i+1,j}}{\Delta r^2} + \frac{2}{r_{i,j}} \frac{u_{i+1,j} - u_{i-1,j}}{2\Delta r} \rightarrow \\ \frac{2(u_{i-1,j} - u_{i,j})}{\Delta r^2} + 2 \left(\frac{1}{\Delta r} + \frac{1}{R} \right) f'(\theta_j),$$

755 where

$$756 \quad (B.15) \quad f'(\theta_j) = \frac{1}{R} - \frac{(n+1)(\Delta\theta)}{R} \sum_{k=1}^{m-1} w_{j,k} u(R, \theta_k, t),$$

757 and

$$758 \quad (B.16) \quad w_{j,k} = \sum_{n=0}^{\infty} (n+1) P_n(\cos \theta_j) P_n(\cos \theta_k) \sin \theta_k.$$

759 From a practical perspective, we cannot, of course, evaluate the series in (B.16) using
 760 an infinite number of terms, but have found that using 10 terms gives sufficient accu-
 761 racy. Furthermore, it is a straightforward exercise to use the DtN method for other
 762 types of far-field boundary conditions such as flux condition for fluid flow whereby
 763 $\partial u / \partial r \sim 1/r^2$ as $r \rightarrow \infty$.

764

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