TWO DIMENSIONAL DENDRITIC CRYSTAL GROWTH FOR WEAK UNDERCOOLING

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INTRODUCTION

Dendritic crystal growth has been a subject of interest to physicists, metallurgists as well as mathematicians. The most common example of such a growth is the well-known ice-crystal. From a physicist's perspective, dendrites constitute a relatively simple but important problem of pattern formation in non-equilibrium growth [3-5]. In metallurgy, dendrites are common to crystal formation in the manufacture of alloy when the growth rate exceeds some critical value. The literature on the subject is vast and reviewed in [3-6], as well as in our paper [1]. Here, we report a summary of the contents of our recent paper [2]; the reader is referred to it for more details.

APPROACH

In the first of a sequence of papers on dendritic crystal growth for weak undercooling [1], we derived asymptotic equations for weak nondimensional undercooling Δ (non-dimensionalized appropriately, through a combination of latent and specific heat) for a dendrite that was asymptotically a parabola in the far-field. A Peclet number, P was introduced in accordance to

$$\Delta = \sqrt{\pi P}\, e^{P}\, erfc\ (\sqrt{P}) \tag{1}$$

which is clearly small for small Δ. Based on the length scale a, associated with the far-field parabola, a velocity scale $U = 2\ D\ P\ /a$ was identified, where D is the diffusion constant. a and a/U are used to nondimensionalize all lengths and times. We determined that if the initial deviations from an Ivantsov state (parabolic dendrite with a corresponding temperature profile) are limited to an $O(1)$ region near the tip, then the dynamic evolution of the dendrite for the nondimensional time $t \ll P^{-1}$ involves the $O(1)$ tip region only; in that region, the temperature is harmonic to the leading order, with appropriate boundary and far-field matching conditions. It is to be noted that the derivation does not assume that the deviation from the Ivantsov state is small; only that it does not extend all the way to the far-field $O(P^{-1})$ region. This tip-region dynamics was recast in terms of the evolution of the conformal mapping function from an upper-half ζ plane ($\zeta = \xi + i\eta$) to the exterior of the dendrite in the z-plane, where $z = x + iy$ (See Fig. 1). This function $z(\zeta, t)$ was shown to satisfy the following nonlinear integro-differential equation for real ζ (i.e. on ξ-axis):

$$z_t = (H + iR) z_\xi, \tag{2}$$

where

$$R(\xi, t) = \frac{1 - B\ Im\ \omega_\xi}{|z_\xi|^2}, \tag{3}$$

$$-H(\xi, t) = \mathcal{H}\{R\}(\xi, t) \equiv -\frac{1}{\pi} \int_{-\infty}^{+\infty} \frac{d\xi'}{\xi' - \xi} R(\xi', t), \tag{4}$$

$$\omega(\xi, t) = K(\xi, t) + i\ \mathcal{H}\{K\}(\xi, t), \tag{5}$$

where

$$K(\xi, t) = (1 + \alpha f(\xi, t))\, \kappa(\xi, t), \tag{6}$$

$$\kappa(\xi, t) = -\frac{1}{|z_\xi|}\, Im\, \frac{z_{\xi\xi}}{z_\xi}, \tag{7}$$

$$f(\xi, t) = 1 - cos\, 4(\theta - \theta_0) = 1 - Re\, \left(\frac{z_\xi^2}{\bar{z}_\xi^2} e^{-i\, 4\theta_0} \right). \tag{8}$$

In the above, the nondimensional surface energy parameter B is given by

$$B = \frac{c_p\, \tilde{d}_0\, T_M}{2\, a\, L\, P} \tag{9}$$

where $\tilde{d}_0$ is the capillary length, c_p the specific heat, T_M the melting temperature of a planar interface, and L the latent heat. Further, in the above, κ is physically the nondimensional curvature and $1 + \alpha\ f$ is a four-fold surface energy anisotropy correction. Here θ the angle between the normal to the interface (pointing towards the melt) and the y axis, while θ_0 is some fixed value denoting a direction along which surface energy is a minimum.

Through a linearization of the equation for $z(\xi, t)$ about some generally arbitrarily time dependent state, we were able to determine expression for growth rate of an initially localized disturbance in terms of the base state, through a Fourier-analysis, when the disturbance is far from the tip. In the special case of a base state that is steady and is close to an Ivantsov state, the expressions for the growth rate were in accordance to prior results [7]. Interestingly enough, it was possible to obtain the same results by analytically continuing the equations (2)-(8) to the lower-half complex ζ plane and carrying out an asymptotic analysis for the linearized equations near singularities of z_ζ.

It is to be noted that while the lower-half ζ plane does not correspond to any part of the physical domain, singularities of $z(\zeta, t)$ approaching the real axis from below correspond to interfacial distortions. In particular, we found that according to the linearized dynamics, surface energy prevents an initially localized disturbance from remaining localized beyond a certain time. Arbitrarily small initial interfacial distortions (noise), representable by some singularity distribution in $Im\ \zeta < 0$, significantly affect the interface later in time when singularities of the associated zero-surface-energy problem approach or cross $Im\ \zeta = 0$, even though surface energy locally smooths out all singularities in the linearized dynamics. The extent to which the zero-surface-energy singularity dynamics relates to growth rate and dispersion of disturbances for small non-zero surface energy was also uncovered. Hence, zero-surface-energy singularity dynamics have both qualitative and quantitative impact on the physical predictions mentioned above.

The relation between complex singularity dynamics and the evolving physical features of a dendrite transcends the restriction posed by linearized dynamics since a singularity of the conformal map in $Im\ \zeta < 0$ can result in large interfacial distortions when that singularity approaches $Im\ \zeta = 0$. In particular, if we consider an isolated singularity $\zeta_s(t)$ of z_ζ in the lower-half plane so that

$$z_\zeta \sim E_0(t)\,(\zeta - \zeta_s)^{-\beta} \qquad (10)$$

near $\zeta = \zeta_s(t)$, then if the singularity is very close to the real axis, we can expect a corner pointed towards the crystal, with included angle $(1-\beta)\,\pi$. This is locally rounded off over a length scale determined by singularity distance from the real axis. The larger the $|E_0|$ (singularity 'strength'), the larger is the impact region on the interface. $arg\ E_0$ determines the orientation of this distortion relative to the y-axis. The physical effect of an isolated complex singularity corresponding to $\beta = 1$ (pole) is to create parallel sided indentation with width $\pi\ E_0$ and depth that scales as $-\ln\ \eta_s$, where $\eta_s = Im\ \zeta_s$.

It is to be noted that the geometrical features at the interface associated with (10), as discussed above, will remain intact for a period of time, even when the actual singularity $\zeta_s(t)$ is smoothed out or replaced by a cluster of other singularities, provided there is some intermediate range: $B^\delta << |\zeta - \zeta_s(t)| << 1$ for some δ and some set of real ζ for which the behavior (10) persists.

Prior work for dendrites [1], as well as by others on the mathematically analagous Hele-Shaw problem, shows that the zero-surface-energy dynamics preserves the form of the singularity – i.e. β in (10) remains invariant with time; only its position $\zeta_s(t)$ and its strength $E_0(t)$ evolve (except for a pole where E_0 is invariant). When $\beta < 0$, the form (10) is not invariant. Generally for an initial singularity of that kind,

$$z_\zeta \sim A_0(t) + E_0(t)\,(\zeta - \zeta_s(t))^{-\beta} \qquad (11)$$

for ζ sufficiently close to ζ_s. Such singularity on the real axis does not introduce discontinuity in slope, except in non-generic cases– $A_0 = 0$ just when $Im\ \zeta_s = 0$. In this exceptional case, the corner is directed towards the melt, in contrast to $\beta > 0$ case, when it is directed towards the crystal.

All singularities, regardless of their type, were shown to continually approach the real axis with time, though for $\beta > \frac{1}{2}$, they do not impinge the real axis in finite time–indeed they slow down significantly as they come close to the real axis.

A point where $z_\zeta = 0$, but z_ζ is otherwise analytic, is referred to as a zero. A zero on the real axis corresponds to a zero-angled cusp on the interface that protrudes into the melt. Prior work, discussed in [1], shows that a zero remains invariant with time, when surface energy is neglected, i.e. the form

$$z_\zeta \sim z_{\zeta\zeta}(\zeta_0(t), t)\,(\zeta - \zeta_0(t)) \qquad (12)$$

remains invariant. The evolution equation for $\zeta_0(t)$, however, is found to be different from that of a singularity $\zeta_s(t)$. In particular $\zeta_0(t)$ may or may not approach the real axis. For some set of initial conditions, a zero does impact the real axis in finite time. The mathematical solution ceases to be physically meaningful beyond this cusp-formation time.

The connection between the dynamics in the extended domain $Im\ \zeta \leq 0$ and the physical features of an evolving dendrite, as described above, is particularly useful, since there is strong evidence that the zero-surface-energy dynamics in the extended domain is well-posed [See [8]-[9] for evidence for the mathematically similar Hele-Shaw problem], in contrast to the interfacial evolution itself. In the latter case, the domain is restricted to $Im\ \zeta = 0$. This well-posedness at the zeroth order mathematically justifies a systematic perturbation procedure in the extended complex domain to study how small but nonzero surface energy (with or without anisotropy) alters the zero-surface-energy dynamics. The viewpoint we followed in [1]-[2], following the Hele-Shaw analysis with isotropy [9]-[10], is that the interfacial dynamics comes as a byproduct of the dynamics in the extended domain.

A necessary drawback to the above mentioned procedure is that now one must specify initial conditions in the extended complex domain $Im\ \zeta \leq 0$, which obviously cannot be done in an experiment where only the initial interface shape, up to some non-zero error, can be controlled. Connection to observed statistical features of an experiment can be made only by studying the statistics of an ensemble of complex-plane initial conditions, allowing for every conceivable singularity distribution, and with each member of the ensemble consistent with the given initial shape to within experimental error. Clearly, many different singularity distribution can result in the same approximate interfacial shape. However, an essential precursor to such a statistical study is the thorough description of the dynamics of *all* possible forms for singularities in $Im\ \zeta < 0$. Once this is clarified, one can proceed with the statistical study for an ensemble of initial conditions. That such an approach may be useful is already demonstrated in [2], where we obtain dendrite coarsening results based on an ensemble of particular singularities. However, in general, the analytic continuation of $z(\zeta, 0)$ into $Im\ \zeta < 0$, corresponding to a general analytic initial shape, can be expected to contain natural boundaries and perhaps other singularities that are not isolated. Further, even the class of all possible forms of isolated singularities is too broad to study; only a small subset of possible initial conditions contains the specific classes of isolated singularities and zeros, as in (10) and (12), are considered. Nonetheless, such isolated-singularity distributions do correspond to a range of interfacial distortion, when they come close to the real axis. For that reason, we believe that the statistical features of the interfacial dynamics within this limited class of initial conditions are not very different from what is observed in experiment – with the additional proviso that a two-dimensional theory is applicable, at least insofar as scaling predictions.

However, even within the class of possible initial singularities studied, there are basic mathematical issues concerning the asymptotic matching of inner and outer regions in the complex plane (as the surface-energy parameter goes to zero) that remain unresolved. In carrying out matching in the neighborhood of a singularity that is preserved by the zero-surface-energy dynamics, it is observed that the matching is necessarily sectorial–the inner solution does not match to the outer solution in every direction in the complex plane; it can be matched in a certain sector only. This is not a surprising result, since the steady dendrite problem is known to have the same features. However, unlike the steady problem where there are well defined global Stokes lines even beyond the immediate vicinity of an inner-region that determine local sectors of matching (See [11] for instance.), no basic mathematical principle exists for the time-evolving flow. Only local Stokes lines, corresponding to local similarity solutions of the partial differential equations in the inner region, can be identified. There, we invoke a matching principle based on one used in the Hele-Shaw context [9] . The only direct evidence that such a matching principle is sound is our prior finding, in [1], that there is consistency between results from a Fourier analysis in the real domain and a complex singularity approach involving inner-outer matching for the linearized problem.

Further mathematical difficulties arise with initial zeros of z_ζ, since the full investigation of the dynamics at different stages is hampered by lack, in many cases, of either analytical or numerical solutions to a set of complicated partial differential equations in the complex plane. It is to be noted that the mathematical theory of nonlinear higher order partial differential equation in the complex plane is quite undeveloped. Progress in this case has been made, as in [8], with additional ansatz on the dynamics at intermediate stage[s]. There is no direct evidence that these ansatz are correct by themselves, though the careful numerical calculations of the interfaces themselves, for a sequence of computations for decreasing surface energy, indirectly confirm the basic features of the analytic theory, both for the associated isotropic Hele-Shaw problem [10], and also for anisotropic Hele-Shaw and dendrite problems. This work will be reported in an upcoming paper.

Despite the qualifiers above and the fact that our method necessarily requires a lengthy investigation of complex dynamics involving many kinds of initial singularities with corresponding inner equations depending on their distance from the real axis as well as the relative ordering of anisotropy and surface energy, this technique is the only one known for the fully nonlinear, time-evolving dendrite in the small-surface-energy limit. This limit is precisely the most difficult to explore computationally , since resolving small capillary lengths necessarily strains the capacities of computers. Further, even for cases where B is not small, the small surface energy limit cannot be avoided at large distances from the dendrite tip, where the curvature of an essentially parabolic keeps decreasing.

ISSUES ADDRESSED

In [2], we continue our study of complex singularities initiated in [1] by including small but nonzero surface energy ($0 < B << 1$) in the nonlinear dynamics in the extended complex domain, generally taking anisotropy into account. The purpose of this paper is to address, partly or wholly the following important issues:

1. How does a non-zero B alter singularities described in (10)? Do the the alterations and modifications to the singularity stay confined to a small cluster around $\zeta_s(t)$? Is there an intermediate spatial scale over which the behavior (10) is relevant as $B \to 0$? If so, is there a limitation on the order of $|E_0(0)|$ and the time for which this is so such behavior persists? How does anisotropy in surface energy come into play?
2. What are the time and spatial scales over which surface energy effects become important to the real axis dynamics for a singularity corresponding to $\beta > \frac{1}{2}$? Recall that according to zero-surface-energy dynamics, such singularities do not impinge the real axis in finite time, though it continually approaches it.
3. For singularities corresponding to $0 < \beta < \frac{1}{4}$, which are known, in the absence of surface energy, to impact the real axis in finite time (leading to corners at the interface), what are the relevant space and time scales associated with a small nonzero B, when $Im\ \zeta_s(t) \to 0$
4. What can be expected about the growth rate of interfacial distortions associated with approaching complex singularities discussed above in (1)-(3) ? How does surface energy dissipation of weak singularities, determine a cut-off in the growth rate ? How does anistropy affect the result ? It is often stated in the literature that interfacial distortions that point towards the crystal appear to remain stationary in the laboratory frame. Is there a limitation on the time scale over which this is true ?
5. What is the effect of anisotropic surface energy on an initial zero? Is there a 'daughter singularity' $\zeta_d(t)$ that emerges from an initial zero $\zeta_0(0)$, as for the isotropic Hele-Shaw problem [8]-[10] ? If so, how does anisotropy alter the structure of the cluster of actual singularities of z_ζ that are centered at $\zeta_d(t)$.
6. How does the impact of $\zeta_d(t)$ on the real axis affect the interfacial features? As with isotropic Hele-Shaw problem, can one expect the daughter singularity impact time to indicate when an actual interface will veer off from the corresponding zero-surface energy solution ?
7. How are interfacial cusps, associated with a zero $\zeta_0(t)$ impacting the real axis in finite time, prevented by small surface energy effects ? One scenario is that small surface energy becomes important only when the interface becomes close to a cusp, i.e. when curvature of the zero-surface-energy solution becomes large. The second is that the interface never comes close to cusp-formation because it necessarily veers off from the corresponding zero-surface-energy solution significantly before any $\zeta_0(t)$ can impact the real axis. In the context of complex singularity dynamics of this paper, the two scenarios are distinguished by the question: does a daughter singularity $\zeta_d(t)$ necessarily impact the real axis before the corresponding zero $\zeta_0(t)$?
8. How does a given disturbance, that may be associated with many different complex singularity distributions cause $O(1)$ localized deviation in interfacial slope from a smooth background state, evolve in time. Is there a rescaling under which the equations remain invariant in the small surface energy limit ? What does such an invariance tell us about the dynamics ?
9. How do surface energy and anisotropy modify or confirm the coarsening scenario that we proposed in [1]? The selection effect of surface energy on an ensemble of assumed singularities of different strengths is examined in [2], resulting in a prediction for the coarsening rate of $|y|^{1/2}$ for an intermediate range of distances where $|y|$ is large compared to some inverse power of B. There is no necessary contradiction with the well known $|y|^{1/3}$ coarsening result [12], since that is valid for all sufficiently large distances from the tip.

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Figure 1: Conformal map from upper-half ζ plane to the dendrite exterior