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Velocity selection in dendritic growth

David A. Kessler*
Department of Physics, Rutgers University, Piscataway, New Jersey 08854

Herbert Levine
Schlumberger-Doll Research, Old Quarry Road, Ridgefield, Connecticut 06877
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We show that the velocity and shape of two-dimensional dendritic crystals can be determined by solving the steady-state evolution equation at *finite* surface tension. We find that in the zero undercooling limit, crystal anisotropy is necessary to obtain finite velocities. Furthermore, the "solvability" condition at zero anisotropy and small undercooling is essentially singular in the velocity. Finally, we comment on the extension of our results to finite Peclet number and to three dimensions.

The problem of velocity selection for dendritic crystals has been studied for many decades, without adequate resolution.[1] Specifically, under careful experimental conditions,[2] the tip of the growing crystal assumes a parabolic profile and moves at constant velocity, both of which are experimentally reproducible functions of the undercooling. Ivantsov[3] solved the steady-state heat-transport equation at zero surface tension and showed that there exist parabolic profiles which uniformly translate; however, the velocity and tip radius were left undetermined.

A new conjecture regarding the mechanism of velocity selection in dendritic growth has been recently advanced.[4,5] This conjecture was motivated by the growing evidence that surface tension acts in a similar way in controlling pattern formation in various diffusive systems. The essential idea, which has been dubbed "microscopic solvability,"[6] is that a unique velocity and shape emerges from solving the steady-state equation of motion for the interface at *finite* surface tension.[7] Specifically, finite surface tension introduces a solvability condition via essentially singular terms, and it is this condition, invisible in perturbation theory, which determines the velocity. The original continuum of solutions seen in the absence of surface tension thus breaks down to a discrete set of solutions, each with a unique velocity. The final selection from this discrete set is then a dynamical question, with typically only the fastest-moving solution being stable.[8]

The first demonstration of this mechanism was in the case of the Saffman-Taylor finger.[9-11] Independently, microscopic solvability was seen to apply to simple models of interfacial evolution[12] designed to mimic the physics of dendritic growth. Surface tension also plays the same role in determining the velocity of a gas bubble rising in a tube of liquid.[13]

In this paper, we present the first demonstration that microscopic solvability also controls velocity selection for the equations governing diffusion-limited dendritic crystal growth. The key to this demonstration is the realization that the method developed by Vanden-Broeck[10] for his analysis of microscopic solvability in the case of Saffman-Taylor fingers can be applied to our problem. To simplify the analysis, we will focus in this paper on the limit of small undercooling, which is the relevant one for most experiments to date. Also, we work exclusively in two dimensions. At the end, we will briefly describe the extension of our methodology to both larger undercoolings and to three dimensions, the details of which will be discussed elsewhere.[14]

The equations of motion for two-dimensional dendritic growth can be written as

$$D\nabla^2 T = \dot{T} ,$$
$$T(\mathbf{x}_{\rm int}(s)) = T_M - \frac{\gamma}{L}\kappa(s) , \tag{1}$$
$$c_p D[(\hat{\mathbf{n}}\cdot\nabla T)_l - (\hat{\mathbf{n}}\cdot\nabla T)_c] = -L\mathbf{v}(s)\cdot\hat{\mathbf{n}} ,$$

where γ is the surface tension, L is the latent heat of fusion, and c_p and D are, respectively, the specific heat and thermal diffusivity, assumed for simplicity to be equal in the solid and liquid. The interface is given by $\mathbf{x}_{\rm int}(s)$, has curvature $\kappa(s)$, and moves with normal velocity $\hat{\mathbf{n}}\cdot\mathbf{v}(s)$, s being the arclength. The last equation relates the discontinuity in the temperature gradient going from liquid (l) to crystal (c) to the heat production. Finally, the temperature approaches $T_M-\Delta$ (T_M the melting temperature, Δ the undercooling) at large distances from the interface.

It is more convenient for our purposes to use a Green's-function representation of these equations.[15] Assuming a steady-state solution, translating in the $\hat{\mathbf{y}}$ direction with velocity v, one can derive the integral equation

$$\tilde{\Delta} - \tilde{v}\kappa(x) = \frac{1}{\pi}\int_{-\infty}^{\infty} K_0(\{(x-x')^2+[y(x)-y(x')]^2\}^{1/2}) \times e^{y(x')-y(x)}\,dx' , \tag{2}$$

where

$$\kappa(x) = \frac{-y''(x)}{[1+y'(x)^2]^{3/2}} ,$$

and the dimensionless parameters are given by

$$\tilde{v} = \frac{v}{2D}\frac{\gamma T_M}{L^2 c_p} ; \quad \tilde{\Delta} = \frac{\Delta}{L/c_p} .$$

In this form, the entire problem has been reduced to finding the function $y(x)$ and the number $\tilde{v}$, both as a function of undercooling $\tilde{\Delta}$.

Before proceeding, we would like to make one modification of the above equation. In previous work on simplified models,[12] a major role was played by the crystal anisotropy.

This underlying microscopic anisotropy enters into the continuum equations in two places. First, the surface tension acquires an orientation dependence. Assuming an underlying cubic symmetry, we can model this by replacing κ by $\kappa[1-\epsilon\cos 4\theta(x)]$, where θ is the angle between the interface normal and the crystal axes.[16] Also, the correction for the effect of attachment kinetics, which we neglect here, acquires a dependence on θ.

At large distances from the tip, the curvature decreases, and it can be shown that the interface must approach the Ivantsov solution $y_I=-x^2/2p$ appropriate to zero surface tension.[17] Here p is the Peclet number, related to the undercooling by the two-dimensional form of the Ivantsov relation,

$$\tilde{\Delta}=\sqrt{\pi p}\,e^{p}\,\mathrm{erfc}(\sqrt{p}) \ . \tag{3}$$

Specifically, the first correction to the Ivantsov parabola is given by $\alpha(p)/x$, where α can be explicitly computed.[14] It is therefore useful to adopt an idea of Pelce and Pomeau[18] and use the Ivantsov solution to eliminate $\tilde{\Delta}$ in Eq. (2). The equation now becomes

$$-\tilde{v}\kappa(x)[1-\epsilon\cos 4\theta(x)]=\frac{1}{\pi}\int_{-\infty}^{\infty}K_0(\{(x-x')^2+[y(x)-y(x')]^2\}^{1/2})e^{y(x')-y(x)}dx'-(y\rightarrow y_I) \ . \tag{4}$$

This form has the feature that the contribution to the integral from large values of x' now vanishes, which is important for the numerics.

We expect that solutions will exist for only a discrete set of values for the velocity $\tilde{v}$. This happens because the required approach to the Ivantsov parabola at large x is, in general, inconsistent with imposing a smooth behavior at the tip. To study this, we make use of an approach pioneered by Vanden-Broeck[10] in his work on the selection of the Saffman-Taylor finger width. Specifically, we relax the above equation at the tip ($x=0$) and allow for the presence of a finite cusp. Then, there will exist solutions at all $\tilde{v}$. Finally, the selected discrete set emerges as the special values of the velocity at which the cusp magnitude vanishes. This is the condition of "microscopic solvability." At the same time, the dependence of the cusp magnitude on velocity can be extremely informative as to the nature of the solvability condition near the small velocity limit.

We change variables to $z(q)$, $y\equiv y_I+z$, $x\equiv\tan q$, $0\leq q\leq\pi/2$. At infinity, $q=\pi/2$, $z(q)=0$, and $z'(q)=-\alpha(p)$. We then assume that the curve is symmetric around the tip, and discretize the interval for q via $q_i=\pi i/2N$. This converts Eq. (3) into a set of coupled nonlinear equations for z_i, $i=0,N-1$. These equations are solved using the IMSL Newton's iteration solver ZSPOW to converge to a solution. A typical computation uses 100 points and takes several minutes on the VAX 8600. Occasionally, we use more points to check on the accuracy of our results.

In the remainder of this work, we will focus on the limit of small undercooling, developed in Ref. 18. This is not essential, but it serves to simplify the analysis somewhat. As shown there, the limit can be taken by rescaling lengths by a factor of $2p$, transforming the Ivantsov parabola to $y_I=-x^2$. Then, defining c via $\tilde{v}=2cp^2/\pi$, and using the small distance expansion of the Bessel function, one finds the equation

$$\frac{cy''}{(1+y'^2)^{3/2}}\{1-\epsilon\cos[4\tan^{-1}(y')]\}=-\int_{-\infty}^{\infty}\ln\left[\frac{(x-x')^2+(y-y')^2}{(x-x')^2+(y_I-y_I')^2}\right]dx' \ . \tag{5}$$

Furthermore, α is zero in this limit; the first correction is $O(\ln x/x^2)$[10] and therefore $z'(\pi/2)=0$. In addition, the subsidiary condition derived by Pelce and Pomeau that $\int_0^{\infty}z\,dx$ must vanish provides an independent check on the accuracy of our numerical solutions. We find that this condition is satisfied to better than a part in 10^6.

Let us call the value of y' at the tip (approached from the right) f. Then, $f(c)=0$ is the solvability condition. We find that, at zero anisotropy, f is always negative, with a magnitude that is a rapidly decreasing function of the velocity c. In Fig. 1, we plot $-\ln[-f(c)]$ vs $1/\sqrt{c}$ at zero anisotropy. Note that the data demonstrate that f does not go through zero at any finite value of the velocity c. Based on the behavior seen in the local models of interface evolution[12] at zero anisotropy and in the $\lambda=\frac{1}{2}$ Saffman-Taylor finger,[11] we expect the behavior of f to be controlled by the function $e^{-a/\sqrt{c}}$ for small c. Caroli, Caroli, Roulet, and Langer[19] have also advanced arguments that this behavior is to be expected, with power-law corrections, in this model at *infinite* undercooling. The graph is consistent with this exponential form, there not being yet sufficient data to test for the nature of power-law corrections, if any. Thus, the lack of solvability is due to terms exponentially small in the rescaled velocity c. This immediately explains why we cannot use any simple asymptotic expansion technique, even

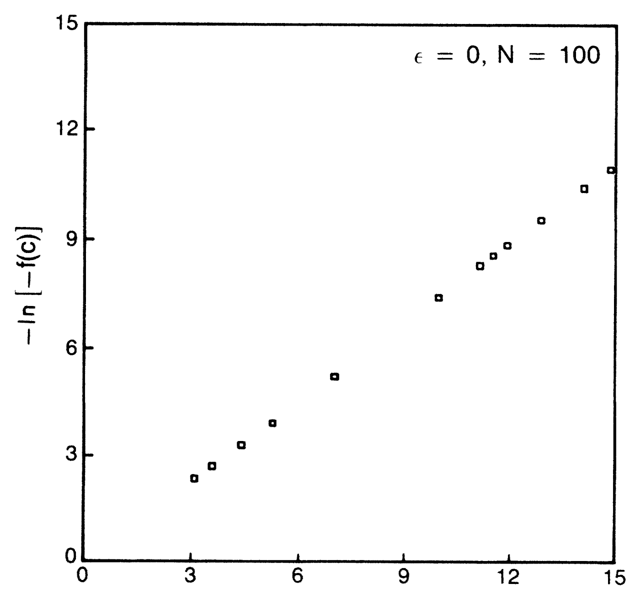

FIG. 1. Cusp magnitude vs velocity at zero anisotropy.

though the physical velocity (even after rescaling by p^2) is small, as we shall now show, for finite crystal anisotropy.

If there is no solution in the small undercooling limit, we must ask why the scaling derived in this limit agrees with experiments. The answer is that the most important correction to the above computation is not finite Peclet number effects, but rather finite crystal anisotropy. In Fig. 2, we plot $f(c)$ vs c at three different values of ϵ: 0.05, 0.1, and 0.15. Now, there is a zero crossing of $f(c)$ at a selected velocity $c=c^*(\epsilon)$, corresponding to an exact steady-state solution. As the anisotropy is increased, the root moves to larger velocity, a qualitatively reasonable result. We can show numerically that the value c^* is the largest possible velocity. We did not find additional solutions but cannot rule out the possibility that these exist for velocities less than 0.002. In any event, we expect that in analogy to Saffman-Taylor fingers, the largest velocity solution will be the only stable solution and hence the physically meaningful one.

To repeat, we have found that at small undercooling, the velocity of the dendrite tip is given by $\tilde{v}=(2p^2/\pi)c^*(\epsilon)$. We have shown how to calculate c^*, and demonstrated that c^* approaches zero for zero anisotropy. Because c^* is small for physically meaningful values of the anisotropy, the curve shape is, in fact, barely distinguishable from the Ivantsov parabola. Our results are in agreement with heuristic arguments that the general Ivantsov solution breaks down at finite surface tension,[19,20] and show that this breakdown is indeed due to a mismatch between conditions imposed on the curve near the tip and infinitely far from the tip.

We expect that in three dimensions, we will again find that the scaling of Pelce and Pomeau,[10] $\tilde{v}=c^*p^2$, will prove to be correct with c^* again a function of the crystal anisotropy which vanishes in the isotropic limit. At finite Peclet number, we expect that solutions will again require finite anisotropy. We will present numerical results on these cases in a forthcoming publication.[14]

Finally, we would like to comment on the idea of "microscopic solvability," which has proven crucial to providing a resolution to the problem of dendrite velocity selection. Without surface tension, there is no length scale with which to derive a velocity and it is therefore immediately apparent that surface tension must be included. The possibility of the breakdown of the continuum of Ivantsov solutions in the presence of surface tension should perhaps not have been unexpected, foreshadowed as it was by the earlier work on singular perturbations in flame propagation.[7] Armed with this notion it is then natural to derive a dimensionless "similarity equation" (in the sense of Barenblatt[21]), in the manner of Eq. (4), in which only the dimensionless "eigenvalue" c appears.

We wish to emphasize, however, the fact that surface tension acts to drive velocity selection in a very distinctive manner. The fact that dendritic growth, Saffman-Taylor fingers, the local interface models, and the rising bubble of gas problem all admit a formulation in terms of a mismatch function, whose vanishing is the subsidiary "solvability" condition which must be imposed on a larger continuum of solutions is by no means an obvious corollary of the singular nature of the surface tension as a perturbation. Even more noteworthy is the existence in all these models of a "critical" end point, at which the mismatch function only vanishes for zero surface tension. This is true for zero anisotropy in the dendrite and local interface models, at $\lambda=\frac{1}{2}$ in Saffman-Taylor and, presumably, at Froude number $=0.23$ in the gas-bubble problem. Furthermore, at these critical values, the mismatch shares a common exponential dependence on the inverse square root of the surface tension.

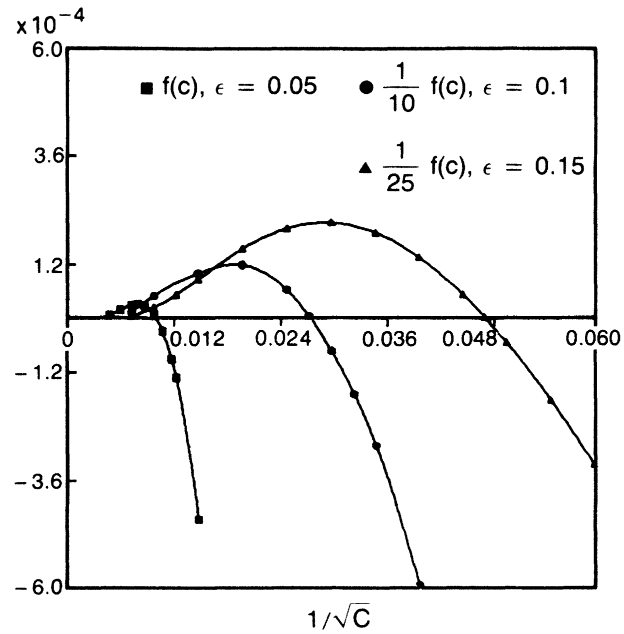

FIG. 2. Cusp magnitude vs velocity at $\epsilon=0.05$, 0.10, and 0.15.

The existence of these "critical" points has a striking consequence. One would naively expect the selected value of c to be of order unity. If this were the case, the corrections to the Ivantsov shape would also be of order unity. In physical terms, this would imply that the dendrite would exhibit large deviations from a pure parabola. However, as mentioned above, the fact that c vanishes at zero anisotropy means that c is small for realistically small anisotropies. The deviations from the Ivantsov shape are then correspondingly small. Also, the shape correction is smooth, exhibiting no structure on the microscopic scale. These two (as yet not fully understood) facts were responsible for the difficulty in understanding the essential role of surface tension in this system in the first place.

After completion of this manuscript, we learned of the independent work of D. Meiron [Phys. Rev. A **33**, 2704 (1986)] which reaches many of the same conclusions regarding velocity selection in dendritic growth.

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*Present address: Department of Physics, University of Michigan, Ann Arbor, MI 48109-1120.

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