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The relationship between dendrite tip characteristics and dendrite spacings in alloy directional solidification

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Abstract

We summarize and discuss a new theoretical model for the directional solidification of alloys in the form of dendrite arrays. Slender body theory is used to obtain an integral equation for the dendrite shape in the asymptotic limit of the dendrite tip radius being much smaller than the solute diffusion length. This equation has a solvability condition that selects the shape and tip undercooling for prescribed solidification conditions and array spacings. A consequence of our results is that we obtain a unique solution to the well-known indeterminacy for the single-dendrite [2] similarity solution by considering the interaction between individual members of an array of dendrites. Further, the dependence of the solutions on the dendrite spacing gives a family of "array solutions," in which the tip radius is related directly to the dendrite spacing. These solutions agree well with experiments in parameter ranges where the slender dendrite theory is expected to be valid. Finally, we discuss how these array solutions, together with surface energy and stability considerations, can describe the selection of dendrite spacings during directional solidification. © 1999 Elsevier Science B.V. All rights reserved.

Keywords: Directional solidification; Dendrite tip characteristics; Dendrite spacing

1. Introduction

Dendrites are a common solidification morphology in both pure materials and alloys [1]. For decades their growth characteristics have been modeled by the similarity solution due to Ivantsov [2]. This solution describes the growth of a single isothermal dendrite into an undercooled liquid of infinite extent. The shape of the dendrite is a paraboloid moving at constant speed V with radius of curvature at the tip ρ. The similarity solution is indeterminate in that, for a prescribed undercooling, the product ρV is determined, but neither ρ nor V is uniquely specified. Despite the indeterminacy (as well as the fact that the Ivantsov solution is smooth and real dendrites have sidebranches) the Ivantsov solution describes the growth of "isolated" dendrites well: for a given undercooling the predicted product ρV corresponds to experimental data [3]. However, in contrast to the indeterminate

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Ivantsov solution, the experiments show a specific tip radius and growth velocity for a given undercooling.

The indeterminacy in the Ivantsov solution is because the mathematical model lacks another length scale that determines the dimensions of the tip. The traditional view is that this missing length scale is the capillary length associated with the surface energy of the solid/liquid interface [4]. Theories that describe tip selection by surface energy include those which incorporate marginal stability theory [5], microscopic solvability theory [6,7], or interfacial wave theory [8]. While these theories have been successful in describing experiments to varying degrees, we show here how to obtain a unique solution by a different mechanism altogether. We show that, neglecting surface energy considerations, the interaction of a dendrite with its neighbors (or container side walls) suffices to eliminate the indeterminacy in the Ivantsov solution. This selection mechanism is generic: the length scale of the dendrite spacing or distance to the container side-walls is present in all real systems. Since all real dendrites have neighbors and/or container walls, the selection of one member of the Ivantsov family by such a length scale may be of wide-ranging importance.

The full mathematical development of the theory involves a substantial amount of both asymptotic and numerical analysis and is published elsewhere [9,10]. The purpose of this paper is to: (1) summarize the predictions of this theory in terms of easily understandable physical parameters; (2) clearly explain the idea behind what we shall call array solutions and the consequential relationship between the tip characteristics and the dendrite spacing; (3) discuss and compare the predictions of the theory relative to existing models of dendritic array growth; (4) advocate the array solutions as a possible alternative/complement to the traditional surface energy selection mechanism for dendritic growth; and (5) explain the possible implications of these array solutions for the determination of primary dendrite spacings.

A main result of our theory is that there is an alternative way to remove the indeterminacy of the single-dendrite similarity solution without appealing to surface energy. However, we do not claim that surface energy is irrelevant to dendrite growth (it is at least necessary to regularize the otherwise ill-posed Ivantsov problem). We do claim, however, that the influence of neighboring dendrites (or a finite system size) which is neglected in the Ivantsov problem, leads to a fundamental effect which is as physically relevant as surface energy. It is likely that both the effect of neighboring dendrites as well as surface energy are important in describing dendritic growth. A comprehensive treatment which includes both is yet to be completed.

In our work we view the spacings as "inputs" to determining the behavior of the tip. Of course a larger question is what determines the spacings in the array. It has been documented experimentally [11,12] and theoretically [13–15] that the spacings of an array of dendrites are determined dynamically by competition/interaction between the dendrites. These interactions result in a range of stable array spacings. Specifically, if the dendrites are too close then the array is unstable to members of the array being left behind by the advancing tips. This overgrowth mechanism leads to an increase in the average dendrite spacing. On the other hand, if the dendrites become too far apart, then the dendrites become unstable to the development of new primary dendrites through the growth of tertiary arms. The tertiary outgrowth mechanism leads to a reduction in the average dendrite spacing. These two mechanisms result in an upper and lower bound on the stable spacings permitted by the array. Our work is directly relevant to the question of dendrite spacings because these instability mechanisms occur at the tip in response to the dendrite spacings. What our results provide is an important connection between the spacings and the behavior of the dendrite tip. At the end of this paper we discuss how our theory relates to the question of stable dendrite spacings. We find that the predictions of our work can be used to determine a bound on the range of stable spacings for given solidification conditions. These results follow directly from our predictions of the relationship between the tip characteristics and the dendrite spacings.

The idea of a direct relationship between the tip characteristics and the array spacing which is found in our results can also be found in simple models of dendrite growth during directional solidification which use ad hoc descriptions of the dendrite shape [16–19]. In these works, a specific dendrite shape (or family of shapes) is assumed a priori, and then the relationship between the tip radius and the spacing is deduced from conservation of heat or conservation of mass. In general the assumed shapes do not solve the free boundary problem for dendrite growth, so the solutions are ad hoc approximations. Our contribution is to determine the actual solution to the free boundary problem for the array and show that the relationship between the tip characteristics and the dendrite spacings is actually a property of the solution to the free boundary problem. We then illustrate how our work relates to these previous theories and compare our predictions with the results from experiments.

While our work derives a result that is similar in spirit to the ad-hoc approximations, we give this result a different interpretation which takes into account recent evidence on existence of a range of stable spacings during dendritic growth. In these earlier models, the relationship between the tip radius and the spacing was augmented by a selection criteria for the tip radius (minimum undercooling in Ref. [16] and marginal stability in Refs. [17,18]) to give a unique spacing for the array. The implication was that there was a unique spacing and tip radius for given solidification conditions. Later, Warren and Langer [13] developed a description of dendrite array growth based on paraboloidal dendrites. In their work, the dendrite tip radius was determined by microscopic solvability with a weak dependence on the dendrite spacings, but without any additional constraints due to the geometry of the array. Thus, they found a family of solutions parameterized by the dendrite spacing. Warren and Langer showed that spacings greater than a critical spacing were stable, giving a range of stable spacings. As with the earlier theories, however, the Warren–Langer dendrites are not solutions to the free boundary problem. We find that if one solves the complete problem for the free boundary, the tip radius is necessarily linked to the spacing of the array without the need for the additional condition of microscopic solvability at the dendrite tip. Despite this fundamental difference, however, our theory gives an end result which is similar to that of Warren and Langer, namely a family of solutions parameterized by the spacing. As in Warren and Langer, we expect that there will be a range of spacings which are stable. In our discussion we compare our results to the Warren–Langer theory and find that, despite having a different selection mechanism, the theories give very similar results over a range of solidification conditions which compare favorably with the experimental results. A related contribution are the numerical calculations of Lu and Hunt [14] which incorporate both the effect of neighboring dendrites and the effect of surface energy. They find that solutions exist if the surface energy is anisotropic, and then there is a family of solutions parameterized by the spacing. In essence, their results are similar to Warren and Langer, with selection being determined by surface energy and not by a geometric constraint.

The rest of the paper is organized as follows. In Section 2 we summarize the theoretical model. In Section 3 we describe the results of the theory, including a comparison to experimental data and a summary of the results as a function of the parameters. Finally, in Section 4 we explain how the tip characteristics are determined by the spacings, compare the predictions of our model to a number of existing models for dendritic growth and discuss the implications of the array solutions for the determination of primary dendrite spacings.

2. Theory

Consider an array of dendrites growing in parallel (Fig. 1.) It is clear that the Ivantsov solution is no longer appropriate for an array because an array of Ivantsov paraboloids will always overlap at some distance behind the tip. Instead, interactions between neighboring dendrites will cause the shape of each dendrite to deviate inwards from the Ivantsov solution and prevent them overlapping. The description of this deviation from the Ivantsov solution due to the presence of neighbors is a significant result of our work. The most important result, however, is that the presence of neighbors determines the dendrite tip characteristics. Without neighboring dendrites we recover the Ivantsov

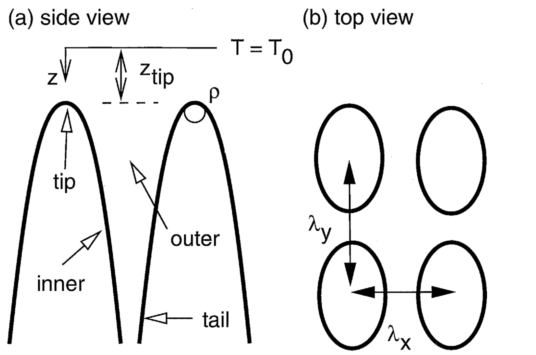

Fig. 1. Schematic diagram of the directional solidification of an array of needle crystal dendrites.

indeterminacy; with neighbors we obtain a unique solution. Of course the more difficult question is how the dendrite spacings themselves are determined. While our theory does not give a complete answer to this question, it provides an important component in that it describes how the tip radius and dendrite spacing are linked. From this relationship we can suggest some possible scenarios for the determination of the dendrite spacings.

Here we present a summary of the theory, with the complete details given in Refs. [9,10]. First, we consider the directional solidification of a binary alloy. While this system is slightly more complicated than the isothermal, one component system for which the Ivantsov solution applies, in the limit of zero temperature gradient and infinite dendrite spacings we recover the solutal Ivantsov problem (isothermal solidification into a supersaturated liquid) as a limiting case. Further, by considering the directional solidification of a binary alloy, we can compare our predictions directly to experiments on dendrite arrays. For simplicity we take the phase diagram to be composed of straight lines with constants k and m_{L} denoting the segregation coefficient and liquidus slope, respectively. We denote the alloy composition as C_{∞} with a liquidus temperature T_0. The alloy then has equilibrium freezing range $\Delta T_0 = m_{\mathrm{L}}(k-1)C_{\infty}/k$, where we use the convention $m_{\mathrm{L}}(k-1) > 0$. The steady-state solidification morphology is assumed to be a rectangular array of identical needle crystals which grows in the $-z$ direction at constant speed V (see Fig. 1). We use a coordinate frame moving with the solidification front and model the heat and solute transport using the one-sided, "frozen-temperature" model.[1] Thus, the temperature field is fixed in the moving frame and denoted by $T = T_0 - Gz$, where G is the constant positive temperature gradient. Two process length scales are determined by the solidification conditions: a diffusion length, $l_{\mathrm{D}} = D/V$, where D is the diffusivity of solute in the liquid; and a thermal length, $l_{\mathrm{T}} = k\Delta T_0/G$, which is related to the vertical extent of equilibrium solidification in the imposed temperature gradient. In addition to the two process length scales there are four length scales which quantify the morphology of the dendrite array: the dendrite tip radius ρ, the spacings of the rectangular array λ_x and λ_y, and the position of the dendrite tip in the imposed temperature gradient relative to the liquidus isotherm, $z_{\mathrm{tip}} = (T_0 - T_{\mathrm{tip}})/G$.

The free boundary problem for the shape of the dendrite consists of equations for the concentration of solute in the liquid, C. These equations describe diffusion of solute in the liquid; conservation of solute and local equilibrium at the dendrite surface; and a condition on the liquid concentration far ahead of the solidification front. In addition, since we are looking for periodic arrays of identical dendrites, we can focus on describing a single dendrite at the center of a repeating "unit cell" with periodic boundary conditions. We nondimensionalize all lengths with l_{D} and describe the dendrite shape in nondimensional cylindrical coordinates relative to the center axis of the dendrite. Denoting the cylindrical coordinates as (r,ζ,θ), where $\zeta = (z - z_{\mathrm{tip}})/l_{\mathrm{D}}$ is the nondimensional distance behind the dendrite tip, the surface of the dendrite is described by $r = R(\zeta, \theta)$. We scale the concentration using $C = C_{\infty}[1 + C^*(1-k)]$ to obtain equations for the nondimensional concentration C^*. Dropping the $*$ superscripts, we obtain the following

[1] In this model the diffusion of solute in the solid is negligible, the diffusion of solute in the liquid is rate-controlling, the diffusion of heat is the same in both phases, and the diffusion of heat is much faster than the diffusion of solute in the liquid. For a further description see Ref. [20].

nondimensional free boundary problem for the steady-state dendrite:

$$\nabla^2 C - \frac{\partial C}{\partial \zeta} = 0 \quad \text{in the liquid,} \tag{1}$$

$$n_z[1 + (1-k)C] = (\boldsymbol{n} \cdot \nabla C) \quad \text{on the dendrite,} \tag{2}$$

$$C = \Theta + g\zeta \quad \text{on the dendrite,} \tag{3}$$

$$C \to 0 \quad \text{as } \xi \to -\infty \tag{4}$$

and periodic boundary conditions at the edges of the repeating unit cell. In the above, $\Theta = (T_0 - T_{\mathrm{tip}})/k\,\Delta T_0 = (C_{\mathrm{tip}} - C_\infty)/(C_\infty(1-k))$ is the nondimensional solute undercooling of the tip relative to the liquidus, $g = l_{\mathrm{D}}/l_{\mathrm{T}}$ is the relative strength of the temperature gradient, $\boldsymbol{n}$ is the outward normal to the dendrite surface, and n_z is the z-component of $\boldsymbol{n}$.

A set of consistent scalings for slender dendrites can be developed when g is small [9]. Defining a slenderness parameter as $\varepsilon \ll 1$, these scalings are $g = \mathrm{O}(\varepsilon)$, $\Theta = \mathrm{O}(\varepsilon)$, $p = \rho/l_{\mathrm{D}} = \mathrm{O}(\varepsilon)$, and $(\Lambda_x, \Lambda_y) = (\lambda_x, \lambda_y)/l_{\mathrm{D}} = \mathrm{O}(1)$. With these scalings we solve the free boundary problem using slender body theory and matched asymptotic expansions to find locally valid solutions in four asymptotic regions (see Fig. 1): the tip region $[|\zeta| = \mathrm{O}(\varepsilon), r = \mathrm{O}(\varepsilon)]$; the inner region $[\zeta = \mathrm{O}(1), r = \mathrm{O}(\varepsilon^{1/2})]$; the outer region $[|\zeta| = \mathrm{O}(1), r = \mathrm{O}(1)]$; and the tail region $[\zeta = \mathrm{O}(1/\varepsilon), r = \mathrm{O}(1)]$. The locally valid solutions are then matched and combined to construct a uniformly valid composite solution.

The four local solutions can be summarized as follows. In the outer region the dendrite appears as a line source of solute, $Q(\zeta) = \mathrm{O}(\varepsilon)$ which extends from $0 < \zeta < \infty$. The tip solution is given by an Ivantsov dendrite, $r = R_{\mathrm{tip}}(\zeta)$, where

$$R_{\mathrm{tip}}^2 = 2p\zeta. \tag{5}$$

The tail region corresponds to two-dimensional solidification in the radial direction. The cross section of the dendrite in the tail region describes the "filling" of the rectangular unit cell as solidification proceeds behind the dendrite tip, and is therefore not axisymmetric. From conservation of solute, the Scheil-type solution gives the cross-sectional area of the dendrite as

$$A_{\mathrm{tail}} = \Lambda_x \Lambda_y \{1 - [1 + (1-k)g\zeta]^{-1/(1-k)}\}. \tag{6}$$

The inner solution contains the interactions with all other dendrites in the array and describes the shape of the dendrite between the tip and tail. For the case of equal dendrite spacings in the x and y directions ($\Lambda_x = \Lambda_y$) the inner solution is axisymmetric to leading order and can be described by $r = R_{\mathrm{in}}(\zeta)$. Conservation of solute on the surface of the slender dendrite gives the shape of the inner solution as

$$\pi R_{\mathrm{in}}^2 = \int_0^\zeta Q(\zeta')\,\mathrm{d}\zeta'. \tag{7}$$

If $\Lambda_x \neq \Lambda_y$ then the lack of 4-fold symmetry in the tail region means that the inner solution and tip solutions may have elliptical cross sections instead of circular cross sections. In this case, R_{in} and R_{tip} each represent an "effective radius" of the cross section.

The line source strength $Q(\zeta)$ is determined by local equilibrium on the surface of the slender dendrite and requires that the solute field generated by the array of line sources $Q(\zeta)$ varies linearly in response to the temperature gradient

$$\begin{aligned}\theta + g\zeta = {} & \frac{-Q(\zeta)}{4\pi}\left[\ln\left(\frac{R_{\mathrm{in}}^2}{4\zeta}\right) + \gamma_{\mathrm{E}}\right] \\ & + \int_0^\infty [Q(\zeta') - Q(\zeta)]G_{00}(\zeta;\zeta')\,\mathrm{d}\zeta' \\ & + \sum_{\substack{i=-\infty \\ i^2+j^2\neq 0}}^{\infty} \sum_{j=-\infty}^{\infty} \int_0^\infty Q(\zeta')G_{ij}(\zeta;\zeta')\,\mathrm{d}\zeta',\end{aligned} \tag{8}$$

where γ_{E} is Euler's constant, $G_{ij}(\zeta;\zeta') = \exp\{-(1/2)[d_{ij} + (\zeta' - \zeta)]\}/(4\pi d_{ij})$, and $d_{ij} = [(i\Lambda_x)^2 + (j\Lambda_y)^2 + (\zeta' - \zeta)^2]^{1/2}$. The matching of the inner solution to the tip and tail solutions generates additional constraints. For $\varepsilon \ll \zeta \ll 1$ the inner solution must match the Ivantsov tip, and for $1 \ll \zeta \ll 1/\varepsilon$ the inner solution must match the Scheil-type tail. These conditions give the additional conditions $Q(0) = 2\pi p$ and $Q(\infty) = g\Lambda_x\Lambda_y$.

The integral (8) has a nonlinear solvability condition [10]: given g and (Λ_x, Λ_y), the unknowns Θ and $Q(\zeta)$ are simultaneously determined. The constraint $Q(\infty) = g\Lambda_x\Lambda_y$ is automatically satisfied by the solution to the integral equation, and the tip radius

p is determined from $p = Q(0)/(2\pi)$. Thus, for directional solidification there is a unique dendrite shape, tip radius, and tip undercooling for given solidification conditions and dendrite spacings. However, if we consider an isolated, isothermal solutal dendrite ($g = 0$, $\Lambda_x = \Lambda_y = \infty$), we recover the paraboloidal Ivantsov solution with $Q = 2\pi p$ (constant) and with the undercooling given by $\Theta = (p/2)[-\ln(p/2) - \gamma_E]$. This relationship between Θ and p is precisely the leading order terms in the expansion of the Ivantsov relationship $\Theta = (p/2)E_1(p/2)\exp(p/2)$ for a slender dendrite ($p \ll 1$). Thus, without neighbors, our theory recovers the Ivantsov solution and associated indeterminacy; with neighboring dendrites we obtain a unique solution.

The dendrite shape is determined by solving Eqs. (7) and (8) numerically to determine the inner solution, and then combining the inner solution with the tip and tail solutions to generate a uniformly valid composite solution (see Ref. [10] for details). Fig. 2 shows the local solutions and composite solution for a typical dendrite shape as calculated by our theory. The figure clearly shows the inward deviation of the composite solution from the Ivantsov tip behavior because of the interaction with the neighboring dendrites. While these results give a unique tip radius for a given spacing, a complete theory needs to address the determination of the dendrite spacing. We discuss this issue following a comparison of our results with experiments and other dendrite theories.

3. Results

To test our theory, we compare our predictions to measured tip radii in directional solidification experiments with SCN-acetone [21]. For the comparison, we use the solidification conditions of the experiment, g, and the measured dendrite spacings, Λ_x and Λ_y. Fig. 3 illustrates the dependence of the tip radius with the spacing as predicted by our theory. There are no adjustable parameters in our theory, and the agreement is quite good for smaller values of g where the asymptotic theory for $\varepsilon \ll 1$ is expected to be valid. That we predict the tip radius this well without regard to surface energy is remarkable. Also shown in Fig. 3 are other theories of dendrite array growth which we will discuss below.

Fig. 4 summarizes the predicted nondimensional tip radius p from our theory for all solidification

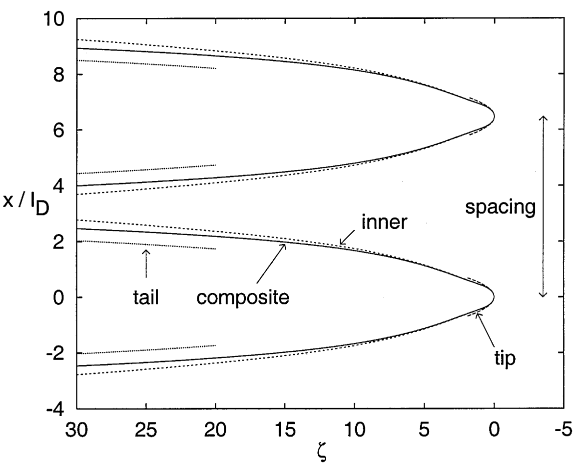

Fig. 2. Composite solution for a slender dendrite. Shown are local solutions for the tip, inner, and tail regions, as well as the composite solution for the entire shape. The parameter values of this solution are $k = 0.1$, $g = 0.0116$, $\Lambda_x = 6.47$ and $\Lambda_y = 7.75$. The nondimensional tip radius is $p = 0.125$ and the nondimensional tip undercooling is $\Theta = 0.116$.

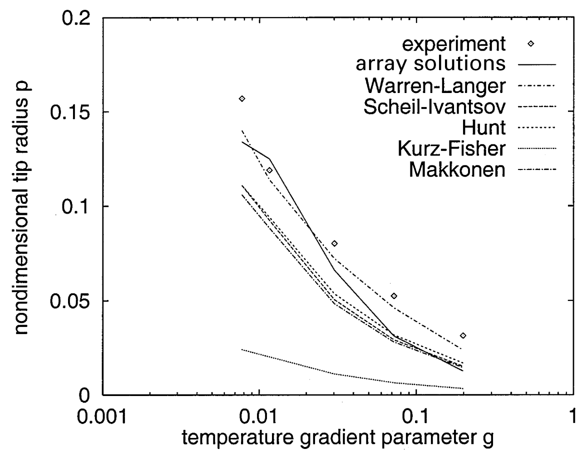

Fig. 3. Comparison of theoretical predictions for the nondimensional tip radius p to data from the Somboonsuk, Mason and Trivedi experiments on SCN-acetone. The slender body theory has no adjustable parameters and assumes that g is small. Also shown are other theories as discussed in the text.

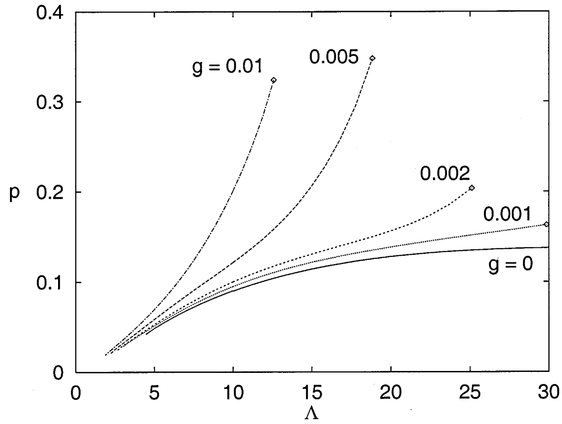

Fig. 4. Theoretical calculations of the nondimensional tip radius p as a function of the dendrite spacing $\Lambda = \Lambda_x = \Lambda_y$ for different values of the temperature gradient parameter g.

conditions and spacings. Note that the tip radius generally increases as a function of the spacing. However, each solution curve terminates at a critical Λ due to a breakdown of our slender body scalings. In particular, following the $g = 0.01$ curve upwards, the dendrite develops a bulbous tip which then pinches off at the point the curve terminates. Thus, it is not possible to consider $\Lambda \to \infty$ at fixed g in our asymptotic theory. Nonetheless, the results in Fig. 4 provide predictions of the tip radius over a wide range of solidification conditions and array spacings.

The tip supersaturation (solute undercooling) θ for the solutions depicted in Fig. 4 are shown in Fig. 5. As $g \to 0$ the solutions collapse onto the small-p Ivantsov relation cited earlier. Thus, for small g and moderately large Λ our theory selects a specific Ivantsov tip solution. At small p, however, the Ivantsov solution is not recovered. This is because at small p the spacings Λ are also getting small in accordance with Fig. 4. Thus, for small p, as in the general case of nonzero g, the tip is paraboloidal but not isolated: the tip undercooling is modified from the Ivantsov undercooling by the neighbors in the array.

4. Discussion

The relationship between the tip characteristics and the dendrite spacing is due to the nonlinear interactions in the array. In the tip and tail regions there are local solutions. The tip solution corresponds to the Ivantsov dendrite and has an indeterminacy. The tail solution, however, is uniquely determined by the spacings because it describes how the dendrites impinge on one another. Of the family of tip solutions, only one is consistent with the tail solution. Thus, the details of the tail solution, which are set by the dendrite spacing, provide the link to the tip characteristics. To determine the transition from tip to tail correctly one must solve for the inner solution which depends on the array interactions as given by the double sum in Eq. (8), but as a simplified explanation as to why the spacing of the array determines a tip radius we can think of directly matching the tip and tail solutions for $\zeta = O(1)$. Expanding Eq. (6) for $g \ll 1$ and using an effective radius of the tail cross section as $r = R_{\text{tail}}(\zeta)$, we obtain

$$\pi R_{\text{tail}}^2 \sim \Lambda_x \Lambda_y g \zeta. \tag{9}$$

To match the Ivantsov tip solution (5) we thus require

$$p = \frac{g\Lambda_x\Lambda_y}{2\pi}. \tag{10}$$

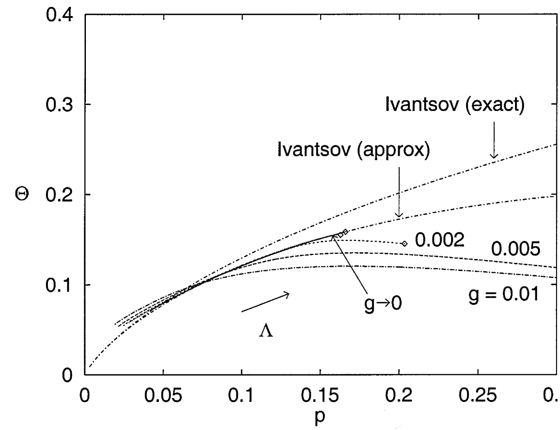

Fig. 5. The nondimensional tip supersaturation (solute undercooling) Θ as a function of the nondimensional tip radius p for the solutions from Fig. 4. Along each curve the spacing Λ increases as p increases. Also shown is the Ivantsov relation $\Theta(p)$ and the small-p approximation appropriate to our slender dendrite theory (see text).

Consequently, the nondimensional tip radius p is uniquely determined by the imposed temperature gradient g and the spacings Λ_x and Λ_y. While the above Ivantsov–Scheil result captures the essence of the selection mechanism, we emphasize that Eq. (10) is not completely correct. The correct relationship between the tip radius and the spacing must be found by solving for the details of the inner transition region from Eqs. (7) and (8). These equations were derived based on slender body theory for the dendrite shape ($\varepsilon \ll 1$), but we believe that this selection mechanism persists even when ε is not small.

The above simple relationship for p is similar to a number of ad hoc theoretical models for dendritic arrays. Hunt [16] used a spherical cap solution for the dendrite tip in conjunction with a 1-D diffusion field for the array to relate the spacings to the tip radius. In terms of our notation, the result is

$$p = \frac{g\Lambda_x\Lambda_y}{4\sqrt{2}[1 - kg + (1-k)g\Lambda_x\Lambda_y/(4\sqrt{2})]}. \tag{11}$$

Trivedi [18] used the Hunt model in his description of array growth. Kurz and Fisher [17] assumed a hexagonal array of ellipsoidal dendrites and determined the tip radius by conservation of mass. Using a geometric factor to convert from a hexagonal to rectangular array, their results can be expressed as

$$p = \frac{2g\Lambda_x\Lambda_y k(1-k)}{3\sqrt{3}[1 - kg]}. \tag{12}$$

Finally, Makkonen [19] employs a conservation of heat argument with the assumption of a parabolic tip to find

$$p = \frac{g\Lambda_x\Lambda_y\Upsilon}{\pi}, \tag{13}$$

where $\Upsilon = k\,\Delta T_0 C/L$ measures the range of equilibrium undercooling to the unit thermal undercooling (C is the heat capacity and L is the latent heat). While the details of these relationships are all different, and each makes different ad hoc assumptions about the dendrite shape, they each share a common feature with the simple Ivantsov–Scheil relationship. Each gives a tip radius which increases monotonically with the dendrite spacing. This feature is preserved in our solution to the free boundary problem. These dendrite theories are shown in Fig. 3, significantly below the experimental results.

The other dendrite theory mentioned earlier is that due to Warren and Langer [13]. Warren and Langer assume the array consists of parabolic dendrites with a tip radius which is determined by a selection criteria based on surface energy. As there is no additional geometric constraints to prevent overlapping dendrites in the array, their results give a unique tip radius and tip undercooling for given solidification conditions and spacings. The Warren–Langer results are also shown in Fig. 3.

Overall, the Warren–Langer and slender body theories reproduce the observations much better than the ad hoc theories. The Warren–Langer theory and our slender body theory are in good agreement with the experiments at small g. At larger g our asymptotic theory is not as accurate, as expected. Thus, even though the Warren–Langer theory and our slender body theory are based on different "selection mechanisms" there is a strong correspondence with the experiments for both. On the basis of this comparison, our array solutions and surface-energy-based tip selection may both be important factors in describing array growth. In order to determine the role of the array solutions and surface energy in array growth, one needs to describe how the array solutions, surface-energy-based tip stability and spacing selection interact in a self-consistent way. We now turn to the issue of how the primary spacing is determined.

While we have demonstrated that prescribing the spacing of the dendrites leads to a unique solution without the need for surface energy, it is not yet clear how the array solutions will interact with the surface energy selection mechanism to determine the spacings in the array. One scenario, depicted in Fig. 6 would be that the array solutions give a family of solutions parameterized by the spacing (or tip radius). The incorporation of a surface-energy-based selection mechanism would require a particular tip radius, thus selecting one member of the family and giving a unique tip radius and dendrite spacing for given solidification conditions. This is the view of array growth employed by Kurz and Fisher [17] and Trivedi [18], but with ad hoc models for the dendrite shape. Our model would

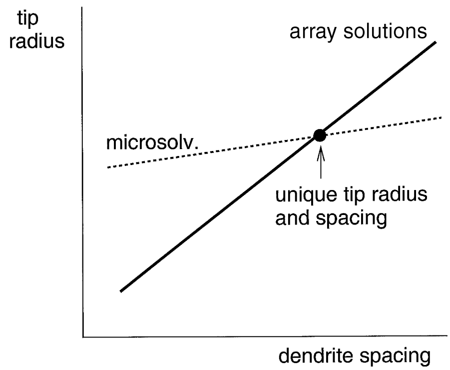

Fig. 6. Array solutions from our model (see Fig. 4 or any of the ad hoc relationships derived from geometric constraints such as the Scheil–Ivantsov model described in the text that have a monotonic relationship between the tip radius and the spacing). The dashed line represents the surface energy selection criteria (either microsolvability or marginal stability) with a relatively weak dependence on the spacing. There is a unique operating state for the array which satisfies both selection criteria and gives a unique dendrite spacing and tip radius.

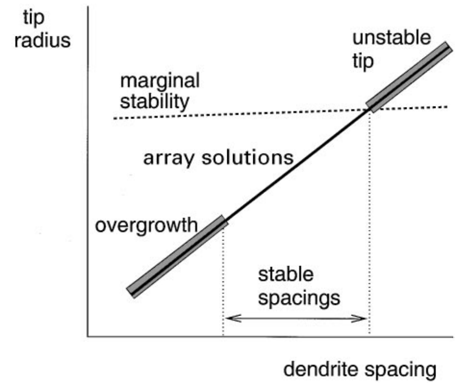

Fig. 7. Array solutions from our model as in Fig. 6. The dashed line represents the marginal stability criteria for the dendrite tip. Dendrites with tip radii above this dashed line are unstable to tip splitting (upper shaded portion of solid line). In addition, spacings below a critical value are unstable to the overgrowth mechanism described in the text (lower shaded portion of solid line). The combined stability boundaries give a range of stable spacings for the array.

give comparable predictions but with a self-consistent dendrite shape.

Another scenario, depicted in Fig. 7, would be that surface energy does not select a unique operating state but rather determines the range of stable spacings. As above, the array solutions give a family of solutions parameterized by the spacing (or tip radius). Marginal stability theory [5] says that the dendrite tip is stable only if its radius does not exceed a critical value determined by surface energy. Thus, of the family of solutions, those with large spacings (corresponding to large tip radii) would be unstable. In essence, this gives an upper bound on the range of stable spacings for the array.

A lower bound on the spacings is set by the overgrowth mechanism. If the dendrites are too closely spaced, the array is unstable to having some members being overgrown by the rest of the array, as shown in the experiments [11] and in theory [13,14]. Thus, of the family of array solutions, the tip instability and overgrowth mechanisms would give a range of stable spacings for the array in agreement with experiments [11,12].

With regard to the upper bound on the spacings, it is acknowledged that in the experiments [11] the instability mechanism is not due to tip splitting but rather due to the outgrowth of tertiary arms. While the tip splitting mechanism described by marginal stability is not the mode of instability, it does provide an *upper bound* on the range of stable spacings: there cannot be an array with a tip radius (and hence spacing) in excess of that prescribed by the marginal stability bound. The actual upper stability bound, corresponding to the outgrowth of tertiary arms, will lie below the tip splitting boundary. Nonetheless, the tip splitting boundary serves as a useful bound on the range of stable spacings in the absence of any theories for tertiary outgrowth.

In either scenario described above, the relationship between the tip characteristics and the array spacings provided by the array solutions would be an important factor in the determination of the dendrite spacing(s).

In summary, we have developed a theory for the directional solidification of an array of dendrites, with the shape of the dendrite influenced by interactions with neighboring dendrites. By determining the explicit details of the solution to the free boundary problem for the dendrite shape, we find that the array interactions determine unique tip characteristics for a given dendrite spacing and remove the indeterminacy in the Ivantsov similarity solution. This family of "array solutions," in which the tip radius is related directly to the dendrite spacing, agrees well with experiment in parameter ranges where the slender dendrite theory is expected to be valid. We suggest that these array solutions, together with surface energy and stability considerations, can describe the selection of a single dendrite spacing or a range of dendrite spacings during directional solidification.

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