

Dendrites, Viscous Fingers, and the Theory of Pattern Formation

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Source: Science, New Series, Vol. 243, No. 4895 (Mar. 3, 1989), pp. 1150-1156

Published by: American Association for the Advancement of Science

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REFERENCES AND NOTES

- 1. R. M. May, Science 241, 1441 (1988).
- 2. F. W. Preston, Ecology 43, 185 (1962); ibid., p. 410; C. B. Williams, Patterns in the Balance of Nature and Related Problems in Quantitative Ecology (Academic Press, New York, 1964); R. M. May, in Ecology and Evolution of Communities, M. L. Cody and J. M. Diamond, Eds. (Harvard Univ. Press, Cambridge, MA, 1975), pp. 81–120; G. Sugihara, Am. Nat. 116, 770 (1980).
- J. E. Cohen, Food Webs and Niche Space (Princeton Univ. Press, Princeton, NJ, 1978);
 S. L. Pimm, Food Webs (Chapman and Hall, London, 1982);
 D. L. DeAngelis, W. M. Post, G. Sugihara, Food Web Theory: Report on a Food Web Workshop (Oak Ridge Natl. Lab., Publ. no. ORNL-5983, Oak Ridge, TN, 1983).
 A. G. Fischer, Evolution 14, 64 (1960);
 G. G. Simpson, Syst. Zool. 13, 413 (1964);
 E. R. Pianka, Am. Nat. 100, 33 (1967);
 R. E. Cook, Syst. Zool. 18, 63 (1969);
 R. H. Machetse, Computing Endow Patterns in the Distriction of Service (Memory).
- H. MacArthur, Geographical Ecology: Patterns in the Distribution of Species (Harper and Row, New York, 1972); J. H. Brown, in Analytical Biogeography, A. A. Myers and P. S. Giller, Eds. (Chapman and Hall, London, in press).
- S. Giller, Eds. (Chapman and Hall, London, in press).
 B. Bruun, Birds of Europe (McGraw-Hill, New York, 1970); E. R. Hall, The Mammals of North America (Wiley, New York, 1980); C. S. Robbins, B. Bruun, H. S. Zim, Birds of North America (Golden Press, New York, 1983); National Geographic Society, Field Guide to the Birds of North America (National Geographic Society, Washington, DC, 1983); J. B. Dunning, Jr., West. Bird Banding Assoc. Monogr. 1, 1 (1984). We also used the North American Breeding Bird Survey, made available by D. Bystrak and C. S. Robbins of the U.S. Fish and Wildlife

- Service.
 J. H. Brown and B. A. Maurer, Am. Nat. 130, 1 (1987).
 G. E. Hutchinson and R. H. MacArthur, ibid. 93, 117 (1959).
 R. M. May, in Diversity of Insect Faunas, L. A. Mound and N. Waloff, Eds. (Blackwell, Oxford, 1978), pp. 188–204.
 J. C. Willis, Age and Area (Cambridge Univ. Press, Cambridge, 1922); E. Rapoport, Aerography: Geographical Strategies of Species (Pergamon, Oxford, 1982).
 G. E. Huchinson, Am. Nat. 93, 145 (1959); M. L. Rosenzweig, J. Mammal. 47, 602 (1966); M. A. Bowers and J. H. Brown, Ecology 63, 391 (1982); T. W. Schoener, in Ecological Communities: Conceptual Issues and the Evidence, D. R. Strong, Jr., D. Simberloff, L. G. Abele, A. B. Thistle, Eds. (Princeton Univ. Press, Princeton, NJ, 1984), pp. 254–281; F. A. Hopf and J. H. Brown, Ecology 67, 1139 (1986). 1139 (1986)
- 11. R. H. MacArthur and E. O. Wilson, *The Theory of Island Biogeography* (Princeton Univ. Press, Princeton, NJ, 1967); T. W. Schoener and D. A. Spiller, *Nature* **330**,

- 474 (1988); D. Simberloff, Annu. Rev. Ecol. Syst. 19, 473 (1988).
- J. H. Brown, Amer. Zool. 21, 877 (1981).
 G. E. Walsberg, in Symposium on Avian Energetics, J. R. King, Ed. (Deutsche Ornithologen-Gesellschaft, Berlin, 1983), pp. 161–220; K. A. Nagy, Ecol. Monogr.
- B. K. McNab, Am. Nat. 97, 113 (1963); T. W. Schoener, Ecology 49, 123 (1968);
 A. S. Harestad and F. L. Bunnell, ibid. 60, 389 (1979).
- W. A. Calder III, Size, Function, and Life History (Harvard Univ. Press, Cambridge, MA, 1984); M. W. Demment and P. J. van Soest, Am. Nat. 125, 641 (1985). J. Damuth, Nature 230, 699 (1981); D. R. Morse, J. H. Lawton, M. M. Dodson, M. H. Williamson, ibid. 314, 731 (1985); D. R. Morse, N. E. Stock, J. H. Lawton, East Engage 13, 25 (1989). Ecol. Entomol. 13, 25 (1988)
- J. H. Brown and Z. Zeng, in preparation.
 R. H. Peters, The Ecological Implications of Body Size (Cambridge Univ. Press, Cambridge 1983); P. H. Harvey and J. H. Lawton, Nature 324, 212 (1987); D. Strayer, Oecologia **69**, 513 (1986).

 19. J. H. Brown and B. A. Maurer, Nature **324**, 248 (1987).

 20. B. A. Maurer and J. H. Brown, Ecology **69**, 1923 (1988).

- M. L. Cody, in Ecology and Evolution of Communities, M. L. Cody and J. M. Diamond, Eds. (Harvard Univ. Press, Cambridge, MA, 1975), pp. 214–257; J. H. Brown, in Organization of Communities: Past and Present, J. H. R. Gee and P. S. Giller, Eds. (Blackwell, Oxford, 1987), pp. 185–203; J. Roughgarden, S. D. Gaines, S. W. Pacala, in Organization of Communities: Past and Present, J. H. R. Gee and P. S. Giller, Eds. (Blackwell, Oxford, 1987), pp. 491-518; T. Root, Ecology 69, 330
- R. P. Neilson and L. H. Wullstein, J. Biogeogr. 10, 275 (1983); J. Roughgarden, S. Gaines, H. Possingham, Science 241, 1460 (1988).
 S. M. Stanley, Macroevolution: Pattern and Process (Freeman, San Francisco, 1979); J. Sepkoski, Jr., Paleobiology 10, 246 (1984); D. Jablonski, Science 231, 129 (1986)
- D. R. Brooks, Ann. Mo. Bot. Gard. 72, 660 (1985).
 P. M. Vitousek, P. R. Ehrlich, A. H. Ehrlich, P. A. Matson, BioScience 36, 368
 - (1986); D. H. Wright, J. Biometeorol. 31, 293 (1987)
- We thank the many people who helped to assemble and analyze the data and those who have discussed these ideas with us. P. Nicoletto, J. F. Merrit, and J. S. Findley contributed data for Fig. 1. G. Ceballos, G. Farley, J. Findley, L. Hawkins, A Kodric-Brown, P. Nicoletto, E. Toolson, and anonymous reviewers made helpful comments on the manuscript. The support of NSF grants BSR-8506729 and BSR-8718139 and Department of Energy grant FG02-86ER60424 is gratefully ac-

Dendrites, Viscous Fingers, and the Theory of **Pattern Formation**

J. S. Langer

There has emerged recently a new theoretical picture of the way in which patterns are formed in dendritic crystal growth and in the closely analogous phenomenon of viscous fingering in fluids. Some interesting questions that arise in connection with this theory include: How broad is its range of validity? How do we understand dynamic stability in systems of this kind? What is the origin of sidebranches? Can weak noise, or even microscopic thermal fluctuations, play a role in determining the macroscopic features of these systems?

HE THEORY OF PATTERN FORMATION IN NONLINEAR DISSIpative systems has taken some surprising turns in the last several years. One of the most interesting developments has been the discovery that weak capillary forces act as singular perturbations which lead to beautifully delicate and very nearly identical selection mechanisms both in dendritic crystal growth and in the fingering patterns which emerge when a viscous fluid is displaced by a less viscous one. It now appears likely that important progress has been made, but pieces of the puzzle still seem to be missing.

For most of us, dendritic crystal growth brings to mind pictures of snowflakes. Materials scientists may think also about metallurgical microstructures, which provide very practical reasons for research in this field; but it is the snowflake that most quickly captures our imaginations. Kepler's 1611 monograph "On the Six-Cornered Snowflake" (1) is often cited as the first published work in which morphogenesis—the spontaneous emergence of patterns in nature—was treated as a scientific rather than a theological topic. At a time in which the existence of atoms was merely speculation, Kepler mused about hexagonal packings of spheres, but concluded that the

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problem was beyond his reach. Its solution would have to be left for future generations. In fact, scientists have waited more than three centuries before finding much hint of an answer to the question that Kepler posed.

One part of the answer, of course, is the understanding of crystalline symmetries and their relation to atomic structure. Another part is our modern statistical theory of the fluctuations and dissipative processes that ultimately govern pattern formation. But it is only in very recent years that we have begun to understand how these irreversible processes can amplify weak anisotropies and even very small noisy fluctuations in such a way as to produce intricate patterns in ostensibly featureless systems.

In the pattern-forming systems of interest here, we are dealing with dynamic processes, not just molecular structures or macroscopic forms. Unlike D'Arcy Thompson (2) (who could describe and measure but not explain) or Nakaya (3) (who produced one of the world's most complete and beautiful catalogues of snowflakes), we now have the experimental and analytic tools that we need to find out, for example, how the growth rate of a dendrite and the spacing between its sidebranches are determined by the temperature and composition of the solidifying substance. We may even have most of the tools—if not yet the information—that we need to understand the growth of biological forms. Of the analytic tools, the two which seem most essential are the theory of morphological instabilities in systems far from equilibrium, and the computer, which enables us to explore quantitatively the nonlinear behavior of such systems. [For both of these we must pay tribute to the remarkable insights of Turing (4).] The work to be described here arises largely from the modern interplay between physical insight, mathematical analysis, and numerical methods.

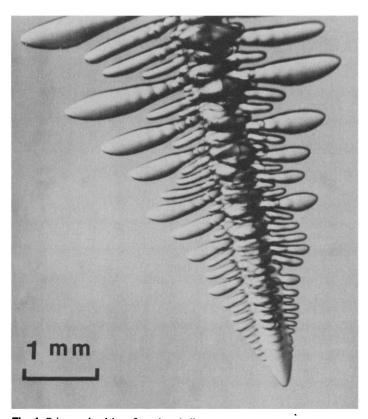


Fig. 1. Primary dendrite of succinonitrile (a transparent plastic crystal with cubic symmetry) growing in its undercooled melt. Note the smooth paraboloidal tip, the secondary sidebranching oscillations emerging behind the tip, and the beginnings of tertiary structure on the well-developed secondaries. (Photograph courtesy of M. E. Glicksman.)

In this article, I shall review briefly the recent history of the dendrite (5) and viscous fingering problems and shall attempt to communicate at least the general flavor of recent developments, specifically, the so-called "solvability theory" (6). As an illustration of this theory, I shall describe Couder's remarkable bubble effect, which, by seemingly turning fingers into dendrites, provides an excellent illustration of the singular perturbation in action. I shall conclude with some conjectures about the range of validity of the solvability theory and its implications for our understanding of more complex dynamical effects such as sidebranching.

Dendritic Solidification of a Pure Substance

In the conventional thermodynamic model of the solidification of a pure substance from its melt, the fundamental rate-controlling mechanism is the diffusion of latent heat away from the interface between the liquid and solid phases. The latent heat that is released in the transformation warms the material in the neighborhood of the solidification front and must be removed before further solidification can take place. This is a morphologically unstable process which characteristically produces dendrites, that is, treelike or snowflakelike structures. In a typical sequence of events, an initially featureless crystalline seed immersed in an undercooled melt develops bulges in crystallographically preferred directions. The bulges grow into needleshaped arms whose tips move outward at constant speed. These primary arms are unstable against sidebranching and the sidebranches, in turn, are unstable against further sidebranching, so that each outward growing tip leaves behind itself a complicated dendritic structure like that shown in Fig. 1.

The dimensionless thermal diffusion field in this model for convenience is chosen to be

$$u = \frac{T - T_{\infty}}{(L/\epsilon)} \tag{1}$$

where T_{∞} is the temperature of the liquid infinitely far from the growing solid, and the ratio of the latent heat L to the specific heat c is an appropriate unit of undercooling. The field u satisfies the diffusion equation

$$\frac{\partial u}{\partial t} = D\nabla^2 u \tag{2}$$

where *D* is the thermal diffusion constant, which can be taken to be the same in both liquid and solid phases. The remaining ingredients of the model are the boundary conditions imposed at the solidification front. First, there is heat conservation:

$$\nu_{\mathbf{n}} = -[D\hat{\mathbf{n}} \cdot \nabla u] \tag{3}$$

where $\hat{\mathbf{n}}$ is the unit normal directed outward from the solid, ν_n is the normal growth velocity, and the square brackets denote the discontinuity of the flux across the boundary. In these units, the left-hand side of Eq. 3 is the rate at which latent heat is generated at the boundary and the right-hand side is the rate at which it is being diffused away. The physically more interesting boundary condition is the statement of local thermodynamic equilibrium, which determines the temperature u_s at the two-phase interface:

$$u_{\rm s} = \Delta - d_0 \kappa \tag{4}$$

where

$$\Delta = \frac{T_{\rm M} - T_{\rm \infty}}{(L/c)} \tag{5}$$

and $T_{\rm M}$ is the melting temperature. Δ is the dimensionless under-

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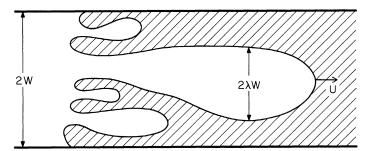


Fig. 2. Schematic illustration of a Hele-Shaw experiment adapted from a photograph by J. Maher. The inviscid fluid is entering from the left and the viscous fluid (shown shaded) is being pushed to the right. The motion is effectively two-dimensional, constrained by narrowly separated glass plates in the plane of the figure. As shown here, the flow takes place in a channel of width 2W. The initially irregular pattern formed by the instability of the interface between the two fluids is developing into a single finger of width $2\lambda W$.

cooling, a measure of the driving force for the processes that we are considering. The second term on the right-hand side of Eq. 4 is the Gibbs-Thomson correction for the melting temperature at a curved surface: κ is the sum of the principal curvatures and $d_0 = \gamma \epsilon T_{\rm M}/L^2$ is a length, ordinarily of order angstroms, which is proportional to the solid-liquid surface tension γ . The latter quantity and, accordingly, d_0 may be functions of the angle of orientation of the interface relative to the axes of symmetry of the crystal. In particular, for a cubic crystal in the (1, 0, 0) plane, d_0 is proportional to $(1 - \alpha \cos 4\theta)$, where θ is the angle just mentioned and α is a measure of the strength of the anisotrophy.

Viscous Fingering

The hydrodynamic analog of dendritic solidification is the fingering instability that occurs when one causes an inviscid fluid to drive a viscous one through a porous medium. The two-dimensional version of this situation (7) is a Hele-Shaw cell in which the two immiscible fluids are constrained to move between narrowly separated parallel plates. The configuration is shown schematically in Fig. 2. The invading inviscid fluid can be visualized as playing the role of the growing solid, and the more viscous fluid that is being pushed away is like the melt. The analog of the thermal field u is the pressure P, which can be taken to be constant in the "solid" and to satisfy Laplace's equation in the "melt." Here is the main difference between fingering and solidification; the Laplace equation is not the diffusion equation. The velocity of the viscous fluid in the porous medium is given by Darcy's law to be simply proportional to $-\nabla P$; thus the expression for the velocity of the interface between the two fluids is precisely the analog of the conservation law, Eq. 3. Finally, the interfacial tension γ causes the pressure at the interface to be reduced by an amount proportional to γκ, in exact analogy to the thermodynamic boundary condition, Eq. 4. There is, however, no crystalline anisotrophy associated with this y. Directional information can be provided only by the interaction between the longranged pressure field and the walls of the container, or else by adding to the model—"by hand," so to speak—some anisotropy of the medium through which the fluids are moving.

Pattern Selection

There are sharply defined problems of pattern selection associated with both of these models. In solidification, it is known that the growth rate ν and the tip radius ρ of a dendrite are determined uniquely by the undercooling Δ . In the hydrodynamic case, specifically, the two-dimensional Saffman-Taylor (7) experiment in which a steady-state finger forms in a long channel, the ratio λ of the width of the finger to the width of the channel is determined uniquely by the flow speed. In both cases, surface tension appears at first glance to be a negligible perturbation; the length d_0 is orders of magnitude smaller than other characteristic lengths. However, the omission of surface tension in either problem leads to continuous families of solutions and, thus, to no explanation whatsoever of the experimentally observed selection principles. It turns out that surface tension is playing an especially subtle role in these processes.

In the case of the dendrite, if one neglects surface tension altogether, one arrives at Ivantsov's paradox (8). Instead of there being a unique growth velocity ν and tip radius ρ at a fixed Δ , as required by experiment, there exists a continuous family of steady-state, shape-preserving solidification fronts—paraboloids of revolution—that satisfy the Ivantsov relation

$$\Delta = p \ e^{p} \int_{p}^{\infty} d\gamma \, \frac{e^{-\gamma}}{\gamma} \tag{6}$$

where $p = \rho v/2D$ is the thermal Péclet number. The tips of dendrites often do look very paraboloidal, and quantitative experiments generally indicate that the Ivantsov relation, Eq. 6, is satisfied. But obviously some essential ingredient of the theory is missing.

Over a decade ago, Müller-Krumbhaar and I (9) explored the idea [originally suggested by Oldfield (10)] that the missing element of the theory might have something to do with stability of the growth form. We performed many complicated calculations, but what was left in the end was a relatively simple conjecture that has since been confirmed remarkably well by experiment. In the simplest possible terms, our conjecture was that the tip radius ρ might scale like the Mullins-Sekerka (11) wavelength $\lambda_s = 2\pi (2Dd_0/v)^{1/2}$. Note that λ_s is the geometric mean of the microscopic capillary length d_0 and the macroscopic diffusion length 2D/v; it is of roughly the right magnitude to characterize dendritic structures. A planar solidification front moving at speed ν is linearly unstable against sinusoidal deformations whose wavelengths are larger than λ_s . Therefore, we reasoned, a dendrite with tip radius ρ appreciably greater than λ_s must be unstable against sharpening or splitting. The dynamical process that leads to the formation of the dendritic tip might naturally come to rest at a state of marginal stability, that is, at a state for which the dimensionless group of parameters

$$\sigma = \frac{2Dd_0}{\nu\rho^2} = \left(\frac{\lambda_s}{2\pi\rho}\right) \tag{7}$$

is a constant, independent of Δ . Moreover, if we take the idea literally and set ρ equal to λ_s , then the value of this constant should be $\sigma^* \cong (1/2\pi)^2 \cong 0.025$. The assumption $\sigma = \sigma^* = \text{constant}$ is consistent with a wide range of experimental observations (12) (when convective effects are eliminated or otherwise taken into account) and the specific value $\sigma^* \cong 0.0195$ for succinonitrile—by far the most carefully studied material—is quite close to the naïve prediction.

What, then, is wrong with the marginal stability theory? It seems that its mathematical foundation has been knocked from under it by the discovery that the Ivantsov family of solutions does not survive in the presence of surface tension (13–17). A nonvanishing d_0 , no matter how small, reduces the continuum of solutions to, at most, a discrete set; and the existence of any solution whatsoever depends on there being some angular dependence of the surface tension, that is, a nonvanishing anisotropy strength α . Thus, the stability calculation that Müller-Krumbhaar and I thought we were performing was

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unfounded because the family of steady-state solutions whose stability we supposedly were testing did not exist.

All is not lost, however, because the mathematics immediately suggests an alternative selection mechanism, albeit one that has little of the intuitive appeal of marginal stability. A natural guess is that the selected dendrite is the one for which a stable solution exists. In more formal language, we guess that the condition for solvability of the steady-state equations is equivalent to a condition for the existence of a stable fixed point with a large basin of attraction in the space of configurations of this dynamical system. If this conjecture is correct, orderly, steady-state dendritic growth does not occur at all in isotropic materials. In suitably anisotropic systems, a growing body of analytic and computational evidence suggests that there is a denumerably infinite set of solutions, and that only the fastest (and thus sharpest) of these solutions can be dynamically stable. The hypothesis that this unique solution exists and that it describes the tip of a dynamically selected dendrite has come to be known as the "solvability theory."

Special Features of the Solvability Theory

This is not the place for a detailed exposition of the mathematics of the solvability theory, but there are several features that do need to be mentioned. In the limit of small Péclet number p, the controlling group of parameters in the theory is the same quantity σ , defined in Eq. 7, that appeared in the stability analysis. This happens because one is looking for a small surface tension—induced correc-

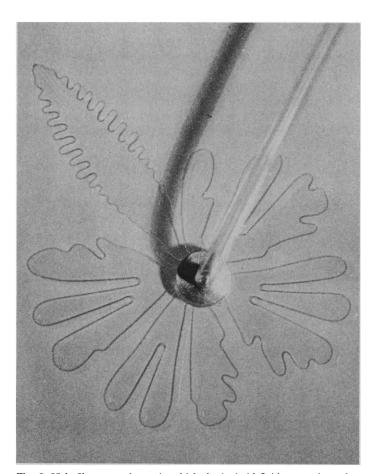


Fig. 3. Hele-Shaw experiment in which the inviscid fluid enters through a central orifice and forms fingers that move radially outward. One of these fingers has trapped a small bubble at its tip. As a result, it is growing stably at constant speed and is emitting sidebranches. (Photograph courtesy of Y. Couder.)

tion to the shape of the Ivantsov parabola and, in computing this correction, one encounters an equation quite similar to the one which arises in linear stability theory. [As shown by Pomeau and coworkers (15), linearization is not a necessary ingredient of the argument for solvability.] To be precise, σ enters the theory as a singular perturbation; it describes the strength of the curvature effect in Eq. 4 and, accordingly, multiplies the highest derivative in the equation for the shape correction once one has reduced this equation to dimensionless form.

There is a very nice way to visualize the effect of this perturbation. In practical numerical calculations (18–20), and also in the analytic approaches that have been applied successfully to this problem (14, 17), one can generally assure the existence of some kind of solution by relaxing a boundary condition—most commonly the condition of smoothness at the tip. Suppose one allows the tip to have a cusp of outer angle Θ and then, either numerically or analytically, computes what value Θ must have in order to achieve a solution at a given value of σ . Because Θ must vanish for a physically acceptable solution, a formally exact statement of the solvability condition is

$$\Theta(\sigma, p, \alpha) = 0 \tag{8}$$

We may think of Θ as a measure of how close we have come to finding a solution at an arbitrary value of σ . The special values of σ for which Eq. 8 is satisfied are denoted $\sigma^*(p,\alpha)$.

If one tries to compute Θ by expanding it in powers of σ , one finds that Θ vanishes at all orders, a result that would be consistent with the original expectation of a continuous family of solutions. If the calculation is performed more carefully, however, the answer— τ small p and zero anisotropy α —has the form

$$\Theta(\sigma,0,0) \propto \exp\left(-\frac{\text{constant}}{\sqrt{\sigma}}\right)$$
 (9)

This function has an essential singularity at $\sigma=0$ and no possible expansion about that point. It is extremely small for small σ , but it does not vanish exactly unless $\sigma=0$. Thus, an arbitrarily small amount of isotropic surface tension destroys all solutions. For small, positive anisotropy α , however, the function $\Theta(\sigma)$ has the same form as Eq. 9 for large σ but oscillates rapidly in the limit $\sigma\to 0$. The largest value of σ at which Θ passes through zero occurs at $\sigma=\sigma^*\propto \alpha^{7/4}$, the latter approximation being valid only in the limit of very small α .

The solvability theory for the Saffman-Taylor (21–23) problem is strikingly similar to the analysis for the dendrite. In this case, the system is automatically in the limit $p \to 0$ because $p = \nu \rho/2D$ and the diffusion equation, Eq. 2, reduces to the Laplace equation in the limit $D \to \infty$. The parameter σ is replaced by the dimensionless group of parameters σ_{ST} :

$$\sigma_{ST} = \frac{\gamma b^2 \pi^2}{12\mu U W^2 (1 - \lambda)^2}$$
 (10)

where γ is the surface tension, b the spacing between the plates, μ the viscosity, U the speed of the finger, 2W the width of the channel, and $2\lambda W$ the width of the finger. All other essential ingredients of the solvability function $\Theta_{ST}(\sigma,\lambda)$ defined in analogy to $\Theta(\sigma,p\to 0,\alpha)$ are the same except that the function $(1-\alpha\cos4\theta)$ is replaced by a function of θ and λ . It then turns out that the boundary-related quantity $\lambda-1/2$ plays a role in this problem that is closely analogous to that played by the anisotropy strength α for the dendrite. For $\lambda<1/2$, Θ_{ST} looks like Θ in Eq. 9 and there are no solutions of $\Theta_{ST}=0$. For $\lambda<1/2$, on the other hand, Θ_{ST} oscillates for small values of σ_{ST} , and the physically meaningful solution of the solvability condition has the form σ_{ST}^* ∞ $(\lambda-1/2)^{3/2}$. The convergence of λ to the value 1/2 at small σ_{ST} (large U) is consistent with experiment (7).

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The picture summarized above—an apparently accurate analytic description of a mechanism for selecting steady-state configurations in two different nonlinear dissipative systems—seems elegant and mathematically compelling. The mathematics looks especially sound in view of recent work of Combescot et al. (22) in which a nonlinear formulation originally suggested by Kruskal and Segur (24) has been developed into an amazingly complete solution of the viscous fingering problem. There remains the possibility, however, that the solvability theory might be mathematically correct but physically irrelevant—that real systems might simply ignore these steady-state solutions and find other, perhaps oscillatory or even irregular, states of motion. With this possibility in mind, let us consider some of the evidence regarding the validity—or lack thereof—of the solvability theory.

Dendritic Growth Rates

First, there is the question of whether the solvability theory really agrees with experiment for the dendrites and, if so, what is its range of validity? Experiments (12) indicate that the parameter σ , as predicted, is a Δ -independent constant at small Δ (apart from corrections at very small Δ where convection in the melt becomes important). The solvability theory also provides a natural explanation for the previously unexplained fact that free dendrites grow only in directions parallel to crystalline axes of symmetry; lack of symmetry precludes the existence of solutions in other directions.

The trouble is that we do not know yet whether the values of σ^* predicted by the theory agree quantitatively with those found experimentally. At the moment, the available evidence seems inconclusive, and we are waiting both for new measurements and for more extensive, three-dimensional calculations. A particularly worrisome aspect of the situation is that the theory predicts a strong dependence of σ^* on the anisotropy strength α ; specifically, σ^* is predicted to be proportional to $\alpha^{7/4}$ in the limit $\alpha \to 0$ and to be roughly linear in α for most of its accessible range of values (19, 25, 26). No such strong dependence on anisotropy has so far been confirmed experimentally.

In my opinion, it is most likely that the solvability theory will turn out to be a correct description of a large but limited class of relatively simple dendritic phenomena. It may break down in complex situations where competing processes such as thermal and solutal diffusion might produce time-dependent behavior that would be invisible in the present steady-state theory. It may also break down at large crystalline anisotropies where the solvability calculations become extremely difficult and perhaps intrinsically impossible. Almost certainly, the solvability theory will fail at small anisotropies, and the $\alpha^{7/4}$ law will turn out not to be physically meaningful. The last conjecture is based on considerations of stability that deserve a few paragraphs of their own. I shall return to that topic shortly.

Couder's Bubbles

A second category of evidence regarding validity of the solvability theory is the bubble effect discovered by Couder and co-workers (27), which indicates that something very much like solvability is occurring in variants of the Saffman-Taylor problem. For both the dendrite and the Saffman-Taylor finger, physically acceptable solutions require $\Theta = 0$, that is, the structure is not allowed to have a cusplike discontinuity at its tip. However, if one were able to perturb the system in such a way as to fix Θ at some nonvanishing positive value, then the mathematics tells us that dendrites should exist in the absence of anisotropy and that viscous fingers should

occur with relative widths λ less than 1/2. Couder *et al.* have produced such perturbations of the fingers by attaching small bubbles to their tips, and in this way have succeeded in observing anomalously small values of λ . Their results for the dependence of λ on the channel width W (a function whose form should not depend on details of the flow in the neighborhood of the tip) are in excellent agreement with the solvability theory (23). Moreover, in the circularly symmetric geometry where radial fingers ordinarily suffer tip-splitting instabilities, they have shown that fingers with bubbles at their tips behave very much like dendrites, complete with sidebranches! A picture of such a finger—behaving like a dendrite—is shown in Fig. 3.

Stability and Sidebranching

Perhaps the most dramatic of the conceptual developments stemming from the solvability theory is a growing understanding of the dynamics of pattern-forming systems. In particular, we are beginning to understand the stability of dendritic tips and the manner in which perturbations of these tips may be amplified to form complex arrays of sidebranches.

Note the following apparent paradox. It has been known for some time that, in the absence of surface tension ($\sigma=0$), Ivantsov's needlelike solutions of the solidification problem are manifestly unstable (9). In fact, the $\sigma=0$ problems for both the dendrite and the viscous finger are not even dynamically well defined because interfaces destabilized arbitrarily rapidly at arbitrarily short-length scales. On the other hand, the most complete stability analyses performed to date (28) indicate that the tips of fingers and dendrites remain linearly stable at all nonzero values of $\sigma=\sigma^*$. How can it happen that an indefinitely small amount of surface tension can so completely change the behavior of this system?

The answer to this question, and to several others of related interest, can be seen in the result of a simple calculation. It will be convenient to describe this calculation in terms appropriate to the dendrite; the analogous result for the viscous finger is slightly different in technical aspects that need not concern us here.

In principle, the correct way to study stability of a moving, openended system like the dendrite is to look at its response to a localized perturbation, for example, a short pulse of heat applied near the tip. The analysis that is needed for this purpose is similar to that used by Zel'dovich and colleagues to study the stability of flame fronts (29). To linear order in the deviation from a steady-state solution determined by solvability, we find that this pulse generates a wave-packet-like deformation whose center moves away from the tip as shown schematically in Fig. 4. More precisely, the center of the wavepacket stays at a fixed position along the side of the dendrite as viewed in the laboratory frame of reference, while the tip grows at constant speed away from the perturbation.

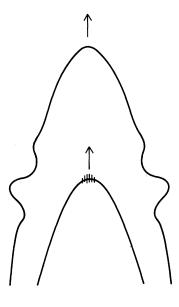
This wavepacket has several important properties (30–32). First, its amplitude A(s) continues to grow as its center moves away from the tip. More specifically,

$$A(s) \approx \exp\left[\frac{0.647}{\left(\sigma^*\right)^{1/2}} \left(\frac{s}{\rho}\right)^{1/4}\right] \tag{11}$$

where s is the distance measured along the front from the tip of the dendrite to the center of the packet. Equation 11 is an asymptotic estimate valid for $s \gg \rho$. Second, the packet spreads and stretches in such a way that, as it grows, it acquires a sharply defined wavelength that increases slowly with distance from the tip. Finally, although this deformation grows as it moves, it leaves the tip of the dendrite unchanged after a sufficiently long time. That is, any point on the

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Fig. 4. Schematic illustration of two stages in the growth of a localized sidebranching deformation. An initial noisy pulse is indicated at the tip of an otherwise unperturbed parabolic needle crystal. At a later time, the tip has regained its shape and has moved beyond the point of perturbation, leaving behind it a smooth wavepacket that will grow into side-



solidification front at a fixed distance from the tip ultimately will return to its original position after the wavepacket has passed. This is the sense in which the front is stable despite the fact that sidebranches continue to grow.

A crucial aspect of Eq. 11 is its singular dependence on σ^* , which is reminiscent of the σ -dependence of the solvability function in Eq. 9. We immediately can see from this how the crossover occurs from stability at $\sigma^* > 0$ to manifest instability at $\sigma^* = 0$; as σ^* becomes small, perturbations become increasingly amplified before leaving the neighborhood of the tip. We also can see why the solvability theory is likely to break down at small anisotropy strength α . If taken literally at arbitrarily small a, the theory predicts arbitrarily small values of σ^* and, according to Eq. 7, tip radii ρ that are much larger than the stability length λ_s . Intuition tells us that such tips should be unstable. According to Eq. 11, the linear instability is controlled because the perturbation moves away from the tip but not until it has grown by an amount which may be large enough to carry it beyond the limits of validity of the linear theory. It seems likely, therefore, that dendrites with small crystalline anisotropies are nonlinearly unstable. Whether or not dendritic behavior occurs in such situations is unknown at present. Perhaps such systems find stable oscillatory modes of growth, or perhaps sufficiently isotropic materials always form chaotic patterns when they solidify in undercooled melts.

The above considerations lead naturally to a theory of sidebranching. Until quite recently, most workers in this field had assumed that the tip of a real dendrite must be weakly—perhaps marginallyunstable against some oscillatory mode of deformation, and that this oscillation must generate the train of sidebranches that seems always to be observed in these systems. Couder's fingers with bubbles at their tips, when driven fast enough, quite definitely do oscillate and emit coherent trains of sidebranches. As mentioned above, however, neither the theorists nor the experimentalists have found any evidence for oscillatory tip modes in the purely thermal dendrites that we have been considering.

One possibility that is suggested by the properties of the wavepacket described above is that dendritic sidebranches are generated by the selective amplification of noise (30, 32-34). In order to construct a satisfactory theory of sidebranching, it seems that we need only to identify the pulses that generate wavepackets with the ambient noise—perhaps just the thermal fluctuations—in the solidifying material. If we look at some fixed distance behind the tip, say, at the point where initially very small deformations have grown out

of the linear regime and are big enough to be visible, then it turns out that only a relatively narrow band of wavelengths has been selected from the original broad-band perturbation. This is what is meant by selective amplification; small, noisy perturbations near the tip produce large deformations away from the tip that look very much like sidebranches. One can even estimate the noise temperature required to generate the sidebranches that are seen experimentally. Purely thermal noise seems too small according to present best estimates, but only by about one order of magnitude. The important lesson is that the dendrite is an extremely sensitive and selective amplifier of weak fluctuations in its environment.

Snowflakes

In conclusion, let us return to Kepler and ask what we now might tell him about snowflakes. We know that snowflakes, at least those that seem aesthetically attractive to us, are flat, hexagonal, ice crystals that have grown under conditions in which dendritic instabilities have taken place at the six corners. (The actual growth mechanisms for real ice crystals are more complex than any I have described in this article, but I do not think that these technical differences are relevant to the main points that I want to make.) We understand why these dendritic arms of snowflakes can grow only along the six preferred crystalline axes, and we know that their precise behaviortheir growth rates, their thicknesses, the spacings of their sidebranches, and so on—are extremely sensitive to small changes in the temperature and humidity of the vapor out of which they are being formed. Because these conditions are very nearly uniform across the millimeter or less that is occupied by a growing snow crystal, the six branches of a single snowflake will be nearly—but usually not quite-identical to one another. On the other hand, because the atmosphere in a snowstorm is generally turbulent on scales of meters and more, each tiny crystal encounters a different sequence of growth conditions. Thus, no two crystals, not even if they have started from neighboring seeds, are likely to be identical to one another. Of all the new ideas we have learned recently about pattern formation, I think it may be this quantitative understanding of the close relationship between instability and diversity that will turn out to be the most important.

REFERENCES

- 1. J. Kepler, The Six-Cornered Snowflake (Clarendon Press, Oxford, 1966) [translated by C. Hardie, originally published as De Nive Sexangula (Godfrey Tampach, Frankfurt am Main, 1611)].
- D'Arcy Wentworth Thompson, On Growth and Form (Cambridge University Press, Cambridge, 1944).
- U. Nakaya, Snow Crystals (Harvard University Press, Cambridge, MA, 1954).
- A. Hodges, Alan Turing, the Enigma (Simon and Schuster, New York, 1983) A review of the physics of pattern formation in crystal growth can be found in J. S.
- Langer, Rev. Mod. Phys. **52**, 1 (1980).

 6. For more detailed reviews of recent developments, see J. S. Langer, in Chance and Matter (Lectures in the Theory of Pattern Formation, Les Houches Summer School, 1986), J. Souletie, J. Vannimenus, R. Stora, Eds. (North-Holland, New York, 1987), pp. 629–711; D. Kessler, J. Koplik, H. Levine, Adv. Phys. 37, 255 (1988); in Dynamics of Curved Fronts, P. Pelcé, Ed. (Academic Press, New York, 1988)
- P. G. Saffman and G. I. Taylor, Proc. Roy. Soc. A 245, 312 (1958); J. W. McLean and P. G. Saffman, J. Fluid Mech. 102, 455 (1981). For reviews of modern developments in the viscous fingering problem, see D. Bensimon, L. P. Kadanoff, S. Liang, B. Shraiman, C. Tang, Rev. Mod. Phys. **58**, 977 (1986) or Kessler et al.
- G. P. Ivantsov, Dokl. Akad. Nauk SSSR 58, 567 (1947).
- J. S. Langer and H. Müller-Krumbhaar, Acta Metall. 26, 1681; 1689; 1697 (1978); H. Müller-Krumbhaar and J. S. Langer, ibid. 29, 145 (1981).
- W. Oldfield, Mater. Sci. Eng. 11, 211 (1973).
 W. W. Mullins and R. F. Sekerka, J. Appl. Phys. 34, 323 (1963); ibid. 35, 444 (1964).
- M. E. Glicksman, R. J. Shaefer, J. D. Ayers, Metall. Trans. A 7, 1747 (1976); S. C. Huang and M. E. Glicksman, Acta Metall. 29, 701 and 717 (1981); M. E. Glicksman, Mater. Sci. Eng. 65, 45 (1984); A. Dougherty and J. P. Gollub, Phys.

3 MARCH 1989 ARTICLES 1155

- Rev. A 38, 3043 (1988). For a counterexample, see J. H. Bilgram, M. Firmann, W. Känzig, Phys. Rev. B 37, 685 (1988).
 13. E. Ben-Jacob, N. Goldenfeld, J. S. Langer, G. Schön, Phys. Rev. A 29, 330 (1984).
 14. J. S. Langer, ibid. 33, 435 (1986).

- P. Pelcé and Y. Pomeau, Stud. Appl. Math. 74, 245 (1986); M. Ben Amar and Y. Pomeau, Europhys. Lett. 2, 307 (1986).
 D. Kessler, J. Koplik, H. Levine, in Proceedings of the NATO Advanced Research
- Workshop on Patterns, Defects, and Microstructures in Non-Equilibrium Systems (Austin, Texas, March 1986), ASI Series E, 121, D. Walgraef, Ed. (Nijhoff, Dordrecht,

- A. Barbieri, D. C. Hong, J. S. Langer, *Phys. Rev. A* 35, 1802 (1987).
 D. Meiron, *ibid.* 33, 2704 (1986).
 M. Ben-Amar and B. Moussallam, *Physica D* 25, 7 (1987); *ibid.*, p. 155.
 D. A. Kessler, J. Koplik, H. Levine, *Phys. Rev. A* 33, 3352 (1986); D. A. Kessler and H. Levine, Phys. Rev. B 33, 7867 (1986).
- B. I. Shraiman, *Phys. Rev. Lett.* 56, 2028 (1986).
 R. Combescot, T. Dombre, V. Hakim, Y. Pomeau, A. Pumir, *ibid.*, p. 2036; *Phys. Rev. A* 37, 1270 (1988).
- D. C. Hong and J. S. Langer, Phys. Rev. Lett. 56, 2032 (1986); Phys. Rev. A 36, 2325 (1987).
- 24. See the discussion of the work of M. Kruskal and H. Segur, in (6).
- 25. A. Barbieri and J. S. Langer, Phys. Rev. A, in press.

- 26. D. Kessler and H. Levine, Phys. Rev. A 36, 4123 (1987).
- Y. Couder, N. Gerard, M. Rabaud, *ibid*. 34, 5175 (1986); Y. Couder, O. Cardoso, D. Dupuy, P. Tavernier, W. Thom, *Europhys. Lett.* 2, 437 (1986).
 D. Kessler and H. Levine, *Phys. Rev. A* 33, 2621 (1986); *ibid.*, p. 2634 (1986);
- Europhys. Lett. 4, 215 (1987).
- Ya. B. Zel'dovich, A. G. Istratov, N. I. Kidin, V. B. Librovich, Combust. Sci. Technol. 24, 1 (1980).
- 30. R. Pieters and J. S. Langer, Phys. Rev. Lett. 56, 1948 (1986); R. Pieters, Phys. Rev. A. 37, 3126 (1988).
- 31. M. Barber, A. Barbieri, J. S. Langer, Phys. Rev. A 36, 3340 (1987).
- 32. J. S. Langer, *ibid.*, p. 3350.
 33. R. Deissler, *J. Stat. Phys.* 40, 371 (1985).
- 34. The relation between noise and sidebranching has been examined experimentally by A. Dougherty, P. D. Kaplan, J. P. Gollub, *Phys. Rev. Lett.* 58, 1652 (1987); see also A. Dougherty and J. P. Gollub, *Phys. Rev. A* 38, 3043 (1988).
- This article is based in large part on the text of the author's Marian Smoluchowski Memorial Lecture presented in Warsaw, Poland, on 17 March 1988. I thank the Polish Academy of Sciences for its hospitality. I also thank Y. Couder, M. Glicksman, and J. Maher for providing photographs and information. The research described here was supported by U.S. Department of Energy grant DE-FG03-84ER45108 and by National Science Foundation grant PHY 82-17853, supplemented by funds from the National Aeronautics and Space Administration.

Unity in Function in the Absence of Consensus in Sequence: Role of Leader Peptides in Export

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Passage of proteins across membranes during export from their site of synthesis to their final destination is mediated by leader peptides that paradoxically exhibit a unity of function in spite of a diversity of sequence. These leader peptides act in at least two stages of the export process: at entry into the pathway and subsequently during translocation across the membrane. How selectivity is imposed on the system in the absence of a consensus among the sequences of leader peptides is the main issue discussed here.

IOLOGICAL MEMBRANES NOT ONLY DEFINE THE BOUNDaries between cells and their environments, but also bring about essential separation of functions within cells by establishing internal compartments. Thus, the main role of membranes, for which the hydrophobic center of the lipid bilayer is responsible, is that of a barrier to the passage of water-soluble molecules. However, membranes cannot be inviolate barriers, since besides being selectively permeable to small molecules and ions, they must also allow passage of selected proteins in order to maintain functional organelles and to mediate secretion.

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Common Themes in Protein Export

The transfer of polypeptides across membranes is thus fundamental to life. Studies of such transfer in a number of different systems have revealed significant similarities, suggesting that there is a single basic mechanism on which secondary system-specific differences are imposed. The passage of polypeptides across the membranes of the endoplasmic reticulum during secretion in eukaryotes and the export of proteins from the cytoplasm to the periplasm and outer membrane in gram-negative bacteria such as Escherichia coli are so alike that each of the two systems appears able to recognize and transfer the proteins handled by the other: thus, findings established in investigations of eukaryotic secretion are usually directly applicable to export from E. coli and vice versa. Here our discussion is based predominantly on information obtained from studies of the latter.

Models for Export

As has long been known, polypeptides that are exported from bacteria or that are secreted from eukaryotic cells transiently carry at their amino termini stretches of amino acids (designated signal or leader sequences) that play a critical role in these processes (1, 2). In spite of intensive investigation over many years, we are not yet in a position to make definitive statements concerning the function of these sequences at the molecular level. Nevertheless, there has been no shortage of informed speculation about their role. Two early models that attributed radically different roles to leader sequences, the signal hypothesis (3) and the membrane trigger hypothesis (4),

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