Dendritic Sidebranching Initiation by a Localized Heat Pulse

X. W. Qian and H. Z. Cummins
Department of Physics, City College of the City University of New York, New York, New York 10031

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The response of a growing crystalline dendrite to a brief localized heat pulse applied near its tip was investigated in a directional solidification experiment on succinonitrile containing 1% acetone. The induced deformation was initially unobservably small, but grew rapidly into a sidebranchlike feature with time. This observation supports the proposed selective amplification of noise mechanism for sidebranching. A preliminary fit to our data of the Gaussian wave-packet description of Pieters and Langer and of Barber, Barbieri, and Langer indicates exponential growth of the perturbation with the $\frac{1}{4}$ power of distance.

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Pattern formation in nonequilibrium dynamical systems has been studied extensively in recent years.[1-5] Dramatic progress has been achieved in understanding how specific dendritic patterns are selected in the solidification of pure materials or alloys. The now widely accepted microscopic solvability theory shows that the selection of a unique velocity and radius of the parabolic dendrite tip (the restabilization process following the initial Mullins-Sekerka and coarsening stages) for a given material under given experimental conditions is primarily controlled by the surface tension and requires anisotropy in either the surface tension itself or in the kinetic coefficient. This theory, and experimental tests of its predictions, are currently the central topic of research in this field.

An associated aspect of dendritic solidification which is not directly addressed by microscopic solvability theory is sidebranching, the appearance of small secondary dendrites which appear several tip radii behind the advancing primary dendrite tip and continue to grow and coarsen as the primary tip moves away. Two quite different explanations for this phenomenon have been proposed. In one, the stable attractor for the dendritic growth process is assumed to be a limit cycle, so that periodic oscillations at the tip generate a coherent train of sidebranches.[6] Alternatively, the attractor has been considered as a stable fixed point with no oscillations, and the sidebranching is then regarded as a stochastic phenomenon resulting from the selective amplification of random noise near the dendritic tip.[7-12].

Various aspects of dendritic sidebranching have been studied previously.[13] A recent crucial study by Dougherty and co-workers[14] of sidebranching in an NH_4Br crystal growing from a supersaturated aqueous solution revealed that (a) the sidebranches are only roughly periodic, (b) there is little correlation between sidebranches on opposite sides of the dendrite, and (c) the sidebranch amplitude increases exponentially with distance from the tip with no apparent onset threshold distance. These observations indicated that the limit-cycle explanation is unlikely, and strongly favor explanations based on the amplification of noise-induced interfacial perturbations. We note that a related noise amplification mechanism has been found to occur at the onset of the Mullins-Sekerka instability at a flat crystal-melt interface.[15] Some aspects of the effects of noise on the stability of Saffman-Taylor fingers were investigated by Tabeling, Zocchi, and Libchaber.[16]

Pieters and Langer and their co-workers have investigated the noise amplification mechanism for sidebranching theoretically using the local boundary-layer model[7,11] (BLM) and a method of analysis related to that employed by Zel'dovich *et al.* in their work on flame fronts.[17] Numerical simulation with the BLM showed that when a small bump is applied to a smooth dendrite tip, the tip is locally stable in the sense that the disturbance at the tip dies out, but the resulting perturbation grows and spreads out as it travels along the smooth side of the dendrite, eventually producing a sidebranch. In addition to these numerical simulations, Pieters and Langer also derived an analytic expression with the form of a Gaussian wave packet for the curvature modification produced by the amplified perturbation, valid asymptotically far from the tip where the intrinsic curvature of the unperturbed dendrite approaches zero. Subsequent calculations by Kessler and Levine,[8] Langer,[9] and Barber, Barbieri, and Langer[10] using more realistic nonlocal models revealed the same qualitative features of selective amplification of noise resulting in sidebranches. Barber, Barbieri, and Langer[10] found that with the two-dimensional symmetric model, a small localized perturbation produces an exponentially growing wave packet identical to the result found with the boundary-layer model.[7]

A direct experimental test of these predictions is hindered by the natural continuous generation of sidebranches which makes it impossible to follow the evolution of a single sidebranch propagating down an otherwise smooth dendrite. We have therefore carried out an experiment in which an externally controlled localized thermal perturbation was applied briefly near the tip, initiating a single sidebranchlike feature which evolved rapidly enough to be observed and analyzed before the spontaneous noise-induced sidebranches grew sufficiently to compete with it.

In our experiment, a 20-cm-long rectangular glass capillary tube (cross section $50\times 1000\ \mu m^2$) was filled in a vacuum-loading apparatus with succinonitrile containing 1% acetone by weight and then sealed. It was then placed in a paraffin-oil-filled-cell assembly attached to a linear motor drive which can move the capillary at constant speed in the range of 0–80 μm/sec, and mounted on a temperature-gradient stage based on the design of Hunt, Jackson, and Brown.[18] The stage was attached to a Nikon Diaphot inverted microscope equipped with both a 35-mm film camera and a video camera interfaced to a digital image-processing board (Data Translation "Quickcapture") in a Macintosh II computer. The drive was typically operated at a growth speed of 0.5 μm/sec, with a temperature gradient of 20 K/cm, resulting (after sufficient time to restabilize in the steady state) in a single dendrite with tip radius $\sim 30\ \mu$m.

The local thermal perturbation was produced by a brief ($\sim\frac{1}{4}$-sec) pulse of 488-nm argon laser light transmitted through an electronic shutter and a single-mode optical fiber to a focusing lens mounted on the stage which produced a spot $\sim 4\ \mu$m in diameter near the tip. Note that this spot is much smaller than the tip radius. The sample was prepared from commercially available succinonitrile (Fluka Industries) purified by multiple vacuum distillation, at least 4 times. 1% by weight of acetone was then added to the purified succinonitrile along with a trace of rhodamine-6G dye to enhance the optical absorption since the succinonitrile-acetone mixture is transparent at 488 nm.

A typical experimental run is illustrated by the sequence of photographs in Fig. 1. The laser pulse was applied close to the tip at $t=0$, corresponding to Fig. 1(a). Initially, there is no visible deformation of the smooth parabolic tip, although the local temperature field has been perturbed. At later times the interface perturbation grows in amplitude and propatates along the dendrite as seen in Figs. 1(b)–1(h). Note that in order to separate the effects of the applied perturbation from the spontaneous noise-generated sidebranches, it is important to start with a stable dendrite having as large a smooth range around the tip as possible. In our experiment, no deformations were initially visible within approximately five tip radii of the dendrite tip. In Fig. 2, we show a sequence of four digitized video images captured by the image-analysis electronics and subsequently transferred to a Vax computer for analysis and plotting. These four images correspond to Figs. 1(b)–1(e).

One possible approach to analyzing our data would be to repeat the numerical BLM simulations of Pieters and Langer[7,11] or, more realistically, to numerically evaluate the integral expressions found with the symmetric model by Barber, Barbieri, and Langer.[10] As a first and simpler approximation, however, we have computed interface profiles from the asymptotic analytic expression of Pieters and Langer[7,11] and compared the results with our data. This analysis is approximate since it cannot be carried out sufficiently far from the tip to be in the correct asymptotic region of validity.

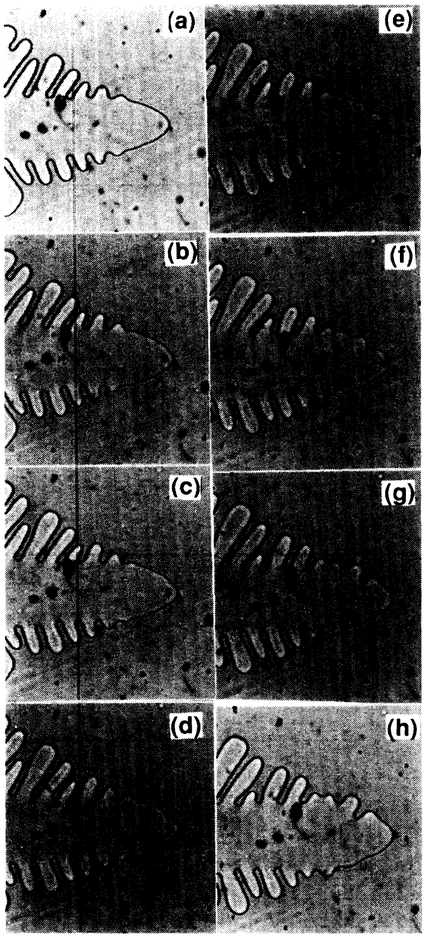

FIG. 1. Microphotographs of a succinonitrile dendrite undergoing directional solidification in a 1% (by weight) acetone solution containing a trace of rhodamine-6G dye. The growth velocity is $V=0.5\ \mu$m/sec and the initial tip radius is $\sim 30\ \mu$m. The time since the application of a $\frac{1}{4}$-sec laser pulse near the tip is (a) $t=0$, (b) 20, (c) 32, (d) 70, (e) 110, (f) 143, (g) 210, and (h) 244 sec.

Pieters and Langer[7,11] analyzed the total interface curvature $\kappa=\kappa_1+\kappa_0$, where κ_0 is the curvature of the unperturbed dendrite. They found that far from the tip (where $\kappa_0\rightarrow 0$) the perturbation-induced modification of the curvature κ_1 could be represented as $\kappa_1 \propto \exp[\Psi(s,s_0)]$, where

$$\Psi(s,s_0)=as^{1/4}\pm(ib/s^{1/4})(s-s_0)-(c/s^{3/4})(s-s_0)^2. \qquad (1)$$

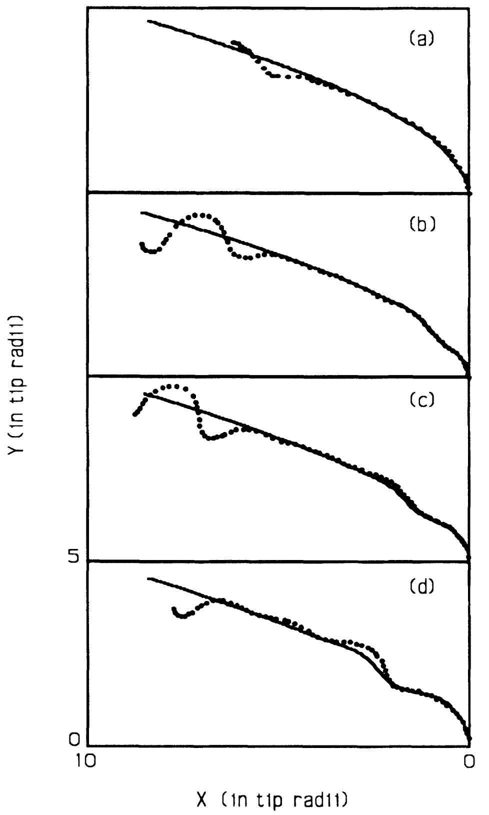

FIG. 2. Dotted lines: Digitized video images of the dendrite of Fig. 1. Images (a)-(d) correspond to the photographs in Figs. 1(b)-1(e). Solid lines: The theoretical predictions of Eq. (1) optimized as described in the text.

In Eq. (1), s is distance along the interface from the tip measured in units of the tip radius, and s_0 is the position at which the perturbation was applied, measured in the reference frame moving with the tip. The coefficients a, b, and c depend on material properties and on the undercooling. Equation (1) describes a Gaussian wave packet whose width and wavelength increase as it grows and propagates down the dendrite. The maximum amplitude of κ_1, at $s = s_0$, is

$$\kappa_1^{\max} \propto \exp(a s_0^{1/4}) , \qquad (2)$$

where $a = 0.647(\sigma^*)^{-1/2}$ and $\sigma^* = 2\tilde{d}_0 D / v\rho^2$. The capillary length $\tilde{d}_0$ is 0.23 μm for 1% succinonitrile-acetone,[1,19] and D, the diffusion constant of acetone in succinonitrile,[19] is 1.27×10^{-5} cm^{2}sec^{-1}. v and ρ are the velocity and radius, respectively, of the steady-state dendrite tip.

In order to compare the predictions of Eq. (1) with our experimental data, we began with the theoretical expression for $\kappa = \kappa_0 + \kappa_1$, treating the coefficients in κ_1 as adjustable parameters, and integrated numerically to obtain the predicted interface shape. The full expression of Eq. (1) did not produce reasonable fits because the experimentally measured packet width remains nearly constant within the range of our measurements rather than increasing with $s_0^{3/8}$ as predicted. We therefore tried two modifications to the full fit. First, we fitted the interface in Fig. 1(d) by adjusting all the parameters to optimize the agreement. All other profiles were fitted with the wavelength constrained as $\lambda \propto s_0^{1/4}$ [as predicted by Eq. (1)] but with the amplitude, width, and s_0 free. The results are shown by the solid lines in Fig. 2 which correspond to the interfaces of Figs. 1(b)-1(e). These fits are reasonably good, but the dependence of the parameters on s_0 was not systematic.

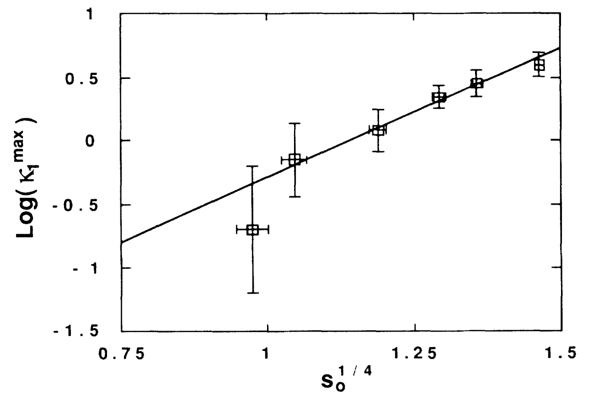

FIG. 3. $\ln \kappa_1^{\max}$ vs $s_0^{1/4}$ from fits to the experimental data with the packet width and wavelength kept constant. The solid line has a slope of $a = 2.0 \pm 0.7$.

We therefore carried out a second fit in which both the packet width and the wavelength were held constant, determined by the fit to Fig. 1(d). For the other interface profiles, only s_0 and $\kappa_1^{\max}$ were varied to optimize the fit to each. These fits were similar to those shown in Fig. 2 for early times [Figs. 1(b)-1(d)] but somewhat worse for later times. In Fig. 3, we show the results for $\kappa_1^{\max}$ vs s_0 obtained from this second fitting procedure. The prediction of Eq. (2) that $\kappa_1^{\max} \propto \exp(a s_0^{1/4})$ is obeyed within the error bars, with $a = 2.0 \pm 0.7$. The prediction of Eq. (2) for our experiment is $a = 0.6$. We similarly plotted the logarithm of the maximum amplitude (rather than curvature) of the perturbation against $s_0^{1/4}$, which is predicted to be a straight line by Eq. (5.6) of Barber, Barbieri, and Langer.[10] The result was essentially identical to Fig. 3.

This analysis is very preliminary since the predictions of Eq. (1) are only expected to become quantitatively accurate farther down the dendrite, in a region where the spontaneous sidebranches interfere with the perturbation-induced feature. Furthermore, the material constants used to calculate the coefficient a may be somewhat modified by the small amount of rhodamine dye we have added. Given these limitations, the results of the analysis are encouraging.

Further experiments are in progress in which periodic heat pulses will be applied at the dendrite tip which should produce coherent trains of sidebranches.[20] The results of these experiments and of an analysis using computer simulations of the BLM will be reported in a future publication.

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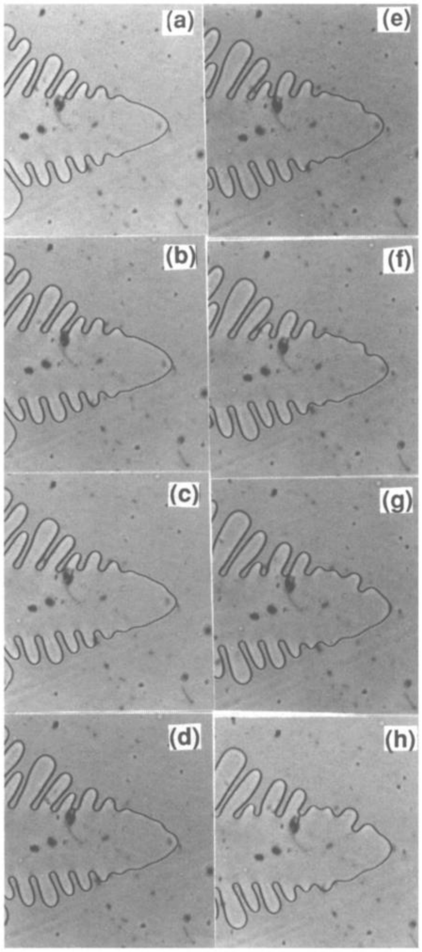

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