

## Direct Measurement of Dendritic Array Stability

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Stability of a uniformly spaced dendritic array against a spatial period doubling instability was studied through UV thermal perturbations of every other dendrite tip. We observed that above a critical pulling speed the array is stable against these perturbations and we measured decreasing decay rates of the perturbed mode as we approached the critical speed. In the linear regime, our measurements are qualitatively consistent with the Warren-Langer linear stability analysis for a dendritic array [J. A. Warren and J. S. Langer, *Phys. Rev. A* **42**, 3518 (1990); *Phys. Rev. E* **47**, 2702 (1993)], while in the nonlinear regime fitting to a third order amplitude equation shows that the transition is subcritical. [S0031-9007(98)06563-6]

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In directional solidification experiments a dilute binary alloy with solute concentration  $C_\infty$  is put in a temperature gradient  $G$  and a flat solid-liquid interface is established. The sample is then pulled with pulling speed  $V_P$  toward the cold side; i.e., the alloy is solidified. Above a critical pulling speed the flat interface becomes cellular and at higher speeds a dendritic pattern is established. The dendrite coarsening process, sidebranching, and the interdendritic spacing  $\lambda_1$  are critical problems in pattern formation, and much work has been devoted to studying the time evolution, the structure, and the stability of the observed patterns.

Detailed studies by Trivedi and co-workers, who started crystal growth from a planar front with constant pulling speed  $V_P$ , indicated that in steady state the interdendritic spacing  $\lambda_1$  is reproducibly determined by the growth conditions and changes smoothly with pulling speed  $V_P$  [1] or solute concentration  $C_\infty$  [2]. Such observations suggested the existence of a dynamical selection mechanism for  $\lambda_1$ . Warren and Langer (WL) carried out a linear stability analysis of an array of dendrites [3,4] using microscopic solvability theory [5,6] to describe individual tip shapes and calculating the interaction between tips through the solute diffusion field. WL found that no dynamical selection mechanism should exist, but for a given  $V_P$  a range of interdendritic spacings is allowed. Their calculations predict that for given growth conditions there is a range of pulling speeds in which an array of spacing  $\lambda_1$  is stable with a lower limit value  $V_{PD}(\lambda_1)$ . Below  $V_{PD}(\lambda_1)$  the array is unstable and a period-doubling bifurcation should be observed, with every other dendrite continuing to grow.

Subsequently, Huang *et al.* [7] observed that if directional solidification is initiated at some  $V_P'$  until a steady state array develops and the pulling speed is then changed from  $V_P'$  to  $V_P$ , different values of  $\lambda_1$  are found for the same final  $V_P$ , depending on the choice of  $V_P'$ . They found that, for a given  $V_P$ , there is a range of  $\lambda_1$  values whose lower limit lies close to the WL prediction. In a more re-

cent experiment Han and Trivedi [8] observed that once a dendritic array has been established,  $V_P$  can be increased considerably without significantly changing  $\lambda_1$ . Direct observation of a period-doubling transition was reported by Losert *et al.* [9]. Starting with a stable array, the pulling speed was decreased in a sequence of small steps until the bifurcation was observed. Similar to measurements of planar interface stability described in [10,11], instabilities develop slowly close to the stability limit and may not be detected by this kind of measurement, since the instability takes a long time to become observable. The pulling speed  $V_P$  at which a period-doubling instability is observed can therefore set only a lower limit for  $V_{PD}(\lambda_1)$ , where the array of spacing  $\lambda_1$  becomes morphologically unstable for a period-doubling instability.

In this Letter we present direct measurements of the stability of a uniformly spaced dendritic array. For this measurement we perturbed a steady state dendritic array with a spatially periodic modulation with a period of  $2\lambda_1$  and observed the decay of this mode as a function of time. As we approach a critical pulling speed from above we observe a slowing of the decay rate in qualitative agreement with the WL theory.

Our sample consists of a ( $100 \mu\text{m} \times 2 \text{ mm} \times 15 \text{ cm}$ ) capillary filled with succinonitrile (SCN) doped with the laser dye coumarin 152 (C152) at  $C_\infty = 0.43 \text{ wt } \%$ . C152 has fluorescence absorption in the UV range so illuminating the sample along the solid-liquid interface with a Hg UV lamp heats the sample locally and visibly melts back the interface. Irradiation in a pure sample causes no observable perturbation, demonstrating that C152 is responsible for the temperature change. For a detailed description of our apparatus see [10]. In order to obtain spatially periodic modulations of the interface, a uniformly spaced row of UV spots was applied along the interface.

The experiments were performed at a fixed temperature gradient  $G = 11.7 \text{ K/cm}$  in a computerized videomicroscopy apparatus mounted on an inverted Nikon

microscope. All experiments were started at the same pulling speed  $V_P = 19.8 \mu\text{m/s}$ . In order to obtain a uniform interdendritic spacing  $\lambda_1$  we apply a row of UV spots with the same spacing  $\lambda_1$  continuously to the interface starting from the time when the motor is switched on. This perturbation influences the coarsening process and improves the uniformity of the interdendritic spacing, as the dendrite tips align themselves in the middle between UV spots. After the spacing has become sufficiently uniform, the UV light is switched off and we allow the dendritic array, growing in the  $z$  direction, to reach steady state (i.e., neither the radius  $\rho$  nor the  $z$  position of the dendrite tips shifts).

UV spots are then focused onto every other dendrite tip producing a small amplitude periodic modulation of the array, whose wavelength  $\lambda$  is twice the array spacing. Images of the dendritic pattern are recorded on videotape for later digital image analysis. Figure 1 shows a sequence of images of the dendritic array just before, during, and after the UV perturbation (visible as white spots at 1:07:00) is applied.

In the linear approximation, the modulation amplitude decays exponentially with a decay rate  $a_0(\lambda)$ , which can be compared to the rate calculated in the WL linear stability analysis for a dendritic array. Superimposed on this modulation with  $\lambda = 2\lambda_1$  is a uniform meltback of all dendrites ( $\lambda = \infty$ ) caused by fast diffusion of the absorbed heat during UV illumination. In first approximation we assume that the two perturbation modes with  $\lambda = 2\lambda_1$  and  $\lambda = \infty$  are independent which yields results very consistent with the weakly nonlinear analysis.

Figure 2 shows a plot of the modulation amplitude, i.e., the difference between perturbed and unperturbed dendrite tip positions  $\xi_{2\lambda_1}(t)$ , after a 20 s perturbation was applied to every other tip at  $V_P = 9.86 \mu\text{m/s}$ . The

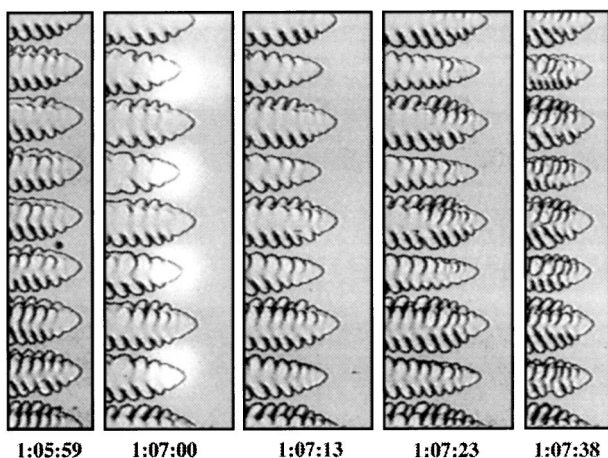


FIG. 1. Image sequence [time in (hour:min:sec)]: Initiation and decay of a spatially periodic modulation of the tip positions in a uniformly spaced dendritic array. Modulation created by a one minute UV-thermal perturbation applied at 1:06:00. (SCN/C152:  $V_P = 8.2 \mu\text{m/s}$ ,  $C_\infty = 0.43 \text{ wt\%}$ ,  $G = 11.7 \text{ K/cm}$ .)

spacing between dendrites in this experiment was  $\lambda_1 \approx 150 \mu\text{m}$ . The modulation amplitude decays exponentially with a negative linear growth coefficient of  $a_0(\lambda = 2\lambda_1) = -0.072 \text{ s}^{-1}$ .

The negative linear growth coefficient of a period-doubling modulation for the same dendrite spacing  $\lambda_1 \approx 150 \mu\text{m}$  was obtained at many different  $V_P$  values between 19.8 and  $6.6 \mu\text{m/s}$ . Figure 3 shows the measured linear growth coefficient as a function of pulling speed. From this plot we can estimate that the stability limit against period doubling [where  $a_0(\lambda = 2\lambda_1) = 0$ ] is reached at  $V_P \approx 4 \mu\text{m/s}$ . An experiment with no perturbation showed that an array with  $\lambda_1 \approx 150 \mu\text{m}$  does indeed become unstable spontaneously at approximately  $V_P \approx 4 \mu\text{m/s}$ . The inset of Fig. 3 shows the WL theory results using the thermodynamic parameters for SCN/C152. Although there is qualitative agreement, the measured decay rates are predicted to occur at pulling speeds about 3 times greater than those used in the experiments. The origin of this quantitative disagreement is a serious concern that should be addressed in future experiments. One possible explanation is that the impurity concentration may have increased from its initial value since the sample was prepared two years ago. Another is that the dendrite axes in this sample were rotated by approximately  $25^\circ$  so that sidebranches of adjacent dendrites do not grow directly towards each other. The influence of the anisotropy of the dendrite shape on the solute diffusion field is not included in the WL analysis, but may lead to increased stability of the dendrite array if sidebranches cannot “collide.”

The growth coefficients for each individual dendrite tip of this uniformly spaced array were also measured for

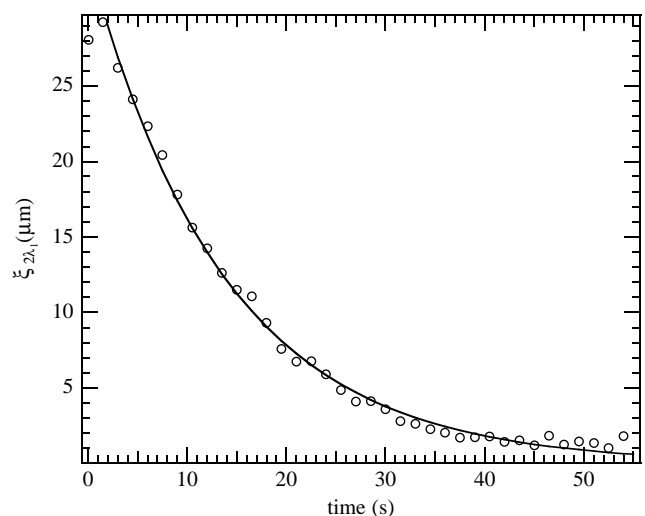


FIG. 2. Average difference in dendrite tip position between perturbed and unperturbed dendrites  $\xi_{2\lambda_1}$  vs time ( $\circ$ ), and exponential fit (—) in the range  $t = 0-52 \text{ s}$  [ $a_0(\lambda = 2\lambda_1) = -0.072 \text{ s}^{-1}$ ]. Modulation produced by a spatially periodic UV thermal perturbation with  $\lambda \approx 300 \mu\text{m}$  applied for 20 s, which melted back every other dendrite tip. (SCN/C152:  $V_P = 9.86 \mu\text{m/s}$ ,  $C_\infty = 0.43 \text{ wt\%}$ ,  $G = 11.7 \text{ K/cm}$ .)

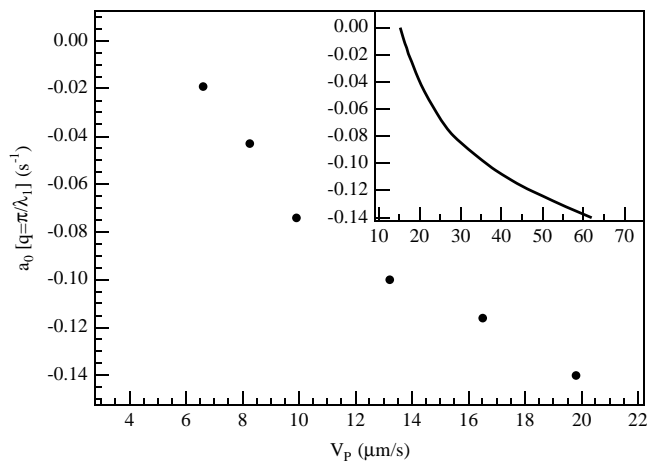


FIG. 3. Linear growth coefficient vs  $V_P$  for an interdendritic spacing of  $\lambda_1 \approx 150 \mu\text{m}$ .  $a_0(\lambda = 2\lambda_1)$  measured as in Fig. 2. Inset: Warren-Langer theory results using the thermodynamic parameters for SCN/C152.

some data points. They were found to be approximately the same, within the uncertainty of the individual fits.

Near the stability limit the linear growth coefficient becomes small and nonlinear terms should be included in the analysis through the third order Landau equation:

$$\frac{d\xi_\lambda(t)}{dt} = a_0(\lambda)\xi_\lambda(t) + a_1(\lambda)|\xi_\lambda(t)|^2\xi_\lambda(t). \quad (1)$$

The solution to Eq. (1) is

$$\xi_\lambda(t) = \left[ \sqrt{\left(\frac{a_1(\lambda)}{a_0(\lambda)} + \xi_\lambda(0)^{-2}\right)e^{-2a_0(\lambda)t} - \frac{a_1(\lambda)}{a_0(\lambda)}} \right]^{-1}. \quad (2)$$

At this speed close to linear marginal stability, small changes in individual spacing between dendrites become important, as they slightly shift the marginal stability point. A quantitative analysis of nonlinear effects was carried out at  $V_P = 9.86 \mu\text{m/s}$  on three individual dendrite tips in a modulated array whose nearest neighbor spacing was  $\lambda_1 = 181 \mu\text{m}$  and deviated from one another by less than 1%. Figure 4 shows  $\xi_\lambda(t)$  of each of those three dendrite tips. Because of nonuniformity of the perturbation in this experiment, the initial values of  $\xi_\lambda(t)$  differed by a factor of about 3. When the curves were offset on the time axis so that the initial amplitudes of the smaller amplitude tips lie on the relaxation curve of the largest amplitude tip, the relaxation curves were found to overlap, independent of the initial amplitude. Exponential fits (---) for amplitude  $\xi_\lambda(t) \leq 20 \mu\text{m}$  gave a relaxation rate of  $a_0 = -0.091 \text{ s}^{-1}$ . The full third order function [Eq. (2)] (—) can be fit well over the full range of amplitudes and yields  $a_0 = -0.090 \text{ s}^{-1}$  and  $a_1 = 1.1 \times 10^{-5} \mu\text{m}^{-2} \text{ s}^{-1}$ . The sign of  $a_1$  determines if the bifurcation is subcritical or supercritical, a point not addressed in the linear analysis of WL.

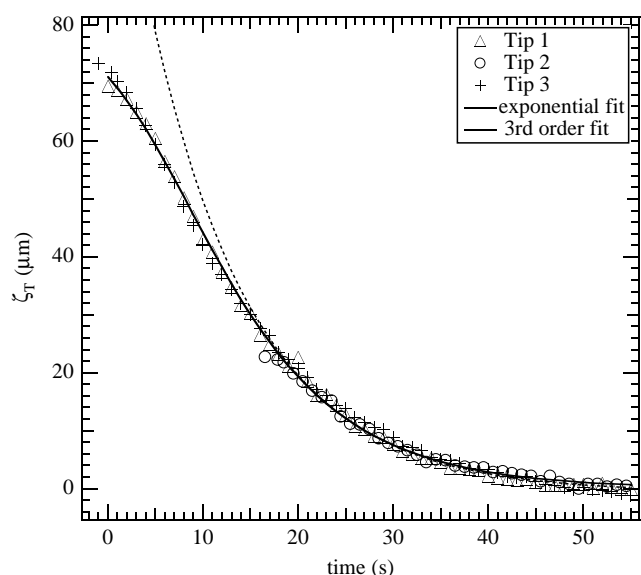


FIG. 4. Nonlinear fitting of the modulation amplitude  $\xi_\lambda$  of three dendrite tips relative to their neighbors ( $\lambda_1 = 181 \mu\text{m}$ ): Exponential fits (---) for amplitude  $\xi_\lambda(t) \leq 20 \mu\text{m}$  ( $a_0 = -0.091 \text{ s}^{-1}$ ) and fits to the full third order function (—) ( $a_0 = -0.090 \text{ s}^{-1}$  and  $a_1 = 1.1 \times 10^{-5} \mu\text{m}^{-2} \text{ s}^{-1}$ ). (SCN/C152:  $C_\infty = 0.43 \text{ wt}\%$ ,  $G = 11.7 \text{ K/cm}$ ,  $V_P = 9.86 \mu\text{m/s}$ .)

Since  $a_1$  is positive, Eq. (1) leads to a subcritical bifurcation. If the initial modulation amplitude  $\xi_\lambda(0)$  is large enough, the instability can occur while the pattern is linearly stable, i.e.,  $d\xi_\lambda/dt > 0$  even though  $a_0 < 0$ . The critical initial amplitude for this effect is [from Eq. (1)] given by  $\xi_C = \sqrt{-a_0/a_1}$ . From the measured values for  $a_0$  and  $a_1$  at  $V_P = 9.86 \mu\text{m/s}$  we obtain  $\xi_C \approx 100 \mu\text{m}$ . When a larger amplitude modulation  $\xi_\lambda(0) \sim 130 \mu\text{m}$  was created at the same pulling speed, we indeed observed that the modulation did not decay but led to a period-doubling instability.

In conclusion, the stability of a uniformly spaced dendritic array against period-doubling perturbations was studied. The relaxation rate of the modulation was found to decrease with decreasing pulling speed in both the linear and the nonlinear regimes. Measured decay rates are in qualitative agreement with the WL theory (although there is quantitative disagreement of a factor of 3). The third order Landau equation fit to the observed decay for large amplitude perturbations indicates that period doubling in a dendritic array close to the cellular regime is subcritical. The subcritical range is large, since the forced transition was observed at more than twice the speed of a spontaneous transition where  $a_0$  reaches zero. Our measurements also indicate that the decay of modulation amplitudes that reach up to 50% of the dendrite spacing can be described well by a linear approximation far from the critical speed. A third order analysis is necessary only for large amplitudes close to the critical speed.

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