LETTER TO THE EDITOR

Role of fluctuations in viscous fingering and dendritic crystal growth: a noise-driven model with non-periodic sidebranching and no threshold for onset

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Abstract. A noise-driven model is developed to describe the role of fluctuations in side-branch phenomena in growth patterns for the fluid displacement problem and for dendritic crystal growth. Simulation results are compared with recent experiments on NH_4Br dendrites. It is found that the RMS sidebranch amplitude is an exponential function of distance from the tip, with no apparent onset threshold. Moreover, the sidebranches are non-periodic (at *all* distances from the tip) with apparently random variations in amplitude.

What is the physical mechanism whereby sidebranches 'spontaneously' appear a short distance behind the growing tip of a dendritic form? For generations, this question has fascinated scientists in a variety of fields, ranging from metallurgy and crystal growth on the one hand to botany and embryology on the other. Recently interest has focused on extremely simple systems that spontaneously develop sidebranches. It has been found that, when a low-viscosity fluid displaces a high-viscosity *anisotropic* fluid under pressure, a sidebranch pattern develops that resembles dendritic crystal growth. For example, Buka *et al* [1] use air to displace a viscous solution of a nematic liquid crystal. The anisotropy can also be in the medium itself. Horvath *et al* [2] have shown that a single scratch in one wall of the confining Hele–Shaw cell is sufficient to produce a dendritic pattern. Similarly, Ben-Jacob *et al* [3] find dendritic fluid patterns when they scratch a triangular lattice onto the cell. Most surprising, perhaps, is the observation of Couder *et al* [4] that dendritic growth patterns can occur when the anisotropy is provided by a simple bubble of air on the tip of the growing viscous finger. Can these similarities between diverse systems be understood in terms of underlying physical principles common to all? Here we tentatively suggest a physical model that seems to account for such sidebranch phenomena.

Although fluid displacement phenomena are striking, a larger number of *quantitative* results is known for dendritic crystal growth. Hence we shall focus attention on the latter. In particular, Dougherty *et al* [5] have recently made a detailed analysis of photographs of growing NH_4Br dendrites, taken at 20 s intervals. They have found three surprising results: (i) sidebranch positions are non-periodic at any distance from the tip, with almost random variations in both phase and amplitude, (ii) sidebranches on opposite sides of the dendrite are essentially uncorrelated in position and length and (iii) the sidebranch amplitude is an exponential function of distance from the tip, *with no apparent onset threshold distance.*

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How can we understand these new experimental facts? Although recent theoretical work has been remarkably successful in explaining many aspects of dendrite growth [6, 7], an explanation of random sidebranching phenomena has been elusive in part, perhaps, because of the difficulty of incorporating in these theories the subtle effects of fluctuations ('noise'). Indeed, some of the best theories [8-11] predict sidebranching that is symmetric and almost perfectly periodic. The model we develop here is essentially the complement of these recent theories and is based entirely on fluctuation phenomena; the resulting patterns can be said to be 'noise-driven'.

Growth of a dendrite from solution is controlled by the random diffusion of solute toward the growing dendrite. In the limit of small Peclet number, the diffusion equation reduces to the Laplace equation. The Laplace equation for a system with an interface not constant with time (the growing dendrite) brings to mind the noise-dominated diffusion-limited aggregation model (DLA) [12]. However, dendrites do *not* resemble DLA patterns which are much too chaotic in appearance, with no obvious anisotropy†.

We discuss here a related model whose asymptotic structure does resemble the patterns found experimentally. Our starting point is the observation that even the tiniest amount of anisotropy becomes magnified as the mass M of a cluster increases [12]. For example, the weak anisotropy of the underlying lattice structure can become so amplified that clusters of 4×10^6 particles grown on a square lattice take on the appearance of a Swiss cross (cf figure 15 of [12]). A real dendrite has a mass of roughly 10^{16} particles; one cannot generate such clusters on a computer, but there is a computational trick—termed *noise reduction*—that speeds the convergence of the pattern toward its asymptotic 'infinite mass' limit [13-16]. We choose boundary conditions (figure 1) corresponding to those of the experimental setup: a long rectangular $L \times L'$ cell oriented parallel to the x axis of an xy coordinate system ($0 < x < L$ and $0 < y < L'$, with $L \gg L'$). The chemical potential $\phi(x, y)$ has the values $\phi(x, y = L') = \phi(x, y = 0) = 1 - x/L$, $\phi(0, y) = 1$, $\phi(L, y) = 0$, and $\phi(x, y) = 1$ for all points (x, y) belonging to the cluster. At time zero, a single seed particle is placed at ($x = 0$, $y = L'/2$), and the Laplace equation $\nabla^2 \phi(x, y) = 0$ is solved numerically with the boundary
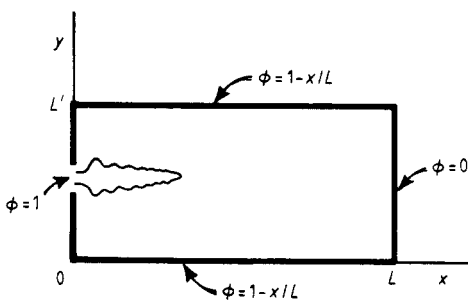

Figure 1. Geometry and boundary conditions of the present model.

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† A critical binary mixture exhibits large concentration fluctuations. Near the growing dendrite there are also large concentration fluctuations (roughly $\pm 10^5$ NH$_4$Br molecule μm^{-3}). Critical mixtures are described by the Ising model (or ϕ^4 field theory) which exhibits fluctuations on all length scales from the microscopic scale of the lattice constant a_0 up to the macroscopic scale of the correlation length ξ; $\xi \to \infty$ as $\varepsilon \equiv (T - T_c)/T_c \to 0$. $\xi_{\text{Ising}} \sim \xi_0 \varepsilon^{-\nu_{\text{Ising}}}$ where $\xi_0 (\approx a_0)$ is a critical amplitude and $\nu_{\text{Ising}} (\approx 0.6)$ is a critical exponent depending only on the system dimension d. Anisotropic DLA has fluctuations (e.g. holes) on all sizes from a_0 to the scale of a 'finger width' ξ_{DLA}; $\xi_{\text{DLA}} \to \infty$ as $\varepsilon_{\text{DLA}} \equiv 1/M \to 0$. $\xi_{\text{DLA}} \sim \xi_0 \varepsilon^{-\nu_{\text{DLA}}}$ where $\xi_0 (\approx a_0)$ and the inverse fractal dimension of DLA, $\nu_{\text{DLA}} (\approx 0.6)$, depends on d.

conditions just given. Each of the N perimeter sites i is assigned a normalised growth probability p_i which is proportional to $\nabla\phi$. The unit interval is partitioned into N subintervals, each of length p_i, which are in one-to-one correspondence with the perimeter sites i. A random number is drawn from the interval $[0, 1]$ and identified with the 'choice' of a perimeter site at which growth could next occur. However, instead of allowing growth to occur, a counter associated with that perimeter site is incremented by unity. A second random number is drawn and the process repeated until one of the N counters reaches a preset threshold value s. The special case $s = 1$ reduces to the conventional DLA model†. The fractal dimension d_f is independent of s for all *finite* values of s, but the computational power gained through the use of noise reduction is enormous. The anisotropic patterns found for huge DLA clusters [12] are reproduced in much less computer time for clusters of smaller size [14–16]. Similarly for the Eden model, previous $s = 1$ results achieved using immense computer effort [17] have been reproduced and improved with much smaller systems using noise reduction $(s > 1)$ [18].

The patterns we obtained (figure 2) with noise-reduced DLA resemble the experiments of [5]. The details of the patterns we obtain can be 'fine tuned' if we introduce slight variations of the original DLA model, as follows.
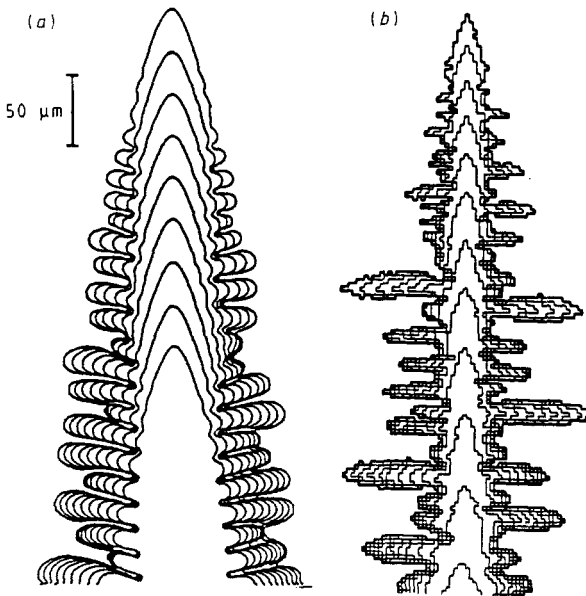

Figure 2. Comparison between (a) experimental dendrite of [5] and (b) typical simulation pattern with $\eta = 1$, $\sigma = 0.01$, $s = 200$, $M = 4000$ and boundary conditions that alternate between DLA and DBM $(k = l = 1)$. We can vary the model parameters over a modest range and still obtain patterns that resemble the experiments of [5].

† As s increases, the growth at the next step—*although still random*—corresponds with better accuracy to the 'Darcy law' (linear response) solution of the Laplace equation. For example, suppose the seed of a cluster were a large disc-shaped object. Then for $s = 1$ subsequent growth would resemble a forest of trees growing off the surface of the disc, while for infinite s the disc would grow into a larger disc. For a very large but finite value of s, the disc would initially grow into a larger disc, and then would undergo tip splitting. The fractal dimension d_f would start at $d_f = 2$ but would eventually cross over to that of the forest of trees.

(a) *Boundary conditions on growth.* A DLA cluster grows when a walker steps on any *perimeter* site i (then i becomes a cluster site). For the dielectric breakdown model (DBM) [19], the random walker must actually step on a *cluster* site in order to grow (then the last visited perimeter site becomes a cluster site). If we solve the Laplace equation directly instead of using random walkers, then DBM boundary conditions imply setting the potential $\phi = 1$ *on the cluster sites*, while DLA boundary conditions imply $\phi = 1$ *on the perimeter sites as well.* Which boundary conditions on growth to use is an open question. Indeed, real growth should probably be intermediate between the extremes of DLA and DBM. Accordingly, we explored the consequences of growth for which k sites were added using DLA boundary conditions, followed by l sites using DBM boundary conditions. Good patterns were obtained for many choices, including the simplest case $k = l = 1$.

(b) *Surface tension.* We found that the effect of tuning a surface tension parameter σ is to thicken the sidebranches and to round the sharper points of the pattern, since the potential ϕ_{1F} on the interface is not constant ($\phi = \phi_0$) but changes with the radius of curvature R_c: $\phi_{1F} = \phi_0 - \sigma/R_c$. We calculated R_c using a variation of the Vicsek form [20]. We increase the growth probability for site i in proportion to the number of cluster sites inside a small box centred about site i, e.g. for a tiny 3×3 box centred on each cluster site, we find $1/R_c = (5 - \text{NN})/4$, where NN denotes the number of occupied neighbours of the central cluster site.

A typical result for a mass of 4000 particles is shown in figure 2(b). After each 333 particles are added, a contour is drawn. Our findings are as follows.

(i) It is apparent from figure 2 that the distance between successive tip positions is a decreasing function of the mass; in fact, we find that $\log x_{\text{tip}}$ is linear in $\log M$ with slope $\frac{2}{3}$. This result is consistent with the belief that $d_f \simeq 1.5$ for anisotropic DLA.

(ii) The tip is parabolic: when we plot $(y_c - y_0)^2$ (where y_c is the contour, and y_0 is the centreline of the dendrite) as a function of $x_{\text{tip}} - x$, we obtain a straight line with a correlation coefficient of 0.997.

(iii) The sidebranches are non-periodic at any distance from the tip, with random variations in both phase and amplitude. To demonstrate this, we analysed our simulations in exactly the same fashion as the experimental patterns were analysed. Thus
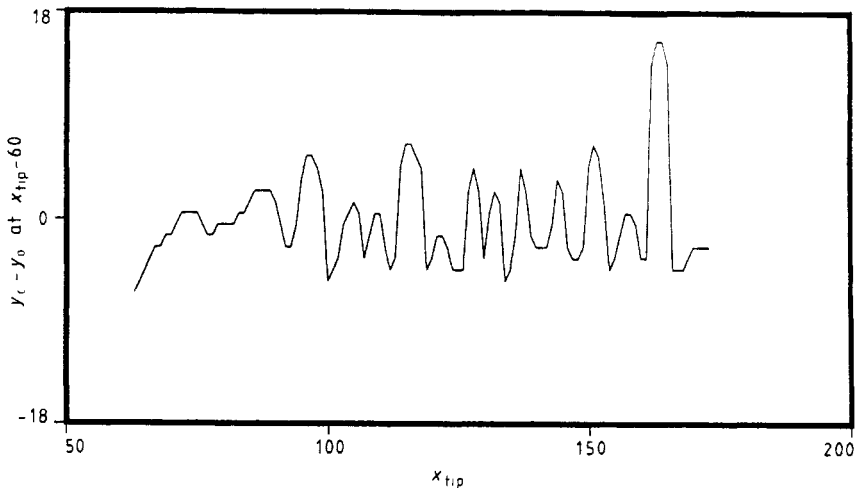

Figure 3. A plot of $y_c - y_0$ as a function of x_{tip} at a fixed distance $z = 60$ from the tip (i.e. at $x = x_{\text{tip}} - 60$) (cf figure 3 of [5]).

we can plot the width y_c of the dendrite at a fixed distance z from x_{tip} as a function of x_{tip}. The sidebranching is not periodic, the amplitude has the same intermittent character as the experimental graph (figure 3) and the power spectrum reveals the presence of many characteristic frequencies.

An open question concerns the microscopic origin of the sidebranch phenomenon. Some models predict that the sidebranch amplitude is periodic and the two sides of the dendrite should have symmetric sidebranching. Rather than supporting this prediction, our simulations (and the experiments of [5]) support the possibility that sidebranches arise from random concentration fluctuations, much as do the 'tree structures' that grow in DLA surface deposition [12]. Indeed, on a microscopic scale the side of the dendrite near the tip resembles an almost flat surface. Growth is small near the tip but increases exponentially with distance from the tip. Specifically, we analysed our simulations exactly as the NH_4Br experiments. As in [5] we found that the 'sidebranch amplitude' (the square root of the area under the broad peak in the noise spectrum—cf our figure 3 and also figure 3(*b*) of [5]) decreases as the distance variable $x_{tip} - x$ decreases, and shows no sign of a threshold distance below which the amplitude is zero! Moreover, close to the tip the sidebranch amplitude appears (figure 4) to approach zero *exponentially* with $x_{tip} - x$, just as found experimentally (figure 4 of [5]).
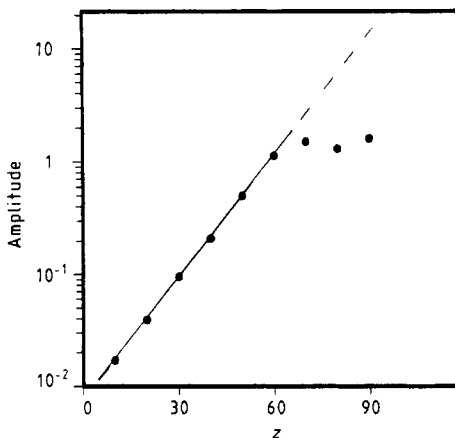

Figure 4. Depencence of the sidebranch amplitude (the square root of the area under the first peak of the power spectrum obtained from Fourier transformation of figure 3) on the distance z from the tip plotted semilogarithmically. The exponential increase is the same as in NH_4Br (cf figure 4 of [5]).

In summary, we have developed a hybrid model (of DLA and DBM boundary conditions) in which noise reduction is used to tune the effect of concentration fluctuations and anisotropy is introduced through the use of an underlying square lattice. The resulting patterns obtained resemble the experimental patterns of [5], both in their qualitative appearance (figure 1) and in the degree of quantitative detail studied experimentally (cf figures 2-4 with figures 1-4 of [5]). Sidebranching arises from the fact that an approximately flat interface in the DLA problem grows 'trees', which resemble 'bumps' in the presence of noise reduction; these compete for the incoming flux of random walkers. If one bump gets ahead, it has a further advantage for attracting the next NH_4Br molecule. Thus some sidebranches get ahead while others do not. The characteristic spacing λ between sidebranches scales with the dendrite mass with

the same exponent $\frac{2}{3}$ that characterises the growth of dendrite length (χ_{tip}). Moreover, the patterns we obtain are reasonably independent of details of the simulation in that similar patterns are obtained when we vary the surface tension parameter σ over a modest range; we can also alter the boundary conditions of the model with some latitude. The significance of the present findings is that the essential physics embodied in the noise-driven DLA model seems sufficient to describe anisotropic fluid displacement phenomena and dendritic growth patterns.

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