Diffusion-limited aggregation and viscous fingering in a wedge: Evidence for a critical angle

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We show that both analytic and numerical evidence points to the existence of a critical angle of $\eta \approx 60° - 70°$ in viscous fingers and diffusion-limited aggregates growing in a wedge. The significance of this angle is that it is the typical angular spread of a major finger. For wedges with an angle larger than 2η, two fingers can coexist. Thus a finger with this angular spread is a kind of building block for viscous fingering patterns and diffusion-limited aggregation clusters in radial geometry. [S1063-651X(98)10506-8]

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The diffusion-limited aggregation (DLA) [1,2] model is a simple idealization of a common natural process, the formation of natural objects where the rate-limiting step is diffusion. In the simplest examples (say, solidification from solution, or diffusion-limited electrochemical deposition), particles random walk and then stick to a growing aggregate. Diffusion-limited growth of this type gives rise to remarkable morphologies which are ramified, disorderly, and, in the case of infinite diffusion length, fractal. It is this complexity which is the major interest in the model. Despite more than a decade of work in this field [2], very little theoretical understanding has been achieved. In this paper we attempt to contribute to such understanding by demonstrating the existence of a kind of building block for the pattern: there seems to be a characteristic angular spread for the fingers which make up the structure.

The fundamental origin of the complexity of DLA patterns has been known from the outset: it is in a *fingering instability*: diffusion-limited growth is generically linearly unstable for flat growing surfaces, and forms fingers. The proliferation of the fingers gives rise to the fractal pattern in a way which we seek to clarify here. Another physical system that displays the fingering instability is the displacement of an inviscid fluid by a viscous one, the viscous fingering problem. It has been suspected since the work of Paterson [3] that the large-scale features of DLA patterns are similar to those in radial viscous fingering. They both obey the *Laplacian growth* equations

$$\nabla^2 \phi = 0, \tag{1}$$

$$\hat{n} \cdot \vec{\nabla} \phi = \hat{n} \cdot \vec{v}. \tag{2}$$

Here ϕ denotes the diffusing field, i.e., the probability density ever to find a random walker at point $\mathbf{r}$ in the case of DLA or the pressure at $\mathbf{r}$ in viscous fingering. The normal velocity of growth of the pattern is $\hat{n} \cdot \vec{v}$. The boundary value on the surface of the growing pattern, ϕ_s, is given by the Gibbs-Thomson relation $\phi_s = \gamma \kappa$ for the case of viscous fingering where γ is the surface tension. DLA differs by having the boundary condition set implicitly by the finite size of the accreting particles, and by the fact that the patterns are affected by shot noise. Some authors have argued [4] that neither of these facts affect the large-scale features of the pattern, and that radial viscous fingering patterns are identical to DLA clusters in a coarse-grained sense. We adopt this point of view.

This idea is attractive because the theory of viscous fingering is quite well developed [5]. In particular, it is clear that viscous fingers in a channel geometry are not fractal [6], and attain a steady state of a single finger. The striking difference from the radial case, where there is no indication that a steady state is ever achieved, led Ben-Amar [7] to investigate the wedge geometry. The general result is that in a wedge of any angle the selected finger grows in a self-similar way. For fixed surface tension they are stable for a finite time, and they then become unstable against tip splitting. This idea was used by Sarkar [8] to give an estimate for the fractal dimension of DLA by counting the tip splittings.

However, we think that Sarkar's estimate left out a crucial effect: that of finger competition. Our view is that this is the key to the whole problem: if fingers split in a wedge that is too narrow, they will compete, and one will die. The result will be a finger with sidebranches. On the other hand, if the wedge is wide enough, then the fingers will not compete, and there will be two branches to the pattern. The wedge angle α at which this begins to happen will be twice the typical angle between fingers, which we call η. As a pattern grows the fingers will split until they form channels of angle η for their neighbors. There is some experimental support for this idea [9] in the mode of tip splitting seen for various angles. However, the experimental evidence is ambiguous because the dynamic range of a real fingering experiment is limited. Here we will try to verify our ideas by giving an analytic estimate for η, and then show that these are reasonable by considering simulations of DLA clusters in a wedge.

To begin, consider two steady-state viscous fingers side by side in a wedge with periodic boundary conditions at the sides. We will attempt to estimate how large the angle must be so that there is no competition between them. For example, for $\alpha = 2\pi$ the fingers grow independently.

We now look at the stability of the two finger solution in order to see if there is competition. We prepare one finger

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slightly longer than the other, and ask, in the linear regime, if there is a different growth rate for the two. We can see how the calculation goes by using the mapping, due to Ben-Amar and Brener [10], between the wedge problem and the problem of diffusive (i.e. not Laplacian) growth in a channel. We first map the wedge to a strip using $\tilde{z}=[2/\alpha]\ln z$. This transforms a wedge of angle α centered around the x axis in the z plane to a strip of width 2 in the $\tilde{z}$ plane. Since the transformation is conformal, the field is Laplacian in the new variables. Equation (2) becomes

$$\hat{\tilde{n}}\cdot\tilde{\nabla}\phi=\exp(\alpha\tilde{x})\hat{\tilde{n}}\cdot\tilde{\vec{v}}. \tag{3}$$

The Gibbs-Thomson condition on the interface is complicated in the new coordinates except for small surface tension, in which case it becomes

$$\phi_s=\gamma\tilde{\kappa}\,\exp(-\alpha\tilde{x}/2). \tag{4}$$

If we define $\Phi\equiv\phi\,\exp(-\alpha\tilde{x})$ then Φ satisfies, up to terms of order α^2, a quasistatic diffusion equation in the frame moving with Peclet number α:

$$\nabla^2\Phi+2\alpha\frac{d\Phi}{dx}=\alpha^2\Phi\approx 0. \tag{5}$$

Equation (3) now reads $\hat{\tilde{n}}\cdot\tilde{\vec{\nabla}}\Phi=\hat{\tilde{n}}\cdot\tilde{\vec{v}}$. Thus we have two steady-state fingers growing in a channel with finite diffusion constant and with boundary condition on the interfaces $\Phi_s=\gamma\tilde{\kappa}\,\exp(-3\alpha\tilde{x}/2)$. This equation implies a space-dependent effective curvature. Thus our problem is not exactly the same as that of dendrites in a channel, but it is qualitatively the same [11]. For the question of competition, the exact form of the surface tension is probably not important. From Eq. (5) it follows that $1/\alpha$ plays the role of a diffusion length: the field is screened over distances larger than $1/\alpha$, and two fingers that are separated by larger distance cannot compete. This is an indication that a critical angle exists.

We have verified this insight by a numerical stability analysis. We found that for small α (weak screening) fingers compete, and for a large wedge angle they do not. The numerical value for the threshold that we compute in this way ($\alpha\approx 0.5$) is too large to be trusted because of the small α approximation.

In order to go further, we do a different estimate which is more qualitative, but not restricted to small angles. Consider, again, two viscous fingers in a wedge of opening angle α. We now replace the problem with a simpler one that we can solve analytically, that of two *needles* in the wedge. In complex notation, the tips of the needles are at $z_1=l_1e^{i\alpha/4}$ and $z_2=l_1e^{3i\alpha/4}$. The Laplacian field ϕ vanishes on the needles, and we suppose that there is a cutoff (finite tip size) a and that the growth rate of the needles is given by the flux of the Laplacian field $\nabla\phi$ at the tips: $z=z_i+ae^{i\theta_i}=z_i+\delta z_i$; $\theta_{1,2}=\alpha/4,3\alpha/4$.

We solve by a series of conformal maps. First we map the z plane into the u plane with $u=z^\beta$ and $\beta=2\pi/\alpha$. The two needles are now one needle along the imaginary axis. Now center the needle. Define $L=[l_1^\beta+l_2^\beta]/2$, and arrange things so that the needle goes from $-L$ to L by putting $w=u-[l_1^\beta-l_2^\beta]/2$. Then we can map the line segment onto the unit circle by putting $w=[Li/2][\tilde{z}+1/\tilde{z}]$. Now the two needles have been mapped onto points on the exterior of the unit circle ($\tilde{z}_1=1$, $\tilde{z}_2=-1$).

It is now clear that the potential can be written $\phi=\mathrm{Re}\,\psi$; $\psi=\phi_o\ln(\tilde{z})$, where ϕ_o is proportional to the incoming flux. This potential satisfies periodic boundary conditions. To obtain the growth rate, it is sufficient to find $d\psi/dz$ because $|d\psi/dz|^2=|\nabla\phi|^2$. By a straightforward computation we can write down the growth rate of tip i:

$$d\psi/dz|_i=|d\Phi/dz|\propto\frac{l_i^{\beta-1}}{[Ll_i^{\beta-1}a]^{1/2}}\propto\frac{l_i^{[\beta-1]/2}}{[l_1^\beta+l_2^\beta]^{1/2}}. \tag{6}$$

Whenever one finger is longer than the other, the longer one will receive more flux, and, it seems, grow faster. However, we know from the computation above that there is a point at which fingers cease competing. Physically this is because the difference between a needle and a finger is that a finger must grow in *area* if it is self-similar. Thus even if the integrated flux to two fingers is the *same*, the fatter one will grow more slowly since it will advance according to $dA_i/dt\propto d(l_i^2)/dt\propto l_idl_i/dt\propto G_i$ where G_i is the flux that finger i receives in competition with the other. We estimate $G_i\propto l_i^{[\beta-1]/2}$ from the needle calculation. That is $dl_i/dt\propto|d\Phi/dz|/l_i$. Thus

$$\frac{dl_1/dt}{dl_2/dt}=[l_i/l_2]^{[\beta-3]/2}. \tag{7}$$

When $\beta=3$, that is, when $\alpha=120°$, the two fingers stop competing. Thus each finger occupies $\eta=60°$. We should note that this is exactly the criterion of Derrida and Hakim [12] who obtained it in a different way, namely, by demanding that, for some fixed a, the *ratio* of the lengths of two spikes remain small (though the difference can be large). That is, for $l_1>l_2$ they made this quantity decrease:

$$\frac{d}{dt}(l_1/l_2)=\frac{l_1}{l_2}[(1/l_1)d\Phi_1/dz-(1/l_2)d\Phi_2/dz], \tag{8}$$

which is our estimate.

We can use this estimate in another way. Suppose that the fingers are fractal, so that we have $A\propto l^D$, where D is the fractal dimension. Now repeating the calculation above, we must have $(\beta-1)/2=D-1$ at the operating point. However, Turkevich and Scher [13] gave another criterion: if the cluster grows so that it has major branches, then the growth and the fractal dimension will be dominated by the tip angle. The result of this consideration, in our notation, amounts to saying that $D=1+\pi/(\pi+\eta)$. Using $\beta=\pi/\eta$ we find that η satisfies a quadratic equation, whose solution is $\eta=(\sqrt{2}-1)\pi$, which corresponds to $\eta\approx 75°$. Thus $D=1+1/\sqrt{2}=1.71$, which is exactly the observed fractal dimension. This estimate was given by Ball [14] some years ago using a different argument.

To verify these estimates we turn to numerical calculations for DLA in a wedge. We grew a large number of off-lattice DLA clusters in wedges of different opening angles α.

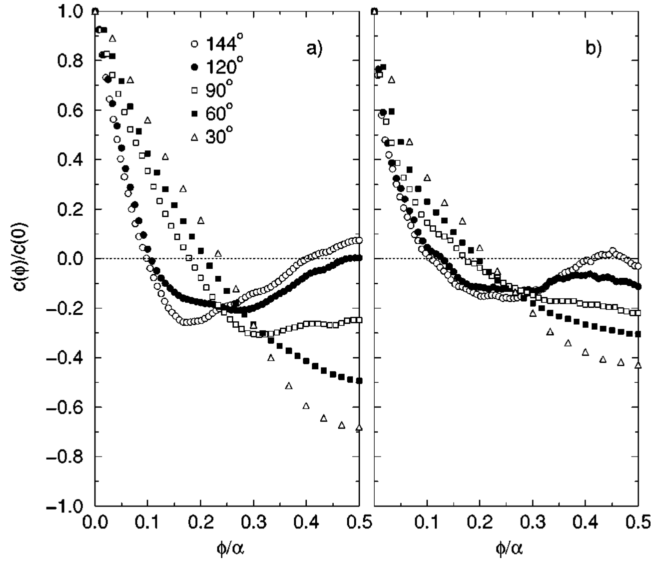

FIG. 1. (a) Angular correlation function $c(\phi)/c(0)$ for DLA clusters in a wedge of angle α as a function of ϕ/α. (b) Correlation functions using the measure M_1.

For greater efficiency we used the method of hierarchical maps [15] adapted to the wedge geometry, so that our wedge was subdivided into sectors whose radii were in geometric ratios. The data which we will report involve averages over 25 realizations for each α, and the number of particles, M, in the wedge was determined so that $M=10^6\alpha/2\pi$. That is, each wedge acted like a slice from a million-particle cluster. We report results for $\alpha=30°$, 60°, 90°, 120°, and 144°.

We measured the fractal dimension of our clusters and find that it depends weakly, if at all, on α. This allows us to understand the remarkable accuracy of the estimate of fractal dimension above. The Turkevich-Scher calculation implies that the fractal dimension of a finger would depend only on the tip velocity, which in turn depends on the tip structure. The invariance of the fractal dimension with α indicates that the tip structure is not affected by boundaries, and thus probably not by the presence of other fingers. However, in the radial case, the large-scale structure (the number of main surviving branches) adjusts via finger competition to be consistent with the local growth rate. In our estimate we gave a representation of the tip which is valid only far away—we replaced the cluster by a needle—but then used self-consistency to find the fractal dimension.

In this work our main interest is not the fractal dimension but the overall shape. To see this, we computed the density-density correlation function for two sectors separated by ϕ:

$$c(\phi)=[\langle\rho(\theta+\phi)\rho(\theta)\rangle-\langle\rho\rangle^2]|_\theta. \qquad (9)$$

Here $\rho(\theta)$ is the density of particles in the cluster in a 1° sector around θ, and we average over the starting angle. All of the angles are taken as periodic with period α, so that the function is reflection-symmetric around $\alpha/2$. In Fig. 1(a), we show the correlation function averaged over 25 realizations, and in Fig. 2 a typical cluster for small and large angles.

There is a very clear difference between large and small α in the behavior of $c(\phi)$. For small angles there is an anticorrelation between the origin and other angles. This corresponds to the matter being clustered in one branch. For α between 90° and 144°, the nature of the correlations changes. The appearance of a second peak and the positive correlation function indicates that there are now two coexisting branches [16].

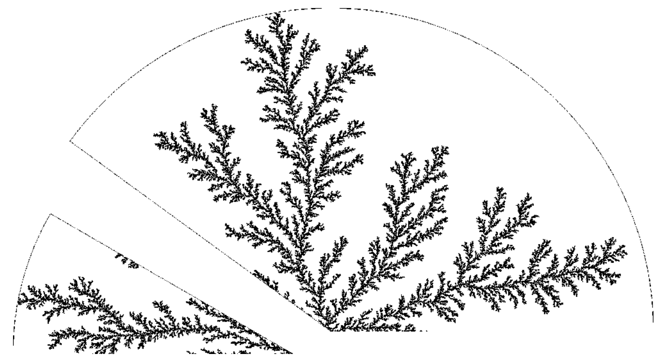

FIG. 2. DLA clusters grown in wedges for $\alpha=30°$ and 144°.

We have examined the individual realizations that make up the average. The appearance of the second peak corresponds to structures which sometimes have one, and sometimes two (or more), large branches. In the case of 120° and 144° there is considerable fluctuation in the correlation functions (and the visual appearance) of each individual realization. This is a further indication that for some α in this range there is a critical point.

We have seen no indication that the correlation functions depend on the cluster size. For the case of 30° we grew clusters ten times larger than those described above to check this, with the result that the correlations were the same. The correlation function depends on the angular spread of the wedge, not on the space available to spread out, which indicates that the branches are self-similar in shape. If we take the point at which the $c(\alpha/2)$ crosses 0 as the criterion for determining η, we find that the typical distance between different major branches is $\eta=\alpha/2\approx60°-70°$. This is in rough agreement with our analytic estimates, and we take this as a verification of our basic idea.

We made another check by trying to quantify exactly what we mean by a ''major branch.'' We focus on the idea that for asymptotic behavior the most important feature is that some branches die, and some survive competition. To see this quantitatively we introduce a measure on DLA clusters which we call the *descendent measure* M_x. For this quantity we weight each point according to the number of descendents it has in the last fraction x of the growth. Thus M_1 measures the total number of points that grow from a given one, and, say, $M_{0.01}$ the number of descendent points in a tiny active zone on the outside of the cluster. The appearance of major branches derived this way is quite robust, and does not depend much on x. Clusters with M_1 weighting are shown in Fig. 3, and Fig. 1(b) illustrates that the correlation functions near the critical angle are not much different with the M_x weighting. The critical angle is robust, but for small angles the measure clearly localizes the main branch much more cleanly than the measure that uniformly weights the mass since it prunes sidebranches. The M_x weighting could be interesting in other contexts, since it provides a definition of a backbone for DLA.

Some aspects of the idea that we have proposed here has

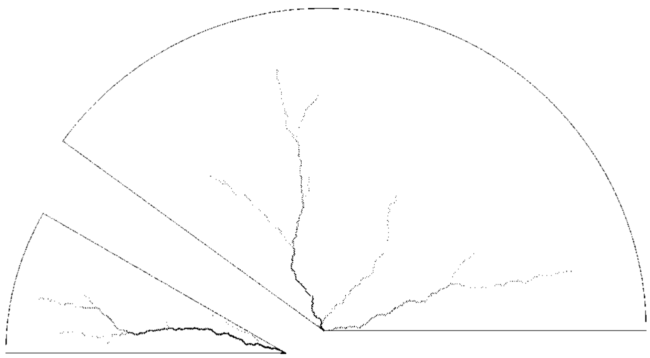

FIG. 3. DLA clusters plotted with gray level equal to M_1 and $\alpha = 30°$ and 144°.

appeared in other forms previously. For example, Arneodo *et al.* observed some hints of a fivefold structure in DLA [17]. This is more or less what we find since our angle η is close to $2\pi/5$. Many workers have noted that DLA clusters seem to have five major arms, but this qualitative impression was not supported by a quantitative estimate of the type we have given here.

We think that we should follow up our idea by checking it for radial viscous fingering in direct simulations. We hope that sophisticated methods such as the vortex sheet technique [18] could allow us to do this, though this is a computation intensive approach. The effect of the exact form of the surface tension can also be checked, although in our opinion the role of the surface tension is only to regularize the equations; its exact form (e.g., the finite size of the DLA particles acts as an effective surface tension) is unimportant. Ideally we should also try to put this idea of a structure made up of building blocks with some typical angle into a more general theoretical context. However, we do not see any obvious relationship between what we have done and the other theoretical approaches to Laplacian growth [19].

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[1] T. A. Witten and L. M. Sander, Phys. Rev. Lett. **47**, 1400 (1981); Phys. Rev. B **27**, 5686 (1983).

[2] T. Vicsek, *Fractal Growth Phenomena* (World Scientific, Singapore, 1992).

[3] L. Paterson, Phys. Rev. Lett. **52**, 1621 (1985).

[4] L. Sander, P. Ramanlal, and E. Ben-Jacob, Phys. Rev. A **32**, 3160 (1985).

[5] P. Pelce (ed.), *Dynamics of Curved Fronts* (Academic, Boston, 1988).

[6] H. Thome, M. Rabaud, V. Hakim, and Y. Couder, Phys. Fluids A **1**, 224 (1989).

[7] M. Ben Amar, Phys. Rev. A **43**, 5724 (1991); **44**, 3673 (1991).

[8] S. K. Sarkar, Phys. Rev. Lett. **65**, 2680 (1990).

[9] M. Ben-Amar, V. Hakim, M. Mashaal, and Y. Couder, Phys. Fluids A **3**, 1687 (1991).

[10] M. Ben-Amar and E. Brener, Phys. Rev. Lett. **75**, 561 (1995); Physica D **98**, 128 (1996).

[11] We have made a further approximation here. In our stability analysis we use the steady state solution to the dendrite problem. For the wedge and for the channel there are two solutions for each surface tension corresponding to different finger widths. In the wedge, the solution with the smaller width is the stable one, and the opposite is true in the channel. However, for the question of finger competition, we think it does not matter which solution one looks at, since the two solutions differ only in their local tip structure. Finger competition is a more global question.

[12] B. Derrida and V. Hakim, Phys. Rev. A **45**, 8759 (1992).

[13] L. Turkevich and H. Scher, Phys. Rev. Lett. **55**, 1026 (1985).

[14] R. C. Ball, Physica A **140**, 62 (1986).

[15] R. C. Ball and R. M. Brady, J. Phys. A **18**, L809 (1985).

[16] Similar indication of coexisting branches has been found in density profiles of DLA clusters grown in wedges with reflective walls [A. Arneodo, J. Elezgaray, M. Tabard, and F. Tallet, Phys. Rev. E **53**, 6200 (1996)].

[17] A. Arneodo, F. Argoul, E. Bacry, J.-F. Muzy, and M. Tabard, Phys. Rev. Lett. **68**, 3456 (1992).

[18] T. Hou, J. Lowengrub, and M. Shelley, J. Comput. Phys. **114**, 312 (1994).

[19] L. Pietronero, E. Erzan, and C. Evertsz, Phys. Rev. Lett. **61**, 861 (1988); T. Halsey, *ibid.* **72**, 1228 (1994).