

## Coarsening of Fractal Viscous Fingering Patterns

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We have measured the coarsening due to surface tension of radially grown fractal viscous fingering patterns. The patterns at late times depend on the structural form at the onset of coarsening, providing information on the age of the fractal. The coarsening process is not dynamically scale invariant, exhibiting two dynamic length scales that grow as  $L_1(t) \sim t^{0.22 \pm 0.02}$  and  $L_2(t) \sim t^{0.31 \pm 0.02}$ . The measured exponents are in agreement with the results of recent numerical studies of diffusion-controlled coarsening of a diffusion-limited aggregation fractal [Lipshtat *et al.*, Phys. Rev. E **65**, 050501 (2002)].

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Domain coarsening has been extensively studied for two-phase systems such as Ising ferromagnets [1], binary alloys [2,3], and binary fluids [4]. Quenching from a stable state into an unstable state initiates phase ordering that is often scale invariant (described by a single time-evolving scale) [5]. In another class of systems, initial unstable configurations are generated by a growth process involving morphological instabilities. An unstable deterministic growth process accompanied by noise can generate fractal surfaces, as in solidification from a supersaturated solution [6], growth of thin films during deposition [7], and the formation of a fracture surface in a brittle material [8] or geological landscape [9]. When the driving force is removed, these far-from-equilibrium systems begin to relax. Relaxation in such systems has been scarcely studied, and it is not known how the fractal scaling affects the relaxation process. Numerical simulations have revealed fragmentation of relaxing fractal clusters [10–12], and similar fragmentation has been observed during the relaxation of thin metal films deposited on a substrate [7]. More recent simulations [13,14] have revealed an absence of scale invariance; instead, the coarsening (due to diffusion) was characterized by two length scales increasing with different powers of time. This loss of scale invariance occurs when a relaxation process locally conserves the order parameter (components of one phase can move around but cannot become part of the other phase as they can in an Ising system).

We have conducted the first study of coarsening of a radial viscous fingering pattern (Fig. 1). The pattern is generated by the penetration of air into a thin oil layer contained between two closely spaced plates. We find that the coarsening is characterized by two length scales that have the same scaling as found to describe the coarsening of a diffusion-limited aggregation (DLA) fractal cluster [13,14]. We first describe the experiment and then present the data analysis that yields two length scales. We conclude by showing that the initial conditions affect the late time structures.

Our experiments use a radial Hele-Shaw cell formed of two 288 mm diameter optically polished glass plates

(60 mm thick) separated by a gap of 0.127 mm. The plates are clamped into an aluminum holder with a 25.4 mm thick Plexiglas clamp, which seals a 12.7 mm annular buffer around the plates. The buffer and gap are filled with silicon oil (viscosity  $\mu = 345$  mPa s, surface tension  $\sigma = 21.0$  mN/m). Fingering patterns are generated by opening the buffer to a reservoir below atmospheric pressure and allowing air at atmospheric pressure to flow into the gap through a center hole in the bottom plate. The high rigidity and flatness of the cell permit a pressure difference up to 1 atm while keeping the variation in the gap below 1%. The resultant highly branched patterns have a minimum finger width more than 2 orders of magnitude smaller than the cell size. When a cluster has grown to the size of the cell, we block both the air and oil lines, and the cluster relaxes due to surface tension. Megapixel images are obtained at a rate decreasing from 12/s to 1 every 250 s for a total time of  $10^5$  s. Subtraction of background and thresholding then provide binary images of the two phases.

The observed coarsening of a viscous fingering pattern is compared in Fig. 1 with the results of Conti *et al.* [13] for simulation of diffusion-controlled coarsening of a DLA cluster. In both cases the initial highly branched fingering pattern at  $t = 0$  becomes smoother as time progresses, and the cluster breaks into fragments, while its radius of gyration  $r_g$  remains constant. (In the experiment  $r_g$  actually decreases slightly, 2% in  $2 \times 10^4$  s.) Conti *et al.* [13] argue that a scale-invariant relaxation *must* involve a significant decrease in  $r_g$ . The lack of such a decrease in our experiments is the first indication of a breakdown of scale invariance.

Properties of the clusters are deduced from the density-density correlation function,  $C(r, t) = \langle [\rho(r', t)][\rho(r' + r, t)] \rangle$ , normalized at  $r = 0$ , with  $\rho = 1$  in the air phase and  $\rho = 0$  in the oil phase. At intermediate length scales  $C(r, t)$  decays as  $r^{-\delta}$  with an exponent  $\delta$  that is related to the fractal dimension of the cluster by  $D = 2 - \delta$ . This power law is cut off at length scales comparable to the system size. At early times we find  $C \sim r^{-0.29}$  over more than a decade of length scales [Fig. 2(a)], indicating

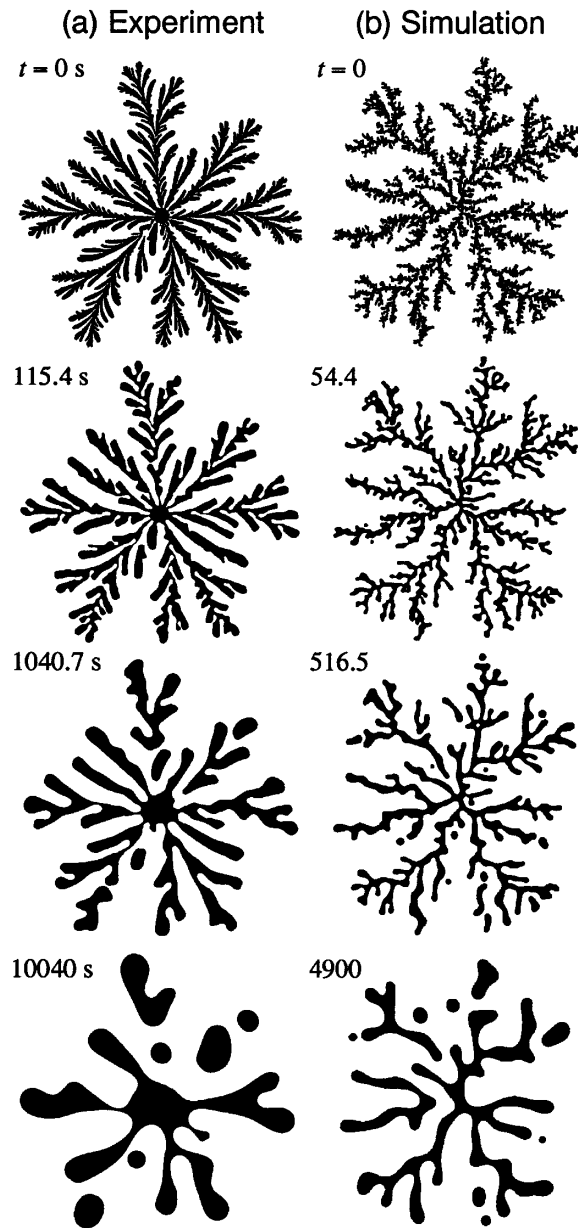


FIG. 1. Coarsening of fractal clusters. (a) Images of an observed fingering pattern (cluster diameter 200 mm) relaxing due to surface tension. (b) A DLA fractal undergoing diffusive coarsening, obtained in a simulation by Conti *et al.* [13]. The simulation used a dimensionless time step that corresponds to 2.1 s in the viscous fingering relaxation. The relaxation in both systems is characterized by the smoothing of small scales, fragmentation of the cluster, and a constant radius of gyration.

fractal scaling with dimension  $D = 1.71 \pm 0.03$ . As a check, we also computed the dimension directly using a box counting method, which yielded  $D = 1.70 \pm 0.03$ . Our result for  $D$  is in accord with the DLA fractal dimension [15],  $D = 1.713 \pm 0.003$ . The question of whether Laplacian growth, including radial viscous fin-

gering, is the continuum limit of DLA is currently the focus of much research [16,17], but this question is beyond the scope of the present work.

At very small scales  $C(r, t)$  decays linearly [Fig. 2(b)] with an inverse slope proportional to the smallest length scale,  $L_1(t)$ , which increases with time (Porod law [5]); for lengths smaller than  $L_1$  the interface appears smooth.

Despite the dramatic time evolution of a cluster (Fig. 1), the correlation function reveals that the large-scale structure remains frozen [see Figs. 2(a) and 2(c)]. The combination of an increasing lower cutoff and a frozen tail must lead to the appearance of a second length scale: Because of the conservation of volume, the integral of  $rC(r, t)$  with respect to  $r$  is invariant in time. The thickening of small scales while the largest scales remain frozen requires the dilution of intermediate scales. Hence a dip forms in  $C(r, t)$ , as can be seen in Fig. 2(a). The position of the minimum corresponds to about half the distance between adjacent arms of a cluster. We define  $L_2$  to be the location of this minimum. This definition of the larger length scale differs from that of Lipshtat *et al.* [14], who obtained a length  $\ell_2$  by finding the smallest radius beyond which  $|C(r, t) - C(r, 0)|$  was smaller than a predetermined threshold. The length scale  $\ell_2$  was thus associated with the boundary between the dilute and the frozen parts of the correlation function. Our method for obtaining  $L_2$  is more robust and less arbitrary than the method for determining  $\ell_2$ , but we find that within the uncertainty the two lengths are proportional, which indicates that the interarm spacing determines the location of the correlation function's frozen tail.

At  $t = 0$ , both  $L_1$  and  $L_2$  are equal to the same cutoff length scale  $L_0$ , but at long times we find  $L_1 \propto t^{0.22 \pm 0.02}$  and  $L_2 \propto t^{0.31 \pm 0.02}$  [Fig. 2(d)]. Nearly the same exponent values were obtained for the two length scales in diffusion-controlled coarsening of a DLA cluster,  $0.22 \pm 0.01$  and  $0.32 \pm 0.01$  [14]. The different growth rates of  $L_1$  and  $L_2$  exclude any global rescaling that would lead to the same form for  $C(r, t)$  at all times, as can be seen in Fig. 2(a).

In a scale-invariant coarsening process, information about the initial structure is lost, but this is not the case in the relaxation of viscous fingering patterns: the longer the system evolves, the larger  $L_2$  is compared to  $L_1$  [Fig. 2(d)], and the initial conditions of the cluster are *not forgotten*. Instead, they determine a unique combination of  $L_1$  and  $L_2$  at any time.

The effect of the initial conditions at late times is illustrated by the clusters in Fig. 3. Different initial pressure differences produced clusters with different minimum length scales  $L_0$  but did not affect the cluster fractal dimension [18]. We allowed each cluster to evolve until the lower length scale reached the same value,  $L_1(t) = 14.0$  mm. Clusters that evolved from a smaller  $L_0$  had to evolve over longer times, so  $L_2$  became larger for those old clusters, which have greater fragmentation

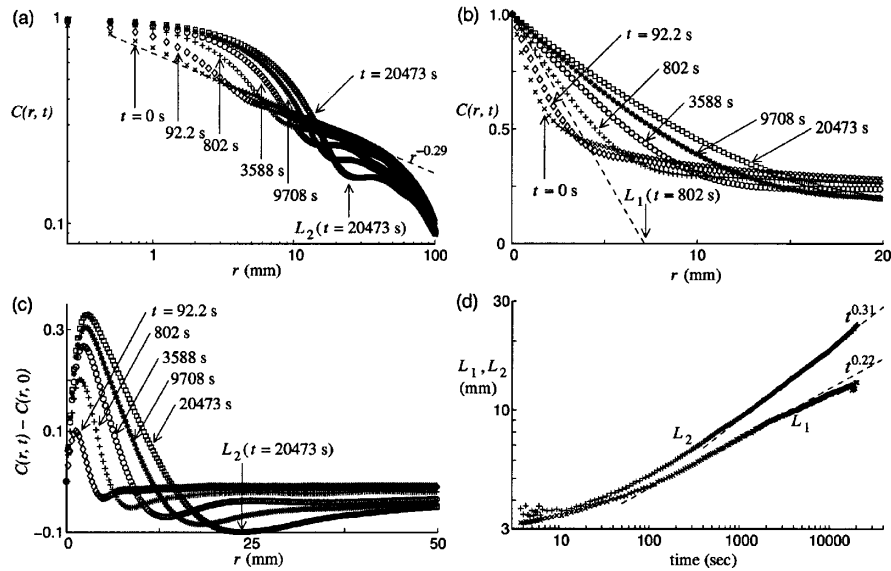


FIG. 2. The time evolution of the correlation functions and length scales. (a) Correlation functions at different times  $t$ , averaged over 16 experimental runs. The dashed line shows a fit to the fractal scaling region for the  $t = 0$  curve, where  $C \propto r^{-0.29}$ , yielding a fractal dimension of  $D = 1.71 \pm 0.03$ . The scaling regime shortens and eventually disappears as  $C(r, t)$  develops a minimum. (b) The same correlation functions on a linear plot, showing a linear decay for the smallest scales.  $L_1(t)$ , the length scale below which the cluster is smooth (nonfractal), is obtained by measuring the inverse slope of a linear fit to  $C(r, t)$  at small scales. The dashed line shows such a fit for  $C(r, t = 802 \text{ s})$ , which yields  $L_1 = 7.14 \text{ mm}$ . (c) The difference  $C(r, t) - C(r, 0)$  has a minimum that is taken to be  $L_2(t)$ , which is about half the interarm distance. (d) The time evolution of the two length scales with power law fits at long times (dashed lines). At late times ( $t \geq 4000 \text{ s}$ ),  $L_1$  grows more slowly as the smaller bubbles approach their equilibrium size after fragmentation.

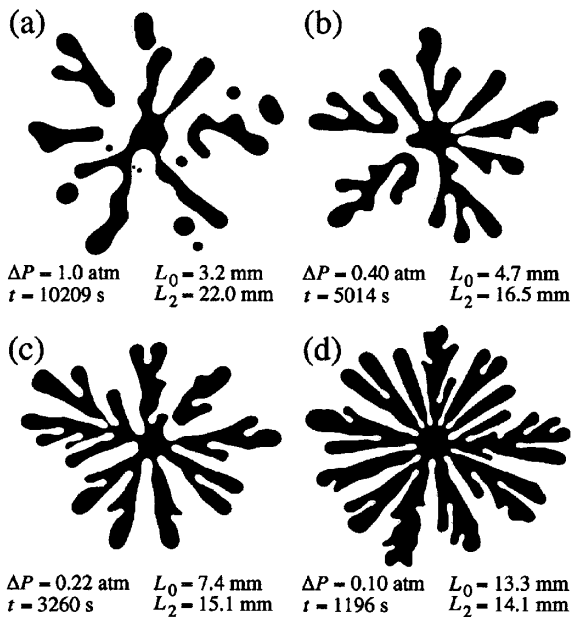


FIG. 3. Four clusters generated with different pressure differences  $\Delta P$ . The clusters have different initial length scales  $L_0$ , but the lower cutoff length has evolved in time to the same value,  $L_1 = 14.0 \text{ mm}$ . The different evolution times lead to different lengths  $L_2$ , mirrored by enhanced fragmentation of older clusters. In contrast to scale-invariant coarsening, the initial conditions are not forgotten.

and arm separation than the young clusters. Thus information about the initial configuration is retained. In principle a cluster's age could be estimated from measurements of  $L_1$  and  $L_2$  at a late time, but a precise determination of age would require an understanding of the evolution at early times, before the onset of the power law scaling ( $t \lesssim 100 \text{ s}$ ).

In conclusion, we observe the loss of scale invariance in the coarsening of fractal viscous fingering patterns: there is a small length scale below which the cluster is smooth, and a larger length scale corresponding to half the distance between adjacent cluster arms, and these two length scales are described by different power laws. The existence of the larger length scale is a consequence of volume conservation together with local smoothing in the relaxation process. The breakdown of scale invariance suggests a scenario in which the present configuration of other fractals, such as in geological systems, might include information about their age and initial morphology.

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