Breakdown of Scale Invariance in the Phase Ordering of Fractal Clusters

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Numerical simulations with the Cahn-Hilliard equation show that coarsening of fractal clusters (FCs) is not a scale-invariant process. On the other hand, a typical coarsening length scale and interfacial area of the FC exhibit power laws in time, while the mass fractal dimension remains invariant. The initial value of the lower cutoff is a relevant length scale. A sharp-interface model is formulated that can follow the whole dynamics of a diffusion controlled growth, coarsening, fragmentation, and approach to equilibrium in a system with conserved order parameter. [S0031-9007(98)06180-8]

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Nonequilibrium driven dissipative systems relax to equilibrium after the driving agent is "switched off" or depleted. In complex systems the relaxation dynamics can be quite complicated, and it is natural to seek for dynamic scaling and universality. An instructive, exactly solvable nonlinear example of dynamic scaling in relaxation (coarsening) of rough (self-affine fractal) surfaces with *nonconservative* dynamics is given by the deterministic (undriven) KPZ equation [1,2]. A much older example is decay of homogeneous and isotropic hydrodynamic turbulence [3,4]. Finally, there is an important class of relaxation problems related to phase ordering dynamics, nonconserved and conserved, in the bulk and on the surface [5–7].

If the system obeys a conservation law, "switching off" of the driving agent occurs naturally. There are many important nonequilibrium systems that exhibit morphological instabilities and ramified growth at an early stage of the dynamics, show phase ordering at an intermediate stage, and finally approach a simple equilibrium. A canonical example is provided by diffusion controlled systems, such as deposition of solute from a supersaturated solution and solidification from an overcooled liquid. The stage of morphological instability and its implications have been under extensive investigation [8–12]. If some noise is present, a fractal cluster (FC) can develop at this stage [12]. The subsequent surface tension driven coarsening of this FC is unavoidable in a closed geometry with a finite amount of mass or heat. This stage has not received much attention, with the exception of the paper by Irisawa *et al.* [13] where two-dimensional Monte Carlo simulations were performed, and a power law found for the perimeter of a diffusion-limited aggregation (DLA) cluster versus time.

We are aware of two additional physical systems with a conservation law, for which numerical simulations showed nontrivial fractal coarsening dynamics and power laws for the cluster perimeter: interface controlled [13,14] and surface diffusion controlled [13,15] systems. Besides, Stokes flow controlled coarsening has been discussed in the context of sintering of fractal matter [15].

No theory is available for any of these fractal coarsening systems, except for the very late postfragmentation stage [16]. On the other hand, a FC is a particular case of disordered media with long-range (power-law) spatial correlations [17]. The scaling hypothesis (SH) (the cornerstone of modern theory of phase ordering [18]) does not exclude FCs when dealing with long-range correlations in the initial condition [6]. Therefore, one is tempted to employ the SH and calculate the growth exponents for the coarsening of FCs. We start with these simple calculations. Then we report our simulations of the diffusion controlled coarsening of a DLA aggregate, as described by the Cahn-Hilliard (CH) equation. Having measured, for the first time, the dynamics of the pair correlation function (which is very close to the average mass density) of the FC, we show that the SH is invalid. On the other hand, we find that a characteristic coarsening length scale and interfacial area of the FC exhibit power-law dynamics (with a *new* growth exponent), while the fractal dimension remains invariant (on an interval of scales shrinking with time). The initial value of the lower cutoff of the FC is shown to be an additional relevant length scale. Finally, a minimalistic sharp-interface model is presented that can follow the whole dynamics of the diffusion controlled system: an unstable growth, coarsening, fragmentation, and approach to equilibrium.

Let the initial state of a conserved system represent a single-connected, statistically homogeneous self-similar mass fractal of the minority phase, characterized by the fractal dimension D on an interval of scales between the lower cutoff l_0 and upper cutoff L. We start with a simple

coarsening scenario [19,20] that is *required* by the SH. It assumes that the fractal dimension of the coarsening cluster remains constant on a (shrinking) interval of scales between the time-dependent lower and upper cutoffs, $l(t)$ and $L(t)$. The interfacial area A and total mass M of the FC are estimated as [17]

$$A \sim l^{d-1}\,(L/l)^D \quad \text{and} \quad M \sim l^d\,(L/l)^D, \tag{1}$$

respectively, where d is the embedding Euclidean dimension. Mass conservation yields $L \sim l^{(D-d)/D}$ [20]. Now assume that $l(t) \sim t^{1/z}$. Then we find the following scaling laws: $A(t) \sim t^{-1/z}$ and $L(t) \sim t^{(D-d)/Dz}$. The scaling of $L(t)$ describes shrinking of the FC in the process of coarsening [20].

Already at this stage a discrepancy appears: no shrinking has been observed in any direct numerical simulations of coarsening of FCs [13–15]. This gives a strong evidence for breakdown of scale invariance [21]. On the other hand, power laws for $A(t)$ reported in Refs. [13–15] indicate that the problem might possess scaling behavior of a more complicated nature.

To clarify the matter, we performed more detailed numerical simulations of a diffusion controlled system. In addition to $L(t)$ and $A(t)$, we followed the evolution of the equal-time pair correlation function (which is very close to the average mass density of the FC, so we will not distinguish between them). Having measured it, one could find the mass fractal dimension and coarsening length scale of the FC for every moment of time.

If one remains, for one more moment, within the framework of the SH, one can easily predict the dynamics of the mass density $\rho(r,t)$. At distances $r \ll l(t)$ from a (typical) reference point inside the cluster, the cluster is nonfractal: $\rho(r,t) \sim$ const. At distances intermediate between l and L, $\rho(r,t) = a(t)\,r^{D-d}$, where $a(t)$ is a function of time. Matching these two asymptotics, we have $a(t)\,l^{D-d} =$ const and hence $a(t) \sim t^{(d-D)/z}$. Therefore, for $l(t) \ll r \ll L(t)$ the SH predicts $\rho(r,t) \sim (r/t^{1/z})^{D-d}$, a simple self-similar expression. It is the absence of this self-similarity that will enable us to utterly disprove the SH.

We concentrated on the diffusion controlled coarsening and employed the CH equation, a standard model of phase ordering with a conserved order parameter (COP) [5,6],

$$\frac{\partial u}{\partial t} + \frac{1}{2}\,\nabla^2[\nabla^2 u + u - u^3] = 0\,. \tag{2}$$

Equation (2) was discretized and solved on the domain $0 \le x \le 512\,, 0 \le y \le 512$ with periodic boundary conditions. We used an explicit Euler integration scheme to advance the solution in time, and second order central differences to discretize the Laplace operator. With a mesh size $\Delta x = \Delta y = 1$ no preferred directions emerged in the computational grid, due to the truncation errors; a time step $\Delta t = 0.05$ was required for numerical stability. The accuracy was monitored by checking the mass conservation that was verified in all the simulations within 0.01%.

We chose a DLA cluster [22] as the initial condition. The fractal properties of DLA clusters are somewhat more complex that those of a simple self-similar fractal [23]. However, it is a DLA-like FC that can develop during the diffusion controlled growth [12], so this choice is physically motivated. The initial clusters (like the one shown in Fig. 1, upper left), with radius of order 250, were prepared by a standard random-walk algorithm on a two-dimensional square grid. To prevent fragmentation at an early stage of the coarsening process, we followed the technique of Irisawa *et al.* [13,14]: the aggregates were thickened by an addition of peripheral sites. The mass fractal dimension was determined from the mass-radius relation [17] and ranged from 1.67 to 1.72.

We identified the cluster as the locus where $u(\mathbf{r},t) \ge 0$. The coarsening process was followed up to a time $t = 5000$. Typical snapshots of the coarsening process are shown in Fig. 1. One can see that smaller features of the FC are "consumed" by larger features, while the global structure of the cluster is not affected. To characterize the coarsening process, the following quantities were sampled and averaged over 10 initial configurations: (1) the gyration radius of the cluster, (2) the circularly averaged pair correlation function $g(r,t) = \langle[u(r',t) + 1][u(r' + r,t) + 1]\rangle$, (3) the cluster perimeter $A_1(t)$, defined as the sum of $|\nabla u(\mathbf{r},t)|^2$ over the whole domain, and (4) the cluster perimeter $A_2(t)$, defined as the number of broken bonds between the aggregate sites.

The gyration radius of the FC has been found to remain constant within possible logarithmic corrections. Evolution of $g(r,t)$ is shown in Fig. 2. One can see that coarsening affects only the smallest lengths, while the intermediate-distance power-law part remains "frozen." It

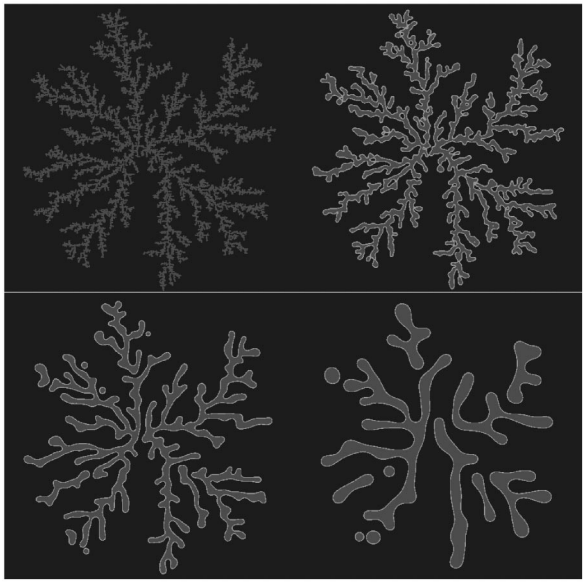

FIG. 1. Evolution of a DLA cluster undergoing coarsening in a conserved, diffusion controlled system. The upper row corresponds to $t = 0$ (left) and 34.7 (right), the lower row to $t = 329.3$ (left) and 4900 (right).

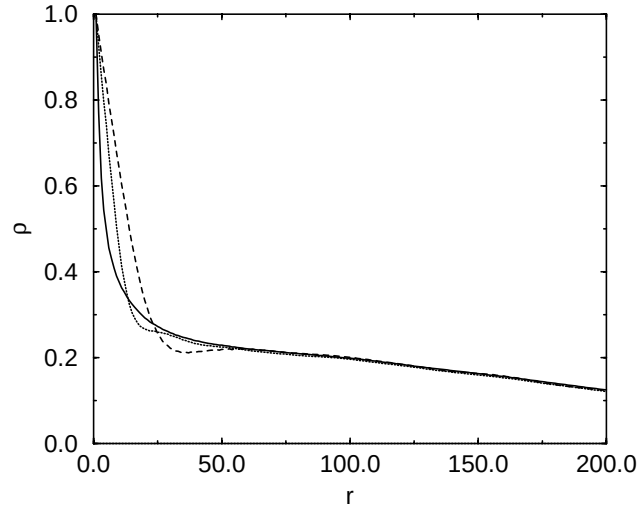

FIG. 2. Dynamics of the pair correlation function $g(r,t)$ for time moments $t = 0$ (solid line), 516.5 (dotted line), and 4900 (dashed line).

is evident that $g(r,t)$ does not acquire a self-similar form, so there is no scale invariance. On the other hand, the mass fractal dimension remains invariant on an interval $l_c(t) \ll r \ll L$. The dynamics of the coarsening length scale $l_c(t)$, extracted for each moment of time from the slope of the linear part of $g(r,t)$ (the Porod law [6]), are shown in Fig. 3. The late-time behavior of the slope versus time shows a power law: $t^{-\alpha_1}$ with $\alpha_1 = 0.19$. Therefore, $l_c(t) \sim t^{\alpha_1}$, and the corresponding growth exponent $z_1 = 1/\alpha_1$ is close to 5 (and not to 3 as could be expected for a diffusion controlled system with a COP [6]). Figure 4 shows the dynamics of the cluster perimeter estimates $A_1(t)$ and $A_2(t)$. The long-time dynamics of each of them is describable by a power law $t^{-\alpha_2}$, with $\alpha_2 = 0.19$ for A_1, and 0.20 for A_2. The corresponding result of Monte Carlo simulations [13] was slightly different: 0.22–0.24. The close proximity of the exponents α_1 and α_2 gives evidence that it is a single exponent.

Absence of scale invariance means presence of an additional length scale. Function $g(r,t)$ gives evidence for the nature of this length scale. For our mass fractal at $t = 0$ we have $g(r,\, t = 0) \sim (r/l_0)^{D-d}$ in the fractal region $l_0 \ll r \ll L$. Preservation of the power-law part of g with time (Fig. 2) implies that the same asymptotics holds, on a shrinking interval of radii, for $t > 0$ (until fragmentation). That is, the small intrinsic length scale l_0 remains relevant. How does it show up in the phenomenology of coarsening? Figure 1 gives evidence that (i) the FC can be regarded as a set of "bars," and (ii) the characteristic bar length l_b grows in time faster than the bar width (identified with l_c). The area of a single bar should scale like $l_b\, l_c$, hence the total area of the FC is $l_b\, l_c\, (L/l_b)^D$. This quantity must be equal to the initial value of the FC area, $l_0^2\, (L/l_0)^D$. This yields $l_b \sim l_0\, (l_c/l_0)^{1/(D-1)} \sim t^{\alpha_1/(D-1)}$.

We will finish this Letter with formulating a sharp-interface model that can describe the *whole* diffusion controlled dynamics, from the stage of growth through coarsening and fragmentation to the final equilibrium. Consider a number of (possibly multiple-connected) mass clusters characterized by a set of their (moving) interfaces γ_i. Now let $u(\mathbf{r},t)$ be the mass concentration of the solution normalized to the (constant) density of solute in the compact solid phase. The field u in the liquid phase is governed by the diffusion equation

$$\frac{\partial u}{\partial t} = \chi \nabla^2 u \tag{3}$$

in a finite d-dimensional domain. We specify a no-flux boundary condition, $\nabla_n u|_\Gamma = 0$ on the external boundary Γ, where index n stands for the normal component of a vector. Assuming that each of the interfaces γ_i is in local thermodynamic equilibrium, we employ the

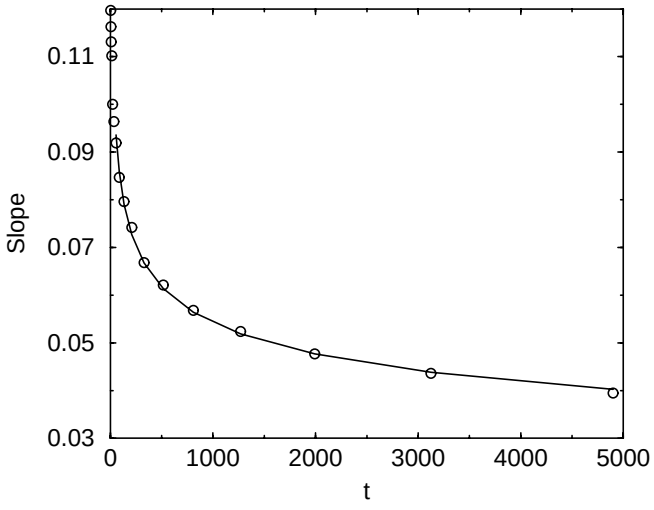

FIG. 3. The slope of the linear part of $g(r,t)$ versus time, and its power-law regression.

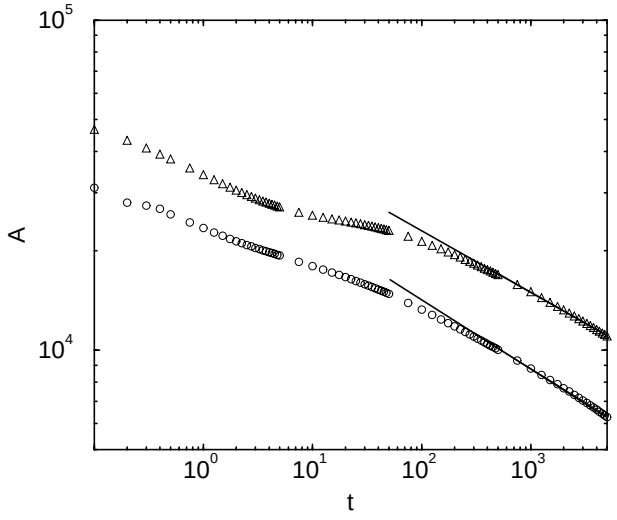

FIG. 4. Two estimates for the FC perimeter versus time: A_1 (triangles) and A_2 (circles).

Gibbs-Thomson relation $u|_{\gamma_i} = u_0(1 + \lambda_0\kappa_i)$, where u_0 is the (normalized) equilibrium concentration of the solution in the bulk, λ_0 is the capillary length, and κ_i is the local curvature for $d = 2$, or the mean curvature for $d > 2$. (We limit ourselves to an isotropic surface tension.) Finally, mass conservation at each of the moving interfaces yields the well-known relation for the normal speed,

$$v_n^{(i)} = \left. \frac{\chi \nabla_n u}{1 - u} \right|_{\gamma_i} . \tag{4}$$

It is easy to check that this model preserves the total mass of the solute. In the normalized form

$$\Omega_c + \int_\Omega u \, d\mathbf{r} = \text{const}, \tag{5}$$

where Ω_c is the total volume (area) of the solid phase, while Ω denotes the region unoccupied by the solid phase. This important conservation law does not appear in the more traditional theoretical formulations of the diffusion controlled growth problem [8–12], where an "infinite" system is studied, and the boundary condition corresponding to a constant (positive) flux or constant supersaturation at $\mathbf{r} \to \infty$ is used. Notice that, even in the limit of strong diffusion, it is the full diffusion equation (rather than its Laplace's equation limit) and no-flux condition on Γ that provide the conservation law. Also, the usually small term u in the denominator of Eq. (4) should be kept to get Eq. (5) right.

In summary, we have demonstrated that diffusion controlled phase ordering of FCs is not a scale-invariant process. In spite of this, the problem possesses nontrivial scaling properties: the coarsening length scale and interfacial area of the FC exhibit power laws in time (with a new growth exponent), and the mass fractal dimension remains invariant. An additional small intrinsic length scale (the initial value of the lower cutoff) remains relevant until the fragmentation stage. We believe that these findings apply to other coarsening mechanisms as well.

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