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Anomalous dynamic scaling in locally conserved coarsening of fractal clusters

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We report two-dimensional phase-field simulations of locally conserved coarsening dynamics of random fractal clusters with fractal dimension $D=1.7$ and 1.5. The correlation function, cluster perimeter, and solute mass are measured as functions of time. Analyzing the correlation function dynamics, we identify *two* different time-dependent length scales that exhibit power laws in time. The exponents of these power laws do not show any dependence on D; one of them is apparently the ''classical'' exponent 1/3. The solute mass versus time exhibits dynamic scaling with a D-dependent exponent, in agreement with a simple scaling theory.

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Many nonequilibrium systems develop morphological instabilities and ramified growth at an early stage of the dynamics, exhibit coarsening at an intermediate stage, and finally approach a simple equilibrium. A classic example is the diffusion-controlled systems, such as deposition of solute from a supersaturated solution and solidification from an overcooled melt. The stage of morphological instability has been under extensive investigation [1–4]. If some noise is present, fractal clusters (FCs) similar to diffusion-limited aggregates (DLA) can develop [3]. If the total amount of mass or heat is finite, the subsequent dynamics are dominated by surface-tension-driven relaxation (coarsening) [5–7]. Coarsening of FCs in systems with conserved order parameter, apart from being interesting in its own right, exemplifies a more general problem of *phase ordering*: emergence of order from disorder, following a quench from a disordered state into a region of phase coexistence [8,9]. This example is nontrivial because of the long-ranged, power-law correlations intrinsic in FCs. The role of the long-ranged correlation in phase ordering dynamics has been under debate in connection to *dynamic scale invariance* (DSI), a major simplifying factor in theory [9]. DSI presumes that there is, at late times, a *single* dynamic length scale $l(t)$ so that the correlation function $C(r,t)$ approaches a self-similar form $g[r/l(t)]$ [9]. In locally conserved systems (model B) $l(t)$ is expected to show dynamic scaling (by which we simply mean a power law in time) with ''classical'' dynamic exponent 1/3 [8–10].

Because of the complexity of the phase-ordering dynamics, DSI has not been proven, except for a very few simple models [9]. However, there is extensive evidence, from experiments and simulations, supporting DSI in conserved systems with *short-ranged* correlations. Recently, phase-field simulations of coarsening of a *globally* conserved, interface-controlled system with *long-ranged* correlations were performed [11]. DLAs with fractal dimension $D=1.75$ served in Ref. [11] as the initial conditions for the minority phase. Notice that a FC is a particular case of systems with long-ranged correlations. D is equal to the exponent σ that appears in the power-law decay of the correlation function at $t=0$: $C(r,t=0)\sim r^{-(d-\sigma)}$, where d is the Euclidian dimension [4]. The simulations [11] have not found any deviations from DSI and yielded the ''classical'' value of the dynamic exponent for the globally conserved model, which is 1/2.

The results [11] stand in sharp contrast with simulation results on *locally* conserved coarsening of DLA clusters, where breakdown of DSI was observed [6]. Breakdown of DSI is related to the fact that the upper cutoff L of the FCs remains almost constant in the process of coarsening [5–7]. While interpreting this finding, one should distinguish between two regimes: diffusion controlled $l_d<L$, and Laplacian $l_d>L$, where $l_d\sim t^{1/2}$ is the diffusion length. It is clear that L should remain almost constant in the diffusion-controlled regime, as interaction between the far-lying parts of the cluster is exponentially small in this case. The inequality $l_d<L$ was satisfied in the simulations of Refs. [6,7], and it is satisfied in the simulations reported in the present work. However, even in the opposite limit of Laplacian coarsening one can expect L to remain almost constant (and breakdown of DSI to persist), this time because of Laplacian screening of transport. An additional nontrivial aspect of *locally* conserved coarsening of DLA clusters is the following. Though not dynamically scale invariant, this coarsening process was shown to exhibit dynamic scaling (with ''unusual'' exponent 0.21–0.22) [5–7]. This implies hidden simplicity of a more complex nature than DSI.

We report phase-field simulations of locally conserved coarsening of three sets of random FCs. The clusters have fractal dimensions $D=1.7$ and 1.5. Our motivation was to check whether the ''anomalous'' scaling persists for clusters different from DLA, and to investigate its possible dependence on D. The simulations confirm breakdown of DSI in all cases. We identify *three* power laws in time. Two of them correspond to dynamic length scales found from the dynamics of $C(r,t)$ at small and intermediate distances. The first length scale, with an exponent 0.21–0.22, is the same as observed earlier. Surprisingly, it does not show any dependence on D. The second length scale has an exponent close to 1/3, and we suggest a simple interpretation for its appearance. The third dynamic exponent is found in the time dependence of the ''solute mass,'' and it is D dependent. We suggest a simple scaling theory for the second and third exponents.

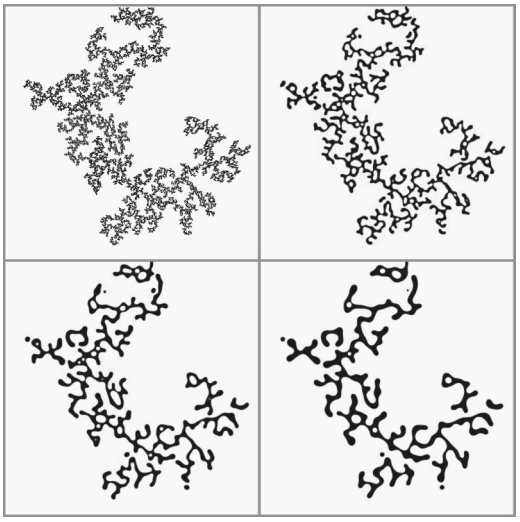

FIG. 1. Images of locally conserved coarsening of a FC with $D=1.7$ at times $t=0$ (upper left), 495 (upper right), 1963 (lower left), and 4920 (lower right).

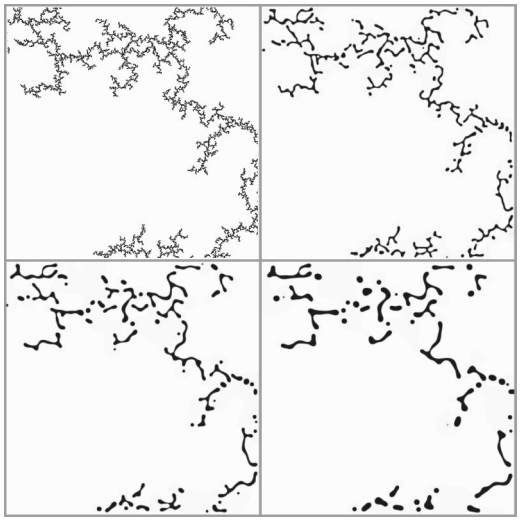

FIG. 2. Same as in Fig. 1, but for $D=1.5$.

Here is a brief description of our simulation techniques. We employed a hierarchical algorithm [12] to build random FCs with tunable fractal dimension. The algorithm starts with a set of 2^n square particles grouped into pairs. These pairs are then grouped into pairs of pairs, and so on, in an iterative procedure. After n iterations the final aggregate of 2^n particles is obtained. The desired fractal dimension D is achieved by an appropriate mutual position in which the aggregates are sticked together in each iteration [12]. The clusters obtained in this way are ''reinforced,'' by an addition of peripheral sites as suggested in Ref. [5], to avoid breakup at an early stage of coarsening. The quality of obtained FCs is controlled by computing the correlation function (see below) and checking the quality of the power law.

A standard phase-field model for locally conserved coarsening is the Cahn-Hilliard equation [8,9],

$$\frac{\partial u}{\partial t}+\frac{1}{2}\nabla^2(\nabla^2 u+u-u^3)=0. \tag{1}$$

This equation was discretized and solved numerically. Two different numerical schemes were employed to advance the solution in time. The first scheme used an explicit Euler integration. The simulated domain Ω was a two-dimensional box 1024×1024 with no-flux boundary conditions (zero normal component of ∇u at the boundaries). The grid size was $\Delta x=\Delta y=1$, the time step $\Delta t=1/25$ ensured numerical stability. The time range of these simulations was $0<t<5000$. The second scheme used a semi-implicit Fourier spectral method [13]. The simulation box was 1792×1792, with periodic boundary conditions. The grid size was $\Delta x=\Delta y=1$, which corresponds to 1792 Fourier modes in each dimension. The time step was $\Delta t=0.3$, the total time range was $0<t<15\,000$. Overall, three series of simulations were performed. In two of them, with the explicit scheme, ten FCs with $D=1.7$ (series A) and ten FCs with $D=1.5$ (series C) served as the initial conditions for the minority phase $u=1$. In addition, seven FCs with $D=1.7$ (series B) were simulated with the spectral scheme. The minority phase area fractions were 10.5% (series A), 7.5% (series B), and 5.8% (series C). Notice that the initial cluster area in series B was almost twice as big as that in series A, while D was the same.

Equation (1) with either no flux, or periodic boundary conditions obey conservation laws: $I_0=\int\int_\Omega u(\mathbf{r},t)\mathbf{d}r=\text{const}$ and $\mathbf{I}_1=\int\int_\Omega \mathbf{r}u(\mathbf{r},t)\mathbf{d}r=\text{const}$. Our numerical schemes preserve I_0 exactly. We checked that they also preserve $\mathbf{I}_1$ with a very high accuracy, better than 5×10^{-3}% up to $t=5000$ (explicit scheme), and better than 0.05% up to $t=15\,000$ (spectral scheme). Additional tests of the two schemes included obtaining the stationary kink solution of Eq. (1) and observing the ''classic'' scaling with exponent 1/3 for the initial condition in the form of ''white noise.''

The cluster was identified as the locus of $B(\mathbf{r},t)=1$, where $B(\mathbf{r},t)=1$ for $u(\mathbf{r},t)\geqslant0$ and zero otherwise. Figures 1 and 2 show snapshots of the coarsening dynamics observed in series A and C. One can see that the larger features grow at the expense of the smaller ones. The global structure of the clusters remains unchanged. In particular, the cluster size is almost constant, in a marked contrast to the globally conserved case [11]. A significant decrease of L in time is a necessary prerequisite of *any* fractal coarsening that obeys DSI [6,11,14,15], therefore its absence clearly implies that DSI is broken. One can also notice a more pronounced breakup of the cluster for $D=1.5$. Still, even in this case the large-scale mass distribution in the cluster does not change much.

To characterize the coarsening dynamics, several quantities were sampled and averaged over the initial conditions in each of the three simulation series.

(1) (Circularly averaged) equal-time correlation function, normalized at $r=0$,

$$C(r,t)=\frac{\langle\rho(\mathbf{r}'+\mathbf{r},t)\rho(\mathbf{r}',t)\rangle}{\langle\rho^2(\mathbf{r}',t)\rangle}.$$

(2) Cluster perimeter $P(t)$ [16].

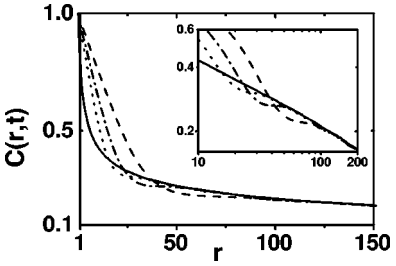

FIG. 3. Equal-time pair correlation function at different times for simulation series *B*. $t=0$ (solid line), $t=658.8$ (dotted line), $t=2787$ (dashed-dotted line) and $t=15\,000$ (dashed line). The inset shows a log-log plot of the same function. The dynamic length scale $l_2(t)$ (the kneelike feature) is apparent.

(3) Cluster mass $M(t)=\int\int_{B(\mathbf{r},t)=1}\rho(\mathbf{r},t)\mathbf{dr}$.

(4) "Solute" mass outside the cluster, $M_s(t)=\int\int_{B(\mathbf{r},t)=0}\rho(\mathbf{r},t)\mathbf{dr}$.

Here an auxiliary "density" field $\rho(\mathbf{r},t)=(1/2)[u(\mathbf{r},t)+1]$ is introduced that varies between 0 and 1. Also, $I_0=$const implies that $M(t)+M_s(t)=$const.

Figure 3 shows $C(r,t)$ at different times for simulation series *B* ($D=1.7$). Coarsening occurs at small and intermediate distances, while the large-r tail remains almost unchanged. The fractal behavior is observed only in the "frozen" tail that shrinks in time. On the typical scales of coarsening, the cluster is not fractal anymore. This implies the breakdown of DSI at $D<2$. The same type of behavior of $C(r,t)$ was obtained for the locally conserved coarsening of DLA clusters [6,7]. At small distances, $C(r,t)$ behaves linearly with r, $C(r,t)\simeq 1-r/l_1(t)$, as expected from the Porod law [9]. This asymptotics yields dynamic length scale $l_1(t)$. One can also see a kneelike feature at intermediate distances, which develops in the course of time. This feature yields dynamic length scale l_2. We define l_2 as the maximum value of r for which the relative difference between $C(r,0)$ and $C(r,t)$ is not greater than 3% (the results are insensitive to the exact value of this threshold). Notice that, by the end of the simulation time, the diffusion length l_d is already greater than 10^2. Still, we do not see any signature of l_d in the shape of $C(r,t)$ at large distances. We expect that the tail of $C(r,t)$ will remain "frozen" until late times, and DSI

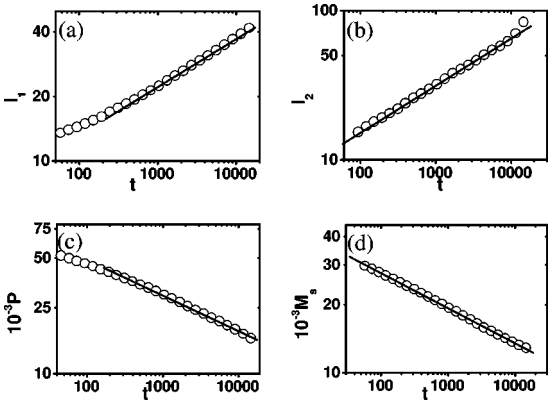

FIG. 4. l_1 (a), l_2 (b), P (c), and M_s (d) vs time for $D=1.7$. The solid lines are power-law fits.

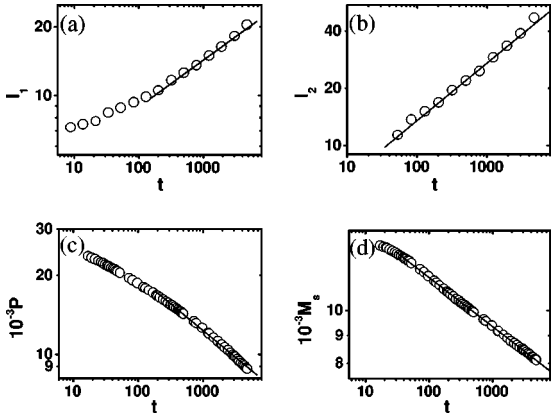

FIG. 5. Same as in Fig. 4, but for $D=1.5$.

broken, in the Laplacian regime as well.

Figures 4 and 5 show the measured quantities l_1, l_2, P, and M_s, averaged over the initial conditions, versus time for simulation series *B* and *C*, respectively. We fitted these time dependencies by power laws with exponents α_1, β, α_2, and γ, respectively. According to the Porod law [9], one expects $\alpha_1=-\alpha_2$. All simulation results are summarized in Table I. α_1 and α_2 are close to the previously reported values [5–7]. We see no evidence of any dependence of these exponents on D. This result is surprising. One can expect a D dependence simply because $\alpha_1=1/3$ when $D=2$ and the DSI is restored [9,10]. Notice, however, significant corrections to scaling (that is, curvature on the log-log plot) in $l_1(t)$ and $P(t)$. This effect (also observed for DLA clusters [5–7]) can mask possible weak D dependence of α_1 in the range of $1.5<D<1.7$. The exponent β is close to the classical exponent 1/3. The appearance of the classical exponent in a situation with broken DSI requires an explanation (see below). The solute mass exhibits dynamic scaling with a D-dependent exponent γ.

Now we report a simple scaling theory for the exponents β and γ. It is based on the fact that, at subdiffusive distances, $r<l_d(t)$, the Cahn-Hilliard dynamics are reducible to a sharp-interface model of Laplacian coarsening [9,17]. The process of Laplacian coarsening can be described as follows. Small branches of the cluster shrink and disappear, and their material is reabsorbed by larger branches. Because of the Laplacian screening, reabsorption occurs locally (therefore,

TABLE I. Dynamic exponents found in the simulations. Also shown is γ_{th}, theoretical prediction for γ.

Series	*A*	*B*	*C*
D	1.7	1.7	1.5
α_1	0.21 ± 0.01[a]	0.22 ± 0.01	0.21 ± 0.01
α_2	-0.21 ± 0.01	-0.21 ± 0.01	-0.21 ± 0.01
β	0.32 ± 0.01	0.32 ± 0.01	0.30 ± 0.01
γ	-0.16 ± 0.005	-0.16 ± 0.005	$-0.09\pm2\times10^{-4}$
γ_{th}	-0.18	-0.18	-0.09

[a]The errors are estimated by shifting the time intervals of fitting. The errors in each fit are much smaller.

the mass distribution on large scales remains unchanged). The reabsorption events cause undulations of the interfaces of the branches. Assuming DSI on length scales $l_1(t)<r<l_d(t)$, we see that, by time t, the characteristic undulation length is of order $t^{1/3}$. This gives a natural explanation to the dynamic length scale $l_2 \sim t^{1/3}$ observed in the simulations. The Gibbs-Thomson condition at the interface [9,17] implies a solute density of order $t^{-1/3}$ in the "contaminated" region within a distance l_d from the cluster. Taking into account the (preserved) fractal structure of the cluster at distances that are large compared to l_d, we estimate the contaminated area as $A_d \sim l_d^2 (L/l_d)^D \propto t^{(2-D)/2}$. Multiplying the "solute density" $t^{-1/3}$ by A_d yields the solute mass $M_s(t) \propto t^{(4-3D)/6}$. The last row in Table I shows the theoretical exponent $\gamma_{th}=(4-3D)/6$. A good agreement with simulations is seen. Notice that γ_{th} changes sign at $D=4/3$. At $D<4/3$ reabsorption should effectively stop, and the cluster should continue dissolving. We performed a single simulation with a random FC with $D=1.3$, which indeed showed a very slow but persistent increase of M_s with time. Notice, however, that FC with a smaller D also implies, in our simulations, a smaller area fraction of the minority phase. At too small area fractions the minority phase can dissolve completely, independently of D. Therefore, a more careful investigation is needed in order to distinguish between these two effects.

In summary, the simulations support earlier results [6] on breakdown of DSI in locally conserved coarsening of systems with long-ranged correlations and reveal new signatures of hidden simplicity in these systems. We confirm the value of the anomalous dynamic exponent $\alpha_1 \simeq 0.21$–0.22. At present we are unaware of any reliable prediction for the dependence of α_1 on D, except the constraint $\alpha_1(D=2)=1/3$. However, in view of our results, this dependence (if any) should be quite weak in the 1.5–1.7 range. Two additional dynamic exponents $\beta \simeq 1/3$ and $\gamma=\gamma(D)$ are identified. They can be interpreted in terms of Laplacian coarsening at subdiffusive distances $r<l_d(t)$, combined with DSI at intermediate distances $l_1(t)<r<l_d(t)$.

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