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 are independent of \( D \); 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 non-equilibrium 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 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 \([1,2]\). 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) \([4]\). 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 \([5,6]\). This example is non-trivial because of the long-ranged, power-law correlations intrinsic in FCs. The role of long-ranged correlation in phase ordering dynamics has been under debate in connection to dynamic scale invariance (DSI), a major simplifying factor in theory \([7]\). 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 \( g[r/l(t)] \). 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 \( 1/3 \).

Because of complexity of phase-ordering dynamics, DSI has not been proven, except for a very few simple models \([8]\). 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 \([9]\). DLAs with fractal dimension \( D = 1.75 \) served in Ref. \([10]\) 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 \( \sigma \) 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 \([7,8]\). 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 far-lying parts of the cluster is exponentially small in this case. The inequality \( l_d < L \) was satisfied in the simulations of Refs. \([7,8]\), 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 non-trivial 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 \( 0.21 - 0.22 \)) \([8,9]\). This implies hidden simplicity of a more complex nature than DSI.

We report phase-field simulations of locally-conserved coarsening of 3 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 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 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.

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 stucked together in each iteration [12]. The clusters obtained in this way are "reinforced", by an addition of peripheral sites as suggested in Ref. [4], 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 [3-5]

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

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 $\Omega$ was a two-dimensional box $1024 \times 1024$ with no-flux boundary conditions (zero normal component of $\nabla u$ 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 [3-5]. The simulation box was $1792 \times 1792$, with periodic boundary conditions. The grid size was $\Delta x = \Delta y = 1$ that corresponds to 1792 Fourier modes in each dimension. The time step was $\Delta t = 0.3$, the total time range was $0 < t < 15000$. Overall, three series of simulations were performed. In two of them, with the explicit scheme, 10 FCs with $D = 1.7$ (series A) and 10 FCs with $D = 1.5$ (series C) served as the initial conditions for the minority phase $u = 1$. In addition, 7 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.

Eq. (1) with either no-flux, or periodic boundary conditions obey conservation laws: $I_0 = \int \int u(r,t) \, dr = \text{const}$ and $I_1 = \int \int u(r,t) \, dr = \text{const}$. Our numerical schemes preserve $I_0$ exactly. We checked that they also preserve $I_1$ with a very high accuracy: better than $5 \times 10^{-3}$% 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".

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 $D = 1.5$.

The cluster was identified as the locus of $B(r,t) = 1$, where $B(r,t) = 1$ for $u(r,t) \geq 0$ and zero otherwise. Figures 1 and 2 show snapshots of the coarsening dynamics observed in series A and C. One can see that larger features grow at the expense of 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, therefore its absence clearly implies that DSI is broken. One can also notice a more pronounced breakup of the cluster for  

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  

2. Cluster perimeter  

3. Cluster mass  

4. “Solute” mass outside the cluster  

Here an auxiliary “density” field  

FIG. 3. Equal-time pair correlation function at different times for simulation series B:  

Figure  shows  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 breakdown of DSI at $D < 2$. The same type of behavior of  

As small distances,  behaves linearly with  

This asymptotics yields dynamic length scale  

in the course of time. This feature yields dynamic length scale  

We define  as the maximum value of  

Still, even in this case the large-scale mass distribution in the cluster does not change much. Notice that, by the end of the simulation time, the diffusion length  is already greater than $10^2$. Still, we do not see any signature of  in the shape of  

We expect that the tail of  

will remain “frozen” until late times, and DSI broken, in the Laplacian regime as well.

Figures  and  show the measured quantities  

averaged over the initial conditions, versus time for simulation series B and C, respectively. We fitted these time dependencies by power laws with exponents  

respectively. According to Porod law  

one expects  

All simulation results are summarized in Table 1.  

Remarkably, they do not depend on  

The exponent  

is close to the classical exponent  

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  

Now we report a simple scaling theory for the exponents  

It is based on the fact that, at subdiffusive distances,  

the Cahn-Hilliard dynamics are reducible to a sharp-interface model of Laplacian coarsening  

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 Laplacian screening, reabsorption occurs locally (therefore, the mass distribution on large scales remains unchanged). The reabsorption events cause undulations of the interfaces of the branches.

Assuming DSI on length scales  

we see that, by time  

the characteristic undulation length is of order  

This gives a natural explanation to the dynamic length scale  

observed in the simulations. The Gibbs-Thomson condition at the interface  

implies a solute density of order $t^{-1/3}$ in the “contaminated” region within a distance  

from the cluster. Taking into account the (preserved) fractal structure of the cluster at distances large compared to  

we estimate the contaminated area as  

Multiplying the “solute density” $t^{-1/3}$ by  

yields the solute mass:  

The last row in the table shows the theoretical exponent  

A good agreement with simulations is seen. Notice that  

changes sign at  

At $D < 4/3$ reabsorption should effectively stop, and the cluster should continue dissolving. We performed a single simulation with a random FC with $D = 1.3$ that indeed showed a very slow but persistent increase of  

with time. Notice, however, that FC with a smaller  

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.

The errors in each fit are much smaller. Therefore, a more careful investigation is needed in order to distinguish between these two effects.

In summary, the simulations support earlier results 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 $\alpha_1 \simeq 0.21 - 0.22$. Surprisingly, it does not show any dependence on $D$. 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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![FIG. 4](image)

**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](image)

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

| Series | $A$ (1.7) | $A$ (1.7) | $A$ (1.5) |
|--------|-----------|-----------|-----------|
| $D$    |           |           |           |
| $\alpha_1$ | 0.21 ± 0.01 | 0.22 ± 0.01 | 0.21 ± 0.01 |
| $\alpha_2$ | -0.21 ± 0.01 | -0.21 ± 0.01 | -0.21 ± 0.01 |
| $\beta$ | 0.32 ± 0.01 | 0.32 ± 0.01 | 0.30 ± 0.01 |
| $\gamma$ | -0.16 ± 0.005 | -0.16 ± 0.005 | -0.09 ± 2 × 10^{-4} |
| $\gamma_{th}$ | -0.18 | -0.18 | -0.09 |

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

[1] J.S. Langer, Rev. Mod. Phys. 52, 1 (1980); and in *Chance and Matter*, edited by J. Souletie, J. Vannimenus, and R. Stora (Elsevier, Amsterdam, 1987).
[2] D.A. Kessler, J. Koplik, and H. Levine, Adv. Phys. 37, 255 (1988); E.A. Brener and V.I. Mel’nikov, Adv. Phys. 40, 53 (1991).
[3] E. Brener, H. Müller-Krumbhaar and D. Temkin, Phys. Rev. 54, 2714 (1996).
[4] J. Feder, *Fractals* (Plenum, New York, 1988); T. Vicsek, *Fractal Growth Phenomena* (World Scientific, Singapore, 1992); P. Meakin, *Fractals, Scaling and Growth Far from Equilibrium* (Cambridge University Press, Cambridge, 1997).
[5] T. Irisawa, M. Uwaha and Y. Saito, Europhys. Lett. 30, 139 (1995).
[6] M. Conti, B. Meerson and P.V. Sasorov, Phys. Rev. Lett. 80, 4693 (1998).
[7] S.V. Kalinin et al., Phys. Rev. E 61, 1189 (2000).
[8] J.D. Gunton, M. San Miguel, and P.S. Sahni, in *Phase Transitions and Critical Phenomena*, edited by C. Domb and J.L. Lebowitz (Academic Press, New York, 1983), Vol. 8, p. 267; H. Furukawa, Adv. Phys. 6, 703 (1985); J. Langer, in *Solids Far From Equilibrium*, edited by C. Godrèche (Cambridge University Press, Cambridge, 1992).
[9] A.J. Bray, Adv. Phys. 43, 357 (1994); also in *Soft and Fragile Matter. Nonequilibrium Dynamics, Metastability and Flow*, edited by M.E. Cates and M.R. Evans (IOP, Bristol, 2000), p. 205.
[10] I.M. Lifshitz and V.V. Slyozov, J. Phys. Chem. Solids, 19, 35 (1961).
[11] A. Peleg, M. Conti and B. Meerson, Phys. Rev. E 64, 036127 (2001).
[12] R. Thouy and R. Jullien, J. Phys. A 27, 2953 (1994).
[13] J. Zhu et al., Phys. Rev. E 60, 3564 (1999).
[14] R. Sempère et al., Phys. Rev. Lett. 71, 3307 (1993).
[15] B. Meerson and P.V. Sasorov, e-print cond-mat/9708036.
[16] J.R. Parker, Practical Computer Vision Using C (Wiley, New York, 1993), p. 51.
[17] R.L. Pego, Proc. R. Soc. Lond. A 422, 261 (1989).