Dynamics of fractal dimension during phase ordering of a geometrical multifractal

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A simple multifractal coarsening model is suggested that can explain the observed dynamical behavior of the fractal dimension in a wide range of coarsening fractal systems. It is assumed that the minority phase (an ensemble of droplets) at $t=0$ represents a nonuniform recursive fractal set, and that this set is a geometrical multifractal characterized by an $f(\alpha)$ curve. It is assumed that the droplets shrink according to their size and preserve their ordering. It is shown that at early times the Hausdorff dimension does not change with time, whereas at late times its dynamics follow the $f(\alpha)$ curve. This is illustrated by a special case of a two-scale Cantor dust. The results are then generalized to a wider range of coarsening mechanisms.

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Fractal growth phenomena have been under extensive investigation during the past two decades [1–3]. The inverse process of fractal coarsening occurs in many physical systems. It has been discussed in the context of sintering of fractal matter [4]. Coarsening of fractal clusters by surface tension in bulk-diffusion-controlled [5,6], interface-controlled [7], and edge-diffusion-controlled [5,8] systems has been investigated. Additional examples include thermal relaxation of rough grain boundaries [9] and smoothing of fractal polymer structure in the process of polymer collapse [10]. Two-dimensional fractal fingering, observed in a Hele-Shaw cell with radial symmetry (for a review, see Ref. [11]), exhibits coarsening at a late stage of the experiment. All these systems are quite different, as they involve nonconserved or conserved order parameters, different transport mechanisms, etc.

A crucial issue related to any phase ordering process is the presence or absence of dynamical scale invariance (DSI) [12]. DSI assumes that there is a single dynamical length scale $\lambda(t)$ such that the coarsening system looks (statistically) invariant in time when lengths are scaled by $\lambda(t)$. Does a fractal cluster or a fractal interface exhibit DSI (on a shrinking interval of distances) in the process of coarsening? Early scenarios of fractal coarsening in systems with nonconserved [13] and conserved [4] order parameters did rely upon the hypothesis of DSI. However, numerical simulations showed that DSI breaks down during the coarsening of fractal clusters in edge- [8] and bulk-diffusion-controlled [6] systems. On the other hand, recent simulations of smoothing of a fractal polymer during collapse [10], and of interface-controlled fractal coarsening under a global conservation law [14], do support DSI. Therefore, a question arises about possible universality classes of fractal coarsening.

Even if DSI holds, the fractal dimension may or may not change with time. Early fractal coarsening scenarios [13,4] assumed that it remains constant (again, on a shrinking interval of distances). Experiments on sintering of silica aerogels (a convenient way of investigating fractal coarsening) have been inconclusive. Some of them [4] gave evidence in favor of constancy of the fractal dimension during coarsening, while others [15] reported a significant change of the fractal dimension with time. Other evidence for a significant decrease of fractal dimension with time was found in experiments on thermal annealing of ferroelectric thin films of lead zirconate titanate [16]. In this experiment, the fractal dimension remained constant at early times, and decreased to its final value at intermediate times. Numerical simulations of a variety of coarsening systems with different growth laws showed that the fractal dimension does not change with time. These simulations include bulk-diffusion-controlled [5,6], edge-diffusion-controlled [5,8], and interface-controlled [7,14] systems.

It is remarkable that in so many systems with widely different coarsening mechanisms the fractal dimension remains constant during the dynamics. Therefore, one is tempted to look for a general scenario that would explain this fact and that would be insensitive to specific coarsening mechanisms. The simple multifractal coarsening model developed in this paper has this property. In addition, this model is an attempt to address the multifractal properties of fractal coarsening.

We shall consider a very simple model of a coarsening fractal system. In this model, the initial condition for the minority phase is an ensemble of droplets that represents a *geometrical multifractal*. We will then assume that the smaller droplets shrink and disappear independently, according to their sizes, and consider discrete time dynamics. Using a well-known theorem of multifractal geometry, we will establish the dynamical behavior of the Hausdorff dimension of this simple coarsening system. This result will be illustrated in a special case, when the droplets are distributed in the form of a two-scale Cantor dust [1,2]. Employing the size distribution function of this fractal set [17], we will follow the dynamical behavior of the d measure in two characteristic limiting cases and show that the Hausdorff dimension's dynamics in this example are consistent with the general result. Then we will relax the discrete time assumption. Furthermore, we will show that the results are essentially independent of the details of the coarsening dynamics as long as the minority-phase droplets do not merge or break up.

The minority phase of our model represents, at zero time, a big but finite ensemble of droplets that form a nonuniform recursive fractal [2] with a constant density distribution in the E-dimensional space. Let us index the droplets in the mth

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generation of the fractal according to their radii. Thus, all the droplets with index k have radius $R_m(k)$ and form a subset of the whole fractal, which we denote by $S_m(k)$. The smallest droplets have index $k=0$ and radius $R_m(0)$, which is the lower cutoff of the fractal. The largest droplets have index $k=m$ and radius $R_m(m)$, which is the upper cutoff. One can work with a size distribution function $n_m(k)$, which is simply the number of droplets with radius $R_m(k)$, and use it to compute the Hausdorff dimension of the fractal (see Ref. [17], where this was done for a two-scale Cantor dust).

Any nonuniform recursive fractal with a constant density distribution can be described as multifractal in the geometrical sense (see Ref. [2], p. 66). In this case one can introduce the measure of the subset $S_m(k)$ in the following way:

$$\mu_m(k)=\frac{R_m^E(k)}{\Sigma_{k=0}^m n_m(k)R_m^E(k)}, \tag{1}$$

where $R_m(k)$ are the radii of the droplets divided by the size of the system. The Hölder exponent of the elements of the subset $S_m(k)$ is defined by [18]

$$\alpha_m(k)=\frac{\ln \mu_m(k)}{\ln R_m(k)}. \tag{2}$$

The $f(\alpha)$ curve for the fractal is constructed in the following way [1,2,19]:

$$f(\alpha)=-\frac{\ln n_m(k)}{\ln R_m(k)} \quad (1\ll k\ll m), \tag{3}$$

where k is supposed to be expressed through α with the help of the equation $\alpha_m(k)=\alpha$. (We assume that this equation gives a one-to-one correspondence between α and k.) $f(\alpha)$ is assumed to have a single maximum that is attained for $\alpha=\alpha_0$, so that $f(\alpha_0)$ is the Hausdorff dimension of the whole fractal. We also assume that $f(\alpha(k))$ is the Hausdorff dimension of the subset $S_m(k)$. This assumption, widely used in the physical literature, was rigorously proved in the case of a two-scale Cantor dust [20], and also for a class of other multifractal measures [21].

We now turn to describe the dynamics. We assume first that the droplets shrink and disappear independently, according to their radius only, and also simplify the governing dynamics by introducing a discrete time τ (later we will relax these two assumptions). In the first time step $\tau=0$ the smallest droplets with radius $R_m(0)$ disappear, *while the sizes of the other droplets do not change*. In the next time step $\tau=1$ the elements with radius $R_m(1)$ disappear, and so on. The set of droplets that survive after each step of these dynamics obviously remains self-similar (on a shrinking interval of distances). The main result of this paper is the following behavior of the Hausdorff dimension D as a function of the discrete time τ. For $\tau\leqslant k(\alpha_0)$ D does not change: $D(\tau)=D_0$, where D_0 is the Hausdorff dimension of the initial condition. For $\tau>k(\alpha_0)$ $D(\tau)=f(\alpha(k_{min}))$ where $k_{min}(\tau)$ is the k value of the smallest droplets that have not yet disappeared by time τ. This dynamical behavior is illustrated in Fig. 1.

The proof of this result is based on the following theorem: *the Hausdorff dimension of a union of two fractal sets S_1 and S_2 with fractal dimensions D_{S_1} and D_{S_2}, respectively, is* $D=max(D_{S_1},D_{S_2})$. (See, for example, [2], p. 17.) In the last time step of the dynamics, $\tau=m$, the coarsening object consists of the subset $S_m(m)$ alone, and its Hausdorff dimension is $f(\alpha(m))$. In the previous time step $\tau=m-1$ the object consists of two subsets: $S_m(m)$ with Hausdorff dimension $f(\alpha(m))$, and $S_m(m-1)$ with Hausdorff dimension $f(\alpha(m-1))$. It follows from the shape of the $f(\alpha)$ curve of the initial fractal that $f(\alpha(m-1))>f(\alpha(m))$. Using the theorem, we get $D(\tau=m-1)=f(\alpha(m-1))$.

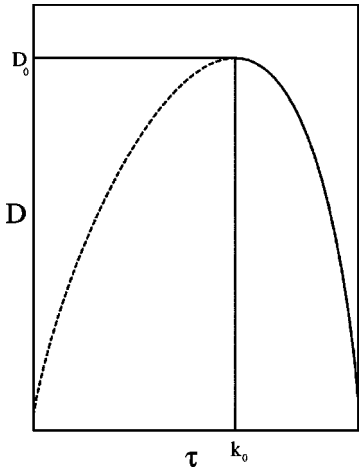

FIG. 1. Hausdorff dimension D of the ensemble of droplets versus discrete time τ (solid curve). The dashed curve is the $f(\alpha(k))$ curve at $\tau=0$. D_0 is the Hausdorff dimension at $\tau=0$, while $k_0\equiv k(\alpha_0)$ is the value of k for which $f(\alpha(k))$ has its maximum.

More generally, consider time $\tau=k_0+s$, where s is a positive integer and $k_0\equiv k(\alpha_0)$. At this time we can regard the object as a union of two fractal subsets $S_m(k_0+s)$ and $S_m(m\geqslant k\geqslant k_0+s+1)$. Here, $S_m(m\geqslant k\geqslant k_0+s+1)$ is the union of all subsets $S_m(k)$ with $k=s+1,\ldots,m$. It is also the whole coarsening object at time $\tau=k_0+s+1$. Assume by induction that $D(\tau=k_0+s+1)=D(S_m(m\geqslant k\geqslant k_0+s+1))=f(\alpha(k_0+s+1))$. It follows from the shape of the $f(\alpha)$ curve that $f(\alpha(k_0+s))>f(\alpha(k_0+s+1))$. Hence, using the theorem, we conclude that $D(\tau=k_0+s)=f(\alpha(k_0+s))$. Since k_0+s is the index of the smallest droplets that have not yet disappeared, we can write this result as

$$D(\tau\geqslant k_0)=f(\alpha(k_{min})). \tag{4}$$

The dynamical behavior of the Hausdorff dimension at times $\tau\leqslant k_0$ can be found in a similar way. For $\tau=k_0-1$ the object can be considered as a union of two fractal subsets $S_m(m\geqslant k\geqslant k_0)$ and $S_m(k_0-1)$. It follows from Eq. (4) that $D(\tau=k_0)=D(S_m(m\geqslant k\geqslant k_0))=D_0$. From the shape of the $f(\alpha)$ curve we get $D_0=f(\alpha(k_0))>f(\alpha(k_0-1))$. Therefore, $D(\tau=k_0-1)=D_0$. More generally, for any time $\tau=k_0-s$ the coarsening object can be considered as a union of the two fractal subsets $S_m(m\geqslant k\geqslant k_0-s+1)$ with Hausdorff dimension D_0 and $S_m(k_0-s)$ with Hausdorff dimension $f(\alpha(k_0-s))$. From the shape of the $f(\alpha)$ curve we deduce $f(\alpha(k_0-s))<D_0$. Hence, by using the above theorem, we conclude that $D(\tau=k_0-s)=D_0$. More generally, we can write:

$$D(\tau \ll k_0) = f(\alpha(k_0)) = D_0. \tag{5}$$

Let us now turn to the particular case when the ensemble of droplets at zero time represents a two-scale Cantor dust. Recall that the initiator of this fractal is an E-dimensional cube of unit side length. The generator consists of n_1 cubes of side l_1 and n_2 cubes of side l_2 where $l_2 > l_1$. In each step of the fractal construction every full cube is replaced by the properly rescaled generator. After the last step of the construction, which is the mth step, all the cubes are replaced by spherical droplets with the same size as the cubes.

Now assume that this two-scale Cantor dust undergoes the simple coarsening dynamics described earlier. For convenience, we will compute the time-dependent d measure of a two-scale Cantor dust that consists of cubes (the ones that were replaced by the spheres after the mth generation of the construction). The only difference in the computed d measure will be in a d-dependent prefactor. Since this prefactor is independent of k and m, it will not affect the dynamical behavior of the d measure and the Hausdorff dimension.

The d measure of the mth generation of a two-scale Cantor dust can be written as [17]

$$M_d = \int_0^m n_m(k) R_m^d(k)\, dk = \left(\frac{m}{2\pi k(m-k)}\right)^{1/2} \int_0^m \exp[g(k)]\, dk, \tag{6}$$

where

$$g(k) = -k \ln\left(\frac{k}{m n_2 l_2^d}\right) - (m-k)\ln\left(\frac{m-k}{m n_1 l_1^d}\right), \tag{7}$$

and $R_m(k) = l_1^{m-k} l_2^k$ is the size of the cubes in the subset $S_m(k)$. The function $\exp[g(k)]$ has a (sharp) maximum at

$$\tilde{k}_0(d) = \frac{n_2 l_2^d m}{n_1 l_1^d + n_2 l_2^d}. \tag{8}$$

For $d = D_0$ one can show that $\tilde{k}_0(D_0) = k(\alpha_0) \equiv k_0$. At time $\tau = k_{min}$ the d measure of the object is

$$M_d(\tau) = \int_{k_{min}(\tau)}^m n_m(k) R_m^d(k)\, dk. \tag{9}$$

As long as $k_{min}(\tau) \ll \tilde{k}_0(d)$, one can apply the saddle point argument used in Ref. [17] and conclude that

$$M_d(\tau \ll \tilde{k}_0(d)) \simeq M_d(\tau = 0) = (n_1 l_1^d + n_2 l_2^d)^m. \tag{10}$$

This implies that during the early stages of the dynamics the d measure remains, with an exponential accuracy, constant. Correspondingly, the Hausdorff dimension, which is computed by solving the same equation

$$n_1 l_1^d + n_2 l_2^d = 1 \tag{11}$$

for d, does not change with time. This is in agreement with Eq. (5) obtained in the general case.

On the other hand, when $\tilde{k}_0(d) \ll \tau = k_{min} \ll m$, the behavior of $M_d(\tau)$ is quite different. Since for $k > \tilde{k}_0(d)$ $g(k)$ is a decreasing function of k, the main contribution to the integral in Eq. (9) comes from a close neighborhood of $k = k_{min}(\tau)$. Therefore, in Eq. (9) we can expand $g(k)$ around $k = k_{min}(\tau)$ to the first order and get

$$M_d \simeq \frac{n_m(k_{min}) R_m^d(k_{min})}{|g'(k_{min})|} = \frac{h(\xi_{min}, d)[y(\xi_{min}, d)]^m}{m^{1/2}}, \tag{12}$$

where $\xi_{min} = k_{min}/m$,

$$y(\xi_{min}, d) = \left(\frac{1-\xi_{min}}{n_1 l_1^d}\right)^{\xi_{min}-1} \left(\frac{\xi_{min}}{n_2 l_2^d}\right)^{-\xi_{min}}, \tag{13}$$

and

$$h^{-1}(\xi_{min}, d) = [2\pi \xi_{min}(1-\xi_{min})]^{1/2} \left| \ln\left(\frac{(1-\xi_{min}) n_2 l_2^d}{\xi_{min} n_1 l_1^d}\right)\right|. \tag{14}$$

The Hausdorff dimension of the subset labeled by ξ_{min} is given by

$$f(\alpha(\xi_{min})) = \frac{\xi_{min}\ln(\xi_{min}/n_2) + (1-\xi_{min})\ln[(1-\xi_{min})/n_1]}{(1-\xi_{min})\ln l_1 + \xi_{min}\ln l_2}. \tag{15}$$

It follows that

$$R_m(k_{min})^{-f(\alpha(k_{min}))} = \frac{n_m(k_{min})}{[m/(2\pi k_{min}(m-k_{min}))]^{1/2}}. \tag{16}$$

Hence, we obtain the following expression for M_d in the limit of $\tilde{k}_0(d) \ll \tau \ll m$:

$$M_d \simeq \left(\frac{h(\xi_{min}, d)}{m^{1/2}}\right) R_m(k_{min})^{d - f(\alpha(k_{min}))}. \tag{17}$$

We see that, up to logarithmic corrections resulting from the factor $h(\xi_{min}, d)$, the d measure obeys a power law of $R_m(k_{min})$ with a time-dependent exponent.

Equations (12)–(14) allow one to calculate the Hausdorff dimension of the ensemble of droplets in the limit of $\tilde{k}_0(d) \ll \tau \ll m$. Taking the logarithm of both sides of Eq. (12) and dividing by m, we get

$$\frac{\ln M_d}{m} \simeq \frac{1}{m}\ln\left(\frac{h(\xi_{min}, d)}{m^{1/2}}\right) + \ln[y(\xi_{min}, d)]. \tag{18}$$

As $m \gg 1$, the first term on the right hand side of Eq. (18) can be neglected. Therefore, the Hausdorff dimension is determined by solving the equation

$$y(\xi_{min},d)=1 \tag{19}$$

for d. The solution is just the Hausdorff dimension of the subset ξ_{min} given by Eq. (15). Therefore, $D(\tau)=f(\alpha(k_{min}))$ for $k_0 \ll \tau \ll m$, in agreement with the general result (4).

We now show that the assumptions of a discrete time and of the independent shrinking of the droplets can be relaxed. It is sufficient to assume only that the dynamics of each droplet are determined by its radius (and possibly by a time-dependent ''critical radius,'' characterizing some mean-field interaction between droplets). We should also assume that the droplets do not merge or break up. Under these assumptions the number of droplets in each subset is constant (until the droplets disappear) and all the droplets belonging to the same subset have the same (time-dependent) radius. In addition, we forbid nucleation, which is a standard assumption for a coarsening stage [12].

Let us denote the radii of the droplets belonging to the kth subset at time t by $R_m(k,t)$. The d measure of the kth subset at time t is given by

$$M_d(m,k,t)=n_m(k)R_m^d(k,t). \tag{20}$$

This can be rewritten as

$$M_d(m,k,t)=M_d(m,k,0)\left(\frac{R_m(k,t)}{R_m(k,0)}\right)^d, \tag{21}$$

where $R_m(k,0)$ and $M_d(m,k,0)$ are the initial values of the radii and d measure. Since the initial condition is a geometrical multifractal, $M_d(m,k,0)$ can be expressed in the following manner:

$$M_d(m,k,0)=\left[Y\left(\frac{k}{m},d,\{P_i\}\right)\right]^m, \tag{22}$$

where the function Y and the parameters $\{P_i\}$ characterize the initial fractal condition considered. (In our example of the two-scale Cantor dust the role of the function Y was played by y, while the set of parameters $\{P_i\}$ included n_1, n_2, l_1, and l_2.) Substituting Eq. (22) into Eq. (21), taking the logarithm of both sides, and dividing by m we get

$$\frac{\ln M_d(m,k,t)}{m}=\ln\left[Y\left(\frac{k}{m},d,\{P_i\}\right)\right]+\frac{d}{m}\ln\left(\frac{R_m(k,t)}{R_m(k,0)}\right). \tag{23}$$

For typical coarsening mechanisms $R_m(k,t)$ grows with time more slowly than exponentially. For example, this is true for nonconserved dynamics (model A) and for the Lifshitz-Slyozov theory of conserved dynamics (model B) [12]. Therefore, when $R_m(k,t) > R_m(k,0)$ the second term on the right side of Eq. (23) is negligible at $m \gg 1$. Similarly, it is negligible when $R_m(k,t) < R_m(k,0)$ as long as $R_m(k,t)$ is not exponentially smaller than $R_m(k,0)$. Equation (23) becomes inconvenient in the case of shrinking droplets at the moment of their disappearance. Equation (21) shows, however, that the d measure of such droplets vanishes. Hence, the d measure of the kth subset does not change during the coarsening dynamics until the droplets belonging to this subset disappear. Consequently, the Hausdorff dimension of this subset does not change until its disappearance. We have therefore shown that the results of our simple discrete time coarsening model apply to a wide range of coarsening mechanisms. It should be noted that for a system with weak multifractal properties our model predicts that the fractal dimension remains approximately constant at all times. Therefore, this model provides a simple explanation for the observation that the fractal dimension does not change in a wide range of coarsening processes [4–10].

In summary, we have considered a simple model of coarsening disconnected droplets forming a geometrical multifractal. We have shown that at early times the Hausdorff dimension of the system does not change, whereas at late times its dynamics follow the $f(\alpha)$ curve of the initial multifractal distribution. These results are insensitive to the particular coarsening mechanism. We hope that they will motivate experimental investigation of multifractal aspects of fractal coarsening.

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