CWI Quarterly Volume 10 (2) 1997, pp. 153 – 178

Aggregation Models of Fractal Growth

Mária Vicsek
Computer and Automation Institute of HAS, Budapest, P.O.B. 63, 1518 Hungary
e-mail: h630vic@ella.hu
Tamás Vicsek
Department of Atomic Physics, Eötvös University, Budapest, Puskin u 5-7, 1088 Hungary
e-mail: h845vic@ella.hu

Recently many growth processes have been shown to result in fractal structures. An important method which can be used to interpret such phenomena is the simulation of the aggregation of similar particles. Aggregation may take place particle by particle, while in other cases the aggregates themselves are also mobile and are joined together to form larger clusters during their motion. In this paper we discuss the basic types of aggregation models leading to fractals. The related results are described in terms of fractal dimension, correlation functions and dynamic scaling. It is demonstrated that by carrying out numerical simulations of the process of aggregation a deeper insight can be gained into the spatial and temporal features of fractal growth phenomena.

1. Introduction

Since the early eighties it has widely been recognized by scientists working in diverse areas that many of the structures common in their experiments possess a rather special kind of geometrical complexity. This awareness is largely due to the activities of Mandelbrot (1982), who called attention to the particular geometrical properties of such objects as the shore of continents, the branches of trees, or the surface of clouds. He coined the name *fractal* for these complex shapes to express that they can be characterized by a *non-integer* (fractal) *dimensionality*. With the development of research in this direction the list of examples of fractals has become very long, and includes structures from microscopic aggregates to the clusters of galaxies.

An important field where fractals are observed is that of far-from-equilibrium growth of interfaces which are common in many areas of science and technology. Examples for such processes include dendritic solidification in an undercooled medium, viscous fingering which is observed when a viscous

FIGURE 1. Example for a complex geometrical structure observed in experiments on a growing unstable interface during electrodeposition.

fluid is injected into a more viscous one, and electrodeposition of ions onto an electrode. Figure 1 demonstrates the complexity of a typical pattern observed in an electrodeposition experiment. In the experiments leading to fractal structures commonly quasi two-dimensional samples are used and the motion of the interfaces is determined by the spatial distribution of a quantity which satisfies the Laplace equation with moving boundary conditions.

In addition to interfacial growth, *aggregation* of similar particles represents another important class of growth phenomena producing complicated geometrical objects. Aggregation may take place particle by particle, while in other cases (for example during the formation of aerogels) the aggregates themselves are also mobile and are joined together to form larger clusters during their motion.

In this paper we shall discuss computer models of aggregation leading to fractals. In most of the cases the structure of the aggregates will be described in terms of the number of particles N in a given region of the fractal aggregate.

A broad class of growing patterns is characterized by a branching fractal structure as is illustrated by Figure 1. Such pictures of real experimental results can also be represented as aggregates by assuming that each pixel of their digitized image is a particle. In the present case the fractality of the structure means that it is *self-similar* (invariant under isotropic dilation) in a statistical sense and the number $N(R)$ of particles within a circle (hypersphere) of radius R scales with R in a non-trivial way

$$N(R) \sim R^D.$$

Here $D < d$ is typically a non-integer number called the *fractal dimension* and d is the Euclidian dimension of the space the fractal is embedded in. Naturally, for a real object the above scaling holds only for length scales between a lower and an upper cutoff.

There are a number of reasons for the recent rapid development in the research of fractal growth. The interest is greatly motivated by the fact that fractal growth phenomena are closely related to many processes of practical importance. Here we shall mention only two examples. The internal texture of alloys due to the dendritic structures developing during their solidification is largely responsible for most of their mechanical properties. Another area of application is secondary oil recovery, where water pumped into the ground through one well is used to force the oil to flow to the neighbouring wells. The effectiveness of this method is influenced by the fractal structure of viscous fingers corresponding to the water-oil interface.

During the past decade several reviews, books and conference proceedings have been published on topics related to fractal growth. For further reading, among others, the following resources can be useful: Vicsek et al (1994), Bunde and Havlin (1994), Barabási and Stanley (1995) and Evertsz et al (1996).

2. Basic principles of simulating aggregation

Because of the complexity of growth phenomena it is usually a difficult task to decide which of the factors affecting the growth play a relevant role in determining the structure of the growing object. In a real system the number of such factors can be relatively large, and this number is decreased to a few by choosing an appropriate model. The investigation of such models helps to detect the most relevant factors, and to demonstrate their effects in the absence of any disturbance.

Structures consisting of connected particles are usually called *clusters* or *aggregates*. In most of the cases the growth will be assumed to take place on a *lattice* for computational convenience, and two particles are regarded as connected if they occupy nearest neighbour sites of the lattice. However, for

studying universality and related questions, *off-lattice* or further neighbour versions of clustering processes can also be investigated. A lattice site with a particle assigned to it is called occupied or filled. An important additional feature included into the majority of models to be described is stochasticity which is typical for growth phenomena.

In general, a stochastic cluster growth model may lead to all possible configurations which can be formed from a given number of particles. What makes these models differ from each other is the weight or probability $P_{N,i}$ associated with a given configuration i consisting of N units. $P_{N,i}$ can be different for the same configuration even in the same model, because generally it depends on the sequence, according to which the individual particles are added to the cluster.

There are two main types of cluster growth processes, depending on the global character of the rule which is used in the course of adding a particle (or a cluster of particles) to the growing cluster. *The rule will be called local if it depends only on the immediate environment* of the position where the new particle is to be added. In other words, when deciding whether to add a particle at site $\mathbf{x}$ only the status (filled or not) of the nearest or next nearest neighbours of this site is taken into account. On the contrary, *in non-local models the structure of the whole cluster* can affect the probability of adding a site at a given position.

In addition to the two types of rules there are two typical geometries considered in the simulations of fractal aggregation. In the first case the growth starts from a single particle and usually results in isotropic aggregates, while in the second geometry the initial configuration is assumed to be a hyperplane (chain, a two dimensional lattice, etc. of seed particles). If the growth starts along a plane, the object we investigate is the advancing and gradually roughening surface of the growing structure. This surface is usually anisotropic and can be described in terms of self-affine fractals (see below).

3. Spreading percolation

In this section we shall consider a model representing perhaps the simplest growth process leading to a branching fractal structure. The process starts with a single seed particle placed onto a site of a lattice. Its neighbouring sites are considered live in the sense that they potentially may become occupied in the future. Next, one of these live sites is chosen randomly and (i) filled with a particle with a probability p or (ii) killed for the rest of time with a probability $1-p$. Occupation of a site with probability p is realized by generating a random number r (with a uniform distribution on $[0,1]$) and filling the site if $r < p$. The filled site becomes part of the growing cluster and its new neighbours become living sites. A large cluster is grown by repeating the same procedure many times, see Figure 2. Variation of this model at each time step all of the living or *growth sites* are considered for occupation, instead of one at a time, therefore, the cluster grows by adding shells to it.

FIGURE 2. Percolation cluster

The above process is relevant to a number of spreading phenomena, including epidemics, chemical reactions, flame propagation, and so on. For example, using the language of epidemics, the live sites are susceptible to infection, the killed sites are immune, while the occupied sites correspond to infected individuals. An epidemics will spread over the whole population if there is always at least one live site.

Note, that although the random numbers r are generated during the growth, the same configurations are obtained as if we had assigned to all of the sites of the lattice a random number previously, then defined equilibrium percolation clusters as connected objects consisting of sites for which $r < p$ and started the process afterwards. Therefore, the above model is equivalent to a simple type of growth on a static percolation cluster and as the available sites of a given configuration are filled we gradually recover an equilibrium percolation cluster. The growth stops when all sites belonging to the cluster containing the seed particle are filled.

Equilibrium percolation is a widely used model for describing various properties of inhomogeneous media (STAUFFER, 1985). Here we only recall those results of percolation theory which are related to the fractal nature of percolation clusters. In particular, when p is increased, at $p = p_c$ a transition takes place which is manifested in the appearance of a connected infinite cluster having a density for $0 < p - p_c \ll 1$

$$P(p) \sim (p - p_c)^{\beta}, \tag{1}$$

where $\beta > 0$ is the critical exponent of the percolation probability, $P(p)$, and p_c is called percolation threshold. In the following we consider the properties of the infinite cluster. The correlation length diverges at p_c according to

$$\xi \sim |p - p_c|^{-\nu_p} \tag{2}$$

where ξ corresponds to the length scale at which the fractal nature of the cluster crosses over into a homogeneous behaviour. We can obtain D from the following simple argument. The number of particles belonging to the cluster within a region of size ξ can be obtained both from $N(\xi) \sim \xi^D$ and from the expression $P(p)\xi^d$ (density times volume). Then from equating these two expressions and taking into account (1) and (2) we obtain

$$D = d - \frac{\beta}{\nu_p}.$$

The exponents β and ν_p are known exactly for $d = 2$ and can be calculated by numerical or theoretical methods for higher dimensions. For example, in two dimensions $\nu_p = \frac{4}{3}$ and $\beta = \frac{5}{36}$ gives for the fractal dimension of the infinite cluster at p_c the value $D = \frac{91}{48} \simeq 1.896$.

In some cases the order according to which the addition of a new particle takes place may play a relevant role. **Invasion percolation** was proposed to account for this type of behaviour. The invasion percolation model is defined on a lattice.

i) A random number drawn from the uniform distribution on the unit interval is assigned to each site of a cell of linear size L.
ii) As in the growing percolation model the process starts with a seed particle or a surface and goes on by subsequent occupation of one of the perimeter sites (empty sites which are nearest neighbours of the cluster).
iii) However, the perimeter site to be occupied is not selected randomly, but the one with the smallest random number r is occupied.

In this version of the invasion percolation model (which simulates the motion of a fluid-fluid interface in a random medium) the process does not stop until the finite cell is filled in completely, since we do not have a temperature like parameter analogous to the occupation probability p of ordinary percolation. On the other hand, as a well defined configuration one can study the structure of the cluster at the point in time when it first percolates, i.e., first forms a connected path between the two opposite edges of the cell. According to the simulations (Wilkinson and Willemsen 1983) the number of sites occupied by the invader at this moment can be expressed as $N \sim L^D$, where $D \simeq 1.89$ in two and $D \simeq 2.52$ in three dimensions. These values for the fractal dimension of the invasion percolation clusters are in good agreement with those obtained for the ordinary percolation clusters, showing the similarity between the static properties of the two models.

4. Diffusion-limited aggregation (DLA)

Many of the growth processes in nature are governed by the spatial distribution of a field-like quantity which is inherently **non-local**, i.e., the value of this quantity at a given point in space is influenced by distant points of the system, in addition to its immediate neighbourhood. For example, such behaviour is exhibited by the distribution of temperature during solidification, the probability of finding a diffusing particle or cluster at a given point, and electric potential around a charged conductor.

The spatial dependence of these quantities in various approximations satisfies the *Laplace equation with moving boundary conditions.* Since the concentration of diffusing particles is also described by the Laplace equation, the above mentioned class of processes is commonly called diffusion-limited growth. Diffusion-limited motion of interfaces typically leads to very complex, branching fractal objects, because of the unstable nature of growth. Thus, as a result of a self-organizing mechanism governed by the Laplace equation, structures with a rich geometry can emerge from the originally homogeneous, structureless medium. This far-from equilibrium phenomenon can be studied by approaches based on aggregation.

It is the non-local character of the probability distribution which plays an essential role in aggregation phenomena, where single particles, or clusters of particles are added to a growing aggregate. The main assumption of the related cluster models is that the particles stick together irreversibly, a condition which is satisfied in a wide variety of growth processes.

Consider an electrolite containing positive metallic ions in a small concentration, and a negative electrode. Whenever a randomly diffusing ion hits the electrode or the already deposited metal on its surface, it stops moving (sticks to the surface rigidly) because of the electrostatic attraction. This experiment results in a complicated, tree-like deposit with scale-invariant structure.

The model called **diffusion-limited aggregation** (DLA) was introduced by Witten and Sander (1981) to simulate in a computer phenomena related to the above mentioned process. The rules of the model are simple: One puts a seed particle at the origin of a lattice. Another particle is launched far from the origin and is allowed to walk at random (diffuse) until it arrives at a site adjacent to the seed particle. Then it is stopped, and another particle is launched which stops when adjacent to the two occupied sites, and so forth. In this way large clusters can be generated whose structure is expected to be characteristic for objects grown under diffusion-limited conditions.

Figure 3 shows several important features. First, the structure of these DLA clusters is remarkably similar to that of the experimental picture shown in Figure 1. Furthermore, Figure 3 demonstrates that these objects i) have a randomly branching, open structure, ii) look stochastically self-similar. By stochastic self-similarity here we mean the following: shrinking a large branch and omitting the finest details one obtains a structure which has the same appearance as a smaller branch. The branching, open structure is a result of "screening". In the case of DLA screening is manifested through the fact

FIGURE 3. Two typical DLA clusters. The top aggregate was grown on a square lattice and contains fifty thousand particles. For comparison a much larger off-lattice cluster (see the text) is also shown. This aggregate (bottom) contains six million particles and is scaled down to the size of the other cluster.

that the tips of the most advanced branches capture the incoming diffusing particles most effectively (particles have very small probability to penetrate into the already spontaneously formed fjords). Thus, small fluctuations are enhanced, and this instability together with the randomness inherent in the model leads to a complex behaviour.

To generate very large DLA clusters one has to use tricks because the original version of the model would require prohibitively large amounts of computer time. In the following, three simple procedures will briefly be described which are helpful in reducing the computational time substantially and allow one to grow aggregates consisting of several millions of particles.

i) The first trick is that we release the particles from a circle of radius R_0 which is just a bit larger than the largest distance between the particles already belonging to the aggregate and the origin. This method is justified

because the particles released very far from the cluster arrive (for the first time) at different points of a circle of radius larger than R_0 and centred at the origin with the same probability.

ii) Whenever a randomly walking particle leaves the region which is inside the above circle, the distance ΔR of the particle from the circle is determined. Then the next step made by the particle is a jump of length ΔR from its actual position made in a randomly selected direction (the final position is the lattice site closest to the coordinates of the particle after the jump has taken place). Inside the circle of radius R_0 the particle always undergoes a random walk jumping one lattice unit choosing one of the nearest neighbour sites randomly. If the particle making large jumps outside of the circle gets too far (e.g. ten times R_0), it is killed and a new particle is released from the circle R_0.

iii) A further improvement can be achieved using the following algorithm. At the beginning one assigns a quite arbitrary value $l' = l_{max}$ to all lattice sites. (A reasonable choice is in the range of 15-30.) Then, for each site from which the random walker could reach the seed (cluster) by making a jump of length l the value assigned to the site is changed to $l' = l$ if $l < l_{max}$. In other words, close to the cluster l' is the maximum step length a random walker may take without crossing the arms of the aggregate. Thus the particles jump a distance which is either l' or ΔR (the latter choice is made if $\Delta R > l_{max}$). After a new particle has been added to the cluster the l' values in the region where it landed are updated so that the next particle could not make a jump which would result in crossing any parts of the aggregate.

Extensive simulations have been carried out to determine the fractal dimension of DLA clusters. The results for $2 \leq d \leq 6$ are summarized in Table 1. Several conclusions can be made from this Table. First, it appears that for all d considered, the following inequalities hold

$$d - 1 < D < d.$$

These values are in good accord with the heuristic prediction

$$D = (d^2 + 1)/(d + 1).$$

The radially spreading and randomly branching structure of DLA clusters can also be quantitatively characterised by using the so called Wavelet Transform (WT). Let us imagine that we have a measure μ along a line. Its WT according to the analyzing wavelet ϕ is defined as

$$W_\phi[\mu](b, a) = \int_A \phi\left(\frac{x - a}{a}\right) d\mu,$$

where a is the scale and b is the space parameter. Usually ϕ is chosen to have vanishing moments up to a certain order. A WT realizes a kind of smoothing;

TABLE 1. The fractal dimension (D) of DLA clusters grown on $2 \leq d \leq 6$ dimensional hypercubic lattices. The prediction $D = (d^2+1)/(d+1)$ is also shown for comparison.

d	D	$(d^2+1)/(d+1)$
2	1.70 ± 0.06	1.667
3	2.53 ± 0.06	2.500
4	3.31 ± 0.10	3.400
5	4.20 ± 0.16	4.333
6	5.3	5.286

in some way the resulting transform looks like a fuzzy picture of the original structure, hence enhancing the main features of the underlying structure on a given scale. ARNEODO et al (1993) have applied this technique to uncover the multiplicative nature of the branching structure of DLA. By using the smoothing Gaussian function $\phi = \exp(-x^2/2)$ they have been able to show that the skeleton of the WT obtained for quasi one-dimensional azimuthal sections of DLA clusters (where μ was associated with the density of the cluster) exhibited a Cantor set type hierarchical structure. From determining the number of WT modulus maxima it was concluded that Fibonacci numbers can be used for describing the branching process during diffusion-limited aggregation.

In addition to its dependence on d, the fractal dimension may be affected by other factors. It is well known from the theory of critical phenomena that the exponents describing the singular behaviour of quantities at a second order phase transition are not changed under the influence of irrelevant parameters such as anisotropy, further neighbour interactions, type of lattice, etc. This property of the exponents, called *universality*, is of special importance. The question of universality of the fractal dimension has been addressed in the context of DLA as well, by investigating modifications of the original model.

As a first approximation to this problem the following versions of diffusion-limited aggregation were considered

i) DLA with *sticking probability* less than 1. In this variation the particles stick to the surface with a probability p_s, and continue to diffuse with a probability $1 - p_s$.
ii) DLA with *next-to-nearest neighbour* interaction. In this version the particles stop moving as soon as they arrive at a site which is next nearest neighbour to the aggregate.
iii) *Off-lattice* DLA (Figure 3, bottom). During the simulations of this variant the centre of a diffusing spherical particle is moved with the same probability to any point within a distance equal to the diameter of the particles. If a particle is found to overlap with another one, the particle is moved back to the position where it first touched the cluster and is incorporated into the aggregate.

FIGURE 4. Clusters consisting of 400 particles generated on the square lattice using the noise-reduced diffusion-limited aggregation algorithm. (a) m=2, random fractal; (b) m=20, dendritic growth, and (c) m=400, noisy needle crystal.

The results have indicated that below approximately 50000 particles the modifications considered in i)-iii) are irrelevant from the point of view of fractal dimension.

However, changes in the basic rules interfering with the instability of the growth process may alter the patterns radically. In particular, controlling the amount of randomness during the growth is likely to enhance the effect of the anisotropy of the underlying lattice and is expected to lead to less stochastic structures.

There is a natural way to decrease the fluctuations in DLA (KERTÉSZ and VICSEK 1986). Instead of adding a particle to the aggregate immediately after it hits a growth site, one keeps a record of how many times each of the perimeter sites (empty sites adjacent to the cluster) becomes a termination point for a randomly walking particle. After a perimeter site has been contacted m times it is filled and the new perimeter sites are identified. The scores (number of contacts) associated with these sites are set to zero. The scores associated with all of the other surface sites remain at their values before this event. Clearly, this procedure decreases the noise with growing m, because probing the surface with many walks provides a better estimate of the expectation value of the growth rate at the given point than a single walk.

Figure 4 shows three representative clusters generated on a square lattice with various values of the noise reduction parameter m. The overall appearance of the aggregates indicates that as a function of decreasing noise (increasing m) two types of morphological changes occur in these small scale simulations. At about $m = 5$ the random, tip-splitting structure typical for DLA clusters

crosses over into a dendritic pattern with well defined but irregularly spaced side branches having stable tips. Further increasing m results in the growth of a structure consisting of four needles growing out from the centre along the lattice axes.

The growth of a DLA cluster is governed by the distribution of the quantity $p(\mathbf{r}_j)$, where $p(\mathbf{r}_j)$ is the probability that the next growth event takes place at the site being at $\mathbf{r}_j$, adjacent to the cluster. This *growth-site probability distribution* (GSPD) is a very complex function changing rapidly in space due to screening. Let us imagine that we proceed along the surface of an aggregate and we record $p(\mathbf{r}_j)$ as a function of the arc length. Whenever we approach a tip in the outer region of the cluster, the growth probability associated with the actual position sharply increases since an advanced tip captures the diffusing particles with a large probability. Leaving this region one may get into a deep fjord which is almost completely screened by the surrounding branches, here $p(\mathbf{r}_j)$ is practically equal to zero. Getting close to another tip the growth probability becomes much larger again.

In general, the exponent describing the singular increase of $p(\mathbf{r}_j)$ depends on the local configuration close to a given tip. Therefore, it is quite natural to look at GSPD as a **fractal measure** or **multifractal** with infinitely many types of singularities (VICSEK 1992). Probing the surface with many random walks is equivalent to the estimation of the corresponding solutions of Laplace's equation which are usually called harmonic functions. Consequently, the name harmonic measure is also used for GSPD.

In addition to describing the distribution of growth probabilities, the harmonic measure is relevant to the physical properties of a fractal. For example, $p(\mathbf{r}_j)$ is proportional to the local charge density on a DLA, assuming that the aggregate is a charged electrical conductor.

To calculate the harmonic measure and its characteristic properties one can do the following. After having generated a DLA cluster one releases further particles whose diffusional motion is simulated by the same technique which is used for growing the aggregate. These probe particles, however, are eliminated when they arrive at the surface, and a record is kept of how many times each of the surface sites is contacted in this way. The normalized number of contacts is then regarded as the growth probability. More complete data can be obtained by solving the Laplace equation $\nabla^2\phi = 0$ with the boundary conditions $\phi = Const$ on the cluster and $\phi = 0$ far from it. Then the growth probabilities are given by $p(\mathbf{r}_j) \sim |\nabla\phi(\mathbf{r}_j)|$ on the basis of the electrostatic analogue.

After having determined the set of $p(\mathbf{r}_j)$ values, the generalized dimensions D_q and the $f(\alpha)$ spectrum of fractal dimensions corresponding to the singularities of strength α can be calculated using the standard expressions for these quantities (see, e.g., the paper by Struzik in this issue). For this purpose one has to cover the cluster with boxes of size l and sum up the $p(\mathbf{r}_j)$ values within the ith box to obtain the accumulated probability p_i associated with it. Then the exponent describing the scaling of the qth moment of the harmonic measure is given by

$$D_q = \lim_{l/L \to 0} \frac{1}{q-1} \frac{\ln \sum_i [p_i (l/L)]^q}{\ln (l/L)}, \tag{3}$$

were L is the linear size of the aggregate. To evaluate (3) one can either change l for a fixed cluster, or keep $l = 1$ and consider the growth probabilities for increasing L values. Note, that in principle both conditions $a/l \ll 1$ (where a is the lattice constant) and $l/L \ll 1$ should be satisfied during the calculations to produce results exactly corresponding to the multifractal spectrum as defined for finite fractals (with no lower cutoff length scale).

Figure 5 shows the results for two-dimensional off-lattice aggregates. These indicate that the so called $f(\alpha)$ spectrum of the growth probabilities behaves anomalously for DLA (no convergence to a well defined hump-shaped $f(\alpha)$ can be observed, scaling can be achieved only by using non-standard assumptions for the dependences). In an extensive subsequent study (SCHWARZER et al 1991) the authors considered clusters up to $N = 20,000$ particles and found that their results for the distribution of growth probabilities $n(\alpha, N)$ could be well described by the expression

$$\ln n(\alpha, N) \sim -\alpha^\gamma / \ln^\delta N,$$

where α denotes now $\alpha = -\ln p / \ln N$ differing from the above used definition only by a constant prefactor $1/D$, $\gamma = 2 \pm 0.3$ and $\delta = 1.3 \pm 0.3$. The validity of this expression is demonstrated by Figure 5, where $M = N$. Note that $\ln n(\alpha, N)$ is different from $f(\alpha)$ only by an additive constant and that the standard $f(\alpha)$ plot would not have the $\log_{10}^{1.3} M$ term in the denominator of the quantity plotted along the horizontal axis. Without this rescaling the $n(\alpha, N)$ (or $f(\alpha)$) curves would be gradually shifted to the left with a *less steep slope* when clusters consisting of increasing number of particles are evaluated. In the asymptotic limit this shift would result in a horizontal line corresponding to a one-sided $f(\alpha)$.

The obtained estimates can be examined using a few theoretical predictions. The value D_1 called information dimension has particular importance. It is equal to the fractal dimension of the set of boxes which give the dominant contribution to the first moment, i.e., to the sum of the box probabilities. According to a recent mathematical theorem, in $d = 2$ the information dimension of the harmonic measure is equal to 1 (MAKAROV 1985), therefore, most of the measure is concentrated on a fractal of dimension $D_1 = 1$. Furthermore, the exponent of the $q = 0$th moment has to be equal to the fractal dimension of the substrate on which the measure is defined. These predictions are consistent with the numerical data.

The so called **Dielectric Breakdown Model (DBM)** model was introduced in order to simulate a variety of dielectric breakdown phenomena which range from atmospheric lightning to electric treeing in polymers. Although the actual physical processes can be quite different in these phenomena, the

FIGURE 5. Results for the distribution of growth probabilities $n(\alpha, N)$, where $\alpha = -\ln p / \ln M$ $(M = N)$. Note that $\ln n(\alpha, N)$ is different from $f(\alpha)$ only by an additive constant and that the standard $f(\alpha)$ plot would not have the $\log_{10}^{1.3} M$ term in the denominator of the quantity plotted along the horizontal axis. The different symbols are for clusters of different sizes ranging from $M = 753$ to $M = 20000$. The inset shows the same data for an extended range of α values (SCHWARZER et al 1991).

global properties of the resulting discharge patterns are very similar: they have a randomly branching, open structure resembling DLA.

The actual model is formulated on a d-dimensional hypercubic lattice, and the Laplace operator, ∇^2, is replaced with its discrete version. For example, in two dimensions

$$\phi_{i,j} = \frac{1}{4}(\phi_{i-1,j} + \phi_{i+1,j} + \phi_{i,j-1} + \phi_{i,j+1}), \tag{4}$$

where $\phi_{i,j}$ is the value of ϕ in the grid site i, j. The boundary conditions are the following

$$\phi_{i,j} = 0 \qquad \text{for sites belonging to the cluster,}$$

and $\phi_{i,j} = -1$ for sites on a large circle of radius r_0 centred at the origin. The boundary condition describing the motion of the interface is represented by an expression for the growth probability at the site i, j adjacent to the cluster

$$p_{i,j} = C\nabla\phi^{\eta}_{i,j} = -C\phi^{\eta}_{i,j},$$

where the normalization factor is given by $1/C = \sum \phi^{\eta}_{i,j}$ with the summation running over all of the nearest neighbour sites to the cluster. It is the exponent η which is an important extra property of DBM with regard to diffusion-limited aggregation, because η turns out to be a relevant parameter from the point of view of the fractal dimension of the patterns. There is another slight difference in the boundary conditions. The absorbing boundary condition used in DLA corresponds to a zero potential (probability) in the sites adjacent to the aggregate (not on the cluster itself as is the case for DBM).

The simulation starts with a seed particle at the origin of a lattice. The potential for each site of the lattice within a circle of radius r_0 is calculated using relaxation methods. Formula (4) represents a system of linear algebraic equations (one equation per site) which can be solved by iteration. Relatively good convergence can be achieved by the Gauss-Seidel over-relaxation scheme which on a square lattice has the form

$$\phi^{k+1}_{i,j} = \phi^{(k)}_{i,j} + \omega\left[\frac{1}{4}(\phi^{(k+1)}_{i-1,j} + \phi^{(k+1)}_{i,j-1} + \phi^{(k)}_{i+1,j} + \phi^{(k)}_{i,j+1}) - \phi^{(k)}_{i,j}\right] \qquad (5)$$

if one sweeps the sites in such a way that i and j increase as one goes to the next site. In (5) ω is the over-relaxation parameter. Finding an optimal value for ω by trial and error may speed up the convergence considerably. Next a perimeter site is chosen randomly, and a random number r drawn from a set of random numbers uniformly distributed between 0 and p_{max}, where p_{max} is the largest growth probability. If $r < p_{i,j}$, the perimeter site i, j is filled, and the whole procedure starts again by calculating the distribution ϕ in the presence of the new configuration.

This procedure for growing an N-site cluster requires much more computer time than generating a diffusion-limited aggregate of the same size since one has to solve the Laplace equation within a large region of radius r_0. Correspondingly, the data for the fractal dimension were obtained for clusters consisting of about 10000 particles. The simulations for $\eta = 1$ led to clusters with $D \simeq 1.70$ in good agreement with the expectation that DLA and DBM with $\eta = 1$ belong to the same universality class ($D_{DLA} \simeq 1.7$ in $2d$).

Varying η results in a non-trivial change of the fractal dimension, a property which makes the DBM model particularly interesting from a theoretical viewpoint (a direct connection between η and physical quantities has not been established).

5. Cluster-cluster aggregation

Aggregation of microscopic particles diffusing in a fluid medium represents a common process leading to fractal structures. If the density of the initially randomly distributed particles is larger than zero, the probability for two "sticky" particles to collide and stick together is finite. It is typical for such systems that the resulting two-particle aggregate can diffuse further and may form larger fractal clusters by joining other aggregates. As a result the mean cluster size increases in time and, in principle, after a sufficiently long period all of the particles in the finite system become part of a single cluster. In many cases the force between two particles is of short range and it is strong enough to bind the particles irreversibly when they contact each other. For example, such behaviour can be observed for iron smoke aggregates formed in air or in aqueous gold colloids.

In the above process each cluster is equivalent with regard to the conditions for their motion, i.e., there is no seed particle as in the case of DLA. Consequently, this process is called *cluster-cluster aggregation (CCA)* to distinguish it from particle-cluster aggregation phenomena discussed in the previous two sections. CCA directly corresponds to the physical situation taking place in a system of aggregating particles, in contrast to DLA which in general should be regarded as a computer model for phenomena not necessarily involving attachment of particles.

The possibility of simulating colloidal aggregation in a computer has been recognized for a few decades (Sutherland 1967). However, large scale numerical investigation of cluster-cluster aggregation has become feasible only in recent years. Simple *computer models* for CCA (e.g. Meakin 1983, Kolb et al 1983) can be successfully used to study the structure of aggregates and the dynamics of their formation. A typical two-dimensional simulation is started by randomly occupying a small fraction of the sites on a square lattice to represent particles. At each time step a particle or a cluster is selected randomly and is moved by one lattice unit in a randomly chosen direction. Two clusters stick when they touch each other. Figure 6 shows four stages of such a process. This figure demonstrates the most important properties of cluster-cluster aggregation. With increasing time the number of clusters decreases, and large, randomly branching aggregates appear in the system. The computer generated clusters and the real aggregates observed in many recent experiments were found to have very similar fractal scaling.

Because of the simultaneous diffusional motion of aggregates, the time is a well defined quantity in CCA (including simulations). Accordingly, the related numerical and experimental investigations have concentrated on both the *geometrical and dynamical* aspects of the aggregation process. The results suggest that in analogy with equilibrium phase transitions, non-trivial scaling can be found in both approaches. Therefore, in addition to the fractal structure of aggregates, in this section we shall discuss the dynamic scaling for the cluster size distribution as well (Vicsek and Family 1984).

Several relevant variants of the simplest version can be considered. A deep

FIGURE 6. Snapshots of configurations taken at various "times" during the computer simulation of diffusion-limited cluster-cluster aggregation in two dimensions.

minimum in the potential and a negligible repulsion part results in the so-called i) *diffusion-limited* regime, when two clusters stick rigidly together as soon as they contact. During ii) *reaction-limited* (or chemically-limited) CCA a small, but relevant repulsive potential barrier can prevent the clusters from joining each other even if they are close. However, after a number of contacts they may become joined irreversibly. If the attractive part is not deep enough, one expects that the event of aggregation of two clusters can be followed by

reorganization (restructuring) or dissociation of the aggregates. In the latter case the irreversible character of the process is lost and one is led to deal with iii) *reversible* CCA. The properties of cluster-cluster aggregates are also affected by the iv) kind of motion they undergo. The trajectory of a cluster can be Brownian or ballistic. In addition, v) the clusters may rotate. Many of these processes have been studied by the three main approaches (simulations, theory and experiments) to be discussed in this section.

One of the most realistic models of CCA includes both particle-cluster and cluster-cluster aggregation (JENSEN et al 1996). The model is aimed at interpreting the various aggregation phenomena taking place during thin film growth by vapour deposition. The process is analogous to CCA with a size dependent diffusion constant in two dimensions except that a permanent flux of single particles onto the surface is taken into account. In the various limits of the model it surprisingly well reproduces the corresponding experimental results.

The actual realization of a cluster-cluster aggregation model in the computer depends on the particular process to be simulated. However, the most widely used simulations are based on the following assumptions. The particles are represented by occupied sites of a d-dimensional hypercubic cell of linear size L. To make the finite-size effects smaller, periodic boundary conditions are used. Initially, $N_0 = \rho L^d$ sites are randomly filled, where $\rho \ll 1$ is the density of the particles in the system. Then the clusters are allowed to move following Brownian or ballistic trajectories. If during their motion two or more particles belonging to different clusters accidentally occupy adjacent (nearest neighbour) sites, the clusters combine to form a single new aggregate with a probability $0 < p_s \leq 1$. Moves resulting in overlap of clusters are not allowed.

Both the related experiments and simulations indicate that cluster-cluster aggregates are typically highly ramified, almost loopless structures exhibiting fractal properties. In contrast to off-lattice DLA clusters, the overall shape of cluster-cluster aggregates is not spherical.

In **diffusion-limited cluster-cluster aggregation** (MEAKIN 1983, KOLB *et al* 1983) the clusters are assumed to undergo random walks on the lattice, and $p_s = 1$. The mobility of the clusters is presumed to depend on the number of particles s they are made of. In particular, it is assumed that the diffusion coefficient D_s of a cluster of size s is given by

$$D_s = Cs^{\gamma},$$

where C is a constant and γ can be used to take into account the effects of cluster geometry. For example, in a typical physical system one expects that $\gamma \simeq -1/D$, because the mobility of a cluster in a fluid is inversely proportional to its hydrodynamic radius which for an aggregate of fractal dimension D is close to its linear extension. For the case $\gamma = 0$, corresponding to a mass-independent diffusion coefficient, clusters are selected randomly and moved by one lattice unit in a direction chosen randomly from the $2d$ possible directions.

If $\gamma \neq 0$ the following procedure is used to decide which of the clusters should be moved next. A random number r uniformly distributed in the range $0 \leq r \leq 1$ is selected and the cluster is moved only if $r < D_s/D_{max}$, where D_s is the diffusion coefficient of the given cluster and D_{max} is the largest diffusion coefficient for any cluster in the system.

The d-dependence of the fractal dimension of aggregates grown by diffusion-limited CCA is presented in Table 2. Clearly, these aggregates have a considerably smaller D than DLA clusters generated on lattices of the same dimension. This result is quite plausible; individual particles can penetrate a DLA cluster easily enough to increase its dimension to at least $d-1$. Cluster-cluster aggregates do not tend to fill holes within each other, being fractal structures themselves.

TABLE 2. Dependence of the fractal dimension of diffusion-limited cluster-cluster aggregates on d .

d	D	N_{max}
2	1.43±0.02	$\sim 10^4$
3	1.75±0.01	$\sim 10^4$
4	2.03±0.04	$\sim 10^4$
5	2.21±0.02	$\sim 10^3$
6	2.38±0.02	$\sim 10^3$

Models for **reaction-limited cluster-cluster aggregation** (JULLIEN and KOLB 1984, BROWN and BALL 1985) are constructed to represent the zero sticking probability limit of CCA. If $p_s \simeq 0$, each of the possible contact configurations of two clusters has the same probability to occur.

A possible realization of reaction-limited CCA on cubic lattices is based on placing two clusters at random positions in a large cell. The resulting configuration is accepted as a new cluster only if the two clusters are adjacent and do not overlap. There are two main possibilities for choosing the above two clusters. i) To produce a *monodisperse* size distribution (JULLIEN and KOLB 1984) one starts with 2^n monomers. At each iteration a dimerization is made until there are no monomers left. Next the dimers are joined to form clusters of 4 particles and so on. ii) In the *polydisperse* case (BROWN and BALL 1985) the clusters are always randomly selected from the ones available at the given stage.

Based on the results for the fractal dimension, the monodisperse and the polydisperse cases are principally different. The following values were obtained

$$D = 1.53 \pm 0.01 \quad (d = 2) \quad \text{and} \quad D = 1.94 \pm 0.02 \quad (d = 3)$$

for the monodisperse distribution and

$$D = 1.59 \pm 0.01 \quad (d = 2) \quad \text{and} \quad D = 2.11 \pm 0.03 \quad (d = 3)$$

for the polydisperse system. It is clear from the above expressions that D is larger for these models than it is for diffusion-limited CCA. This is well illustrated by the most important three-dimensional case for which $D \simeq 1.75$ in the diffusion-limited and $D \simeq 2.11$ in the reaction-limited version of cluster-cluster aggregation, in good agreement with the experimental results.

Simulations of off-lattice ballistic aggregation corresponding to cases i) and ii) resulted in practically unchanged fractal dimensions for a given d (MEAKIN 1987). For $d = 2$ and $d = 3$ the values $D \simeq 1.55$ and $D \simeq 1.91$ were obtained, respectively.

The fractal dimension conveys information about the static or geometrical properties of a single aggregate. However, in a typical cluster-cluster aggregation process there are many clusters simultaneously present in the system, and the evolution of this ensemble of aggregates is of interest as well. This time dependence can be investigated by determining the **dynamic cluster-size distribution function** $n_s(t)$, which is the number of clusters in a unit volume consisting of s particles at time t. The behaviour of $n_s(t)$ is *non-universal* as a function of γ. For $\gamma > 0$ the large clusters move faster, and relatively many small ones do not take part in the aggregation process. In this case $n_s(t)$ is characterized by many small clusters and a gradually decreasing number of clusters for growing sizes. If $\gamma < 0$, the small clusters have a higher velocity, so they die out (aggregate) quickly, forming larger clusters. Thus there will be only a few small and very large clusters in the system, resulting in a non-monotonic, bell-shaped distribution.

The study of the statistics of clusters is a common approach to the description of ensembles of clusters. In many equilibrium systems n_s is known to decay as a power law at the critical point. Analogously, $n_s(t)$ has been shown to exhibit static (as a function of s) and dynamic (as a function of t) scaling in a number of close-to or far-from equilibrium systems. Next we first treat computer simulations of the diffusion-limited and related cluster-cluster aggregation models together with the **dynamic scaling** picture emerging from these numerical investigations.

In the diffusion-limited CCA model to make the cluster size distribution function independent of the cell size we use the definition $n_s(t) = N_s(t)/L^d$, where $N_s(t)$ is the number of s-clusters at time t in the cell. The elapsed time is measured by increasing t by an amount Δt each time a cluster is selected to move. Simulation results for $n_s(t)$ can be well represented by the *scaling assumption* (VICSEK and FAMILY 1984)

$$n_s(t) \sim s^{-2} f(s/t^z), \tag{6}$$

where $f(x)$ depends on the mobility exponent γ. In particular,

$$f(x) \sim x^2 g(x) \qquad \text{for} \quad \gamma < \gamma_c$$

FIGURE 7. Scaling of the dynamic cluster-size distribution function. (6) is supported by the fact that after a relatively short time the data determined for various times fall onto a single curve. In these three-dimensional simulations $\gamma = -2$ was used (MEAKIN *et al* 1985).

with $g(x)$ exponentially small for both $x \ll 1$ and $x \gg 1$, and

$$f(x) \sim \begin{cases} x^{\delta}, & \text{if } x \ll 1 \\ \ll 1, & \text{if } x \gg 1 \end{cases} \qquad \text{for} \quad \gamma > \gamma_c.$$

The scaling (6) can be checked by plotting the quantity $s^2 n_s(t)$ as a function of s/t^z. If (6) is valid, then the results obtained for a given γ should fall onto a single curve corresponding to the scaling function $f(x)$. Figure 7 demonstrates that for the simulations carried out in $d = 3$ with $\gamma = -2$ this is indeed the case.

6. GROWING SELF-AFFINE SURFACES

In many cases rough surfaces are generated by a growing interface which advances as new parts are added according to some dynamical process. Examples include, crystal growth, vapor deposition, electroplating, spray painting and coating, and biological growth. Fractally rough surfaces may also be formed during the removal of material, as in chemical dissolution, corrosion, grinding, erosion, blasting, wear and all types of polishing. The growth of interfaces is intimately related to a variety of other processes, including the propagation of flame fronts, the long time behavior in randomly stirred fluids, impurity roughening and pinning of interfaces, and the problem of directed polymers in random media. These relations provide powerful connections among seemingly

different phenomena, which can be exploited in the development of various approaches for understanding the evolution of rough interfaces.

During the growth of compact (non-fractal) objects the motion of the interface is directed outward, and this orientation plays a special role. Typically, the interface can be well approximated by a *single valued* function $h(\mathbf{r})$ of $d-1$ variables, e.g., one can describe the properties of the surface by examining only those points of the object which are the farthest from the centre of the structure in a given direction. The scaling properties of such surfaces (with irrelevant overhangs) are *direction dependent*: parts of various sizes can be rescaled into an object with the same overall behaviour using a rescaling factor in the direction parallel to the growth which is different from that needed to rescale the perpendicular lengths.

For the 1+1 dimensional case (growth from a line) this can be symbolically expressed as

$$h(x) \simeq b^{-H} h(bx) \tag{7}$$

where $H > 0$ is some exponent. The plot of the distance of a randomly walking particle in one dimension as a function of time can be shown to satisfy the above scaling. An equivalent feature of the self-affine functions is that their height-height correlation function $c(\mathbf{r})$ scales as

$$c(\mathbf{r}) = \langle |h(\mathbf{r}) - h(\mathbf{r} + \Delta\mathbf{r})| \rangle \sim \Delta\mathbf{r}^H,$$

where H is the same exponent as in (7).

The two most studied aggregation models leading to self-affine surfaces are the Eden and the ballistic aggregation models. The first of these was introduced by Eden in 1961 to simulate the growth of tumors. In addition to its biological applications, this model has relevance to many other types of stochastic growth phenomena with stable or marginally stable interfaces. When growing an *Eden cluster* one of the empty sites next to the aggregate (perimeter sites) is chosen randomly, and it is added to the cluster. A large cluster is obtained after having repeated this procedure many times.

In ballistic aggregation models the particles move along straight trajectories until they encounter the growing aggregate and stick to its surface irreversibly. This kind of kinetics is typical for experimental situations when molecules move in a low density vapour.

In an actual simulation of ballistic deposition the *strip geometry* with periodic boundary conditions is used. The linear size of the substrate is denoted by L. The particles are launched at randomly selected positions at a height of $h_{max}+1$ which is the maximum height of any particle in the deposit. Then the particles follow a straight trajectory with a prescribed angle of incidence until they contact either a particle in the deposit or reach the original surface. At the point of the first contact they are stopped and become part of the growing deposit. In the lattice version of the model with vertically dropped particles it is easy to see that there is only one active perimeter site (a site which has

FIGURE 8. Ballistic aggregate growing in the strip geometry.

the possibility to be filled in the next step) in each column. Since one needs to record only the height of these L sites such an algorithm is fast and does not require much computer time.

Both models lead to surfaces exhibiting interesting scaling both in space and time. The corresponding **dynamic roughening** of interfaces is an example of a far-from-equilibrium phenomenon without a complete theory based entirely on first principles. However, the analysis of the scaling behaviour of the time and spatial dependence of the surface properties has led to the development of a general **dynamic scaling** approach for describing growing interfaces (FAMILY and VICSEK 1985).

A fruitful approach to the characterization of self-affine surfaces changing in time is the determination of various *correlation functions*. The most convenient quantity is the time dependent so-called height-height correlation (or height difference) function $c(r,t)$ which is defined as

$$c(r,t) = \langle |h(\mathbf{r}',t') - h(\mathbf{r}'+\mathbf{r},t'+t)| \rangle_{\mathbf{r}',\mathbf{t}'},$$

which is the average height difference measured for a time difference t at two points whose coordinates on the substrate are separated by $\mathbf{r}$. For surfaces isotropic in the directions perpendicular to the growth the correlation function depends only on the absolute value of this vector, r. On the basis of the simulation results and general scaling considerations $c(r,t)$ behaves as

FIGURE 9. Schematic plots of the height correlation functions $c(r,0)$ and $c(0,t)$.

$$c(r,t) \sim r^H f\left(\frac{t}{r^z}\right) \tag{8}$$

with an $f(x)$ given by

$$f(x) \sim \begin{cases} x^\beta, & \text{if } x \ll 1 \\ \text{Const}, & \text{if } x \gg 1. \end{cases} \tag{9}$$

Correspondingly, for long times and for $r << L$ the correlation function $c(r,0)$ is given by

$$c(r,0) \sim r^H,$$

and for fixed r and short times

$$c(0,t) \sim t^\beta.$$

The above discussed expressions can be used to determine the exponents H and β for experimentally observed surfaces. The schematic behaviour of $c(r,0)$ and $c(0,t)$ is shown in Figure 9. The average height difference can be considered as the correlation length $\xi_\perp$ in the growth (perpendicular to the substrate) direction. According to (8) and (9)

$$\xi_\perp \sim t^\beta, \quad \text{for} \quad t \ll \tau \qquad \text{and} \qquad \xi_\perp = \text{Const}, \quad \text{for} \quad t \gg \tau,$$

where the relaxation time τ is proportional to L^z. The spread of fluctuations in the direction parallel to the substrate is characterized by the correlation length

$\xi_{\|}$. This shows that how far the effect of a perturbation can get after some time t along the surface. On the basis of the dynamic scaling form (8)

$$\xi_{\|} \sim t^{H/\beta}, \quad \text{for} \quad t \ll \tau \qquad \text{and} \qquad \xi_{\|} = L, \quad \text{for} \quad t \gg \tau.$$

Most of the studies in the field of self-affine growth concentrate on the determination of the exponents H and β entering the above definitions characterizing the time development of surfaces.

The growth of rough surfaces represents a quickly developing field, where one of the main directions is to construct and study models closely corresponding to actual physical or technological processes. In particular, wetting fronts were described in terms of directed percolation depinning models in which the inhomogeneity of the medium in which the growth occurs is taken into account (Buldyrev et al 1996). Such models are expected to describe a wide class of processes including such examples as paper tearing (Kwon 1997) or the penetration of ink into a paper towel (Horváth and Stanley 1995).

7. Conclusion

We have described the basic types of fractal growth phenomena and the computer models which help to understand their formation. These are models of the evolving patterns originally formed under more complex experimental conditions. We have demonstrated that the theoretical interpretation of aggregation processes can be based on the principle of dynamical scaling. Most of the introduced models can easily be programmed and are contained in our Fractal Growth software package (Vicsek and Vicsek, 1989) (including other models as well) together with their source code in Pascal.

References

1. Arneodo, A., Argoul, F. J., Muzy, F. and Tabard, M. (1993).*Fractals*, **1**, 629
2. Barabási A. L. and Stanley H.E. (1995). *Fractal Concepts of Surface Growth*. Cambridge UP,
3. Brown, W.D., Ball, R.C. (1985). *J. Phys. A* **18**, L517
4. Buldyrev, S. V., Amaral, L. A. N., Barabasi, A-L., Harrington, S. T., Havlin, S., Kertesz, J., Sadr-Lahijany, R. and . Stanley, H. E. (1996). *Fractals* **4**, 307.
5. Bunde A., and Havlin S., (editors) (1994).*Fractals in Sciences*. Springer-Verlag, Berlin,
6. Evertsz, C. J. G., Peitgen, H-O and Voss, R. F. (editors) (1996). *Fractal Geometry and Analysis*, World Scientific, Singapore, New Jersey.
7. Family, F. and Vicsek, T. (1985). *J. Phys. A* **17**, L639
8. Horváth, V. K. and Stanley, H. E. (1995). *Phys. Rev.* **E52**, 5166
9. Jensen P., Barabasi, A-L., Larralde, H., Havlin, S. and Stanley, H. E.(1996). *Fractals* **4**, 321

10. JULLIEN, R., KOLB, M. (1985).*J. Phys. A* **18**, L517
11. KERTESZ, J. and VICSEK,T. (1986). *J. Phys.* , **A19**, L257
12. KOLB,M., BOTET,L. and JULLIEN, R. (1983). *Phys. Rev. Lett.*, **51**, 1123.
13. KWON, T. H. (1997) *Fractals*, **5**, 121
14. MAKAROV, N. G. (1985). *Proc. London Math. Soc.* , **51**, 369
15. MANDELBROT, B.B. (1982). *The Fractal Geometry of Nature*. Freeman, San Francisco.
16. MEAKIN, P. (1983). *Phys. Rev. Lett.*, **51**, 1119.
17. MEAKIN, P. VICSEK, T. and FAMILY, F. (1985).*Phys. Rev.*, **B31**, 564
18. STAUFFER, D. (1985).*Introduction to Percolation Theory*. Taylor and Francis, London,
19. SUTHERLAND,D. N. (1967). *J.Colloid Interface Sci.* , **25**, 373
20. SCHWARZER,S., LEE,J., HAVLIN, S., STANLEY, H.E. and MEAKIN, P. (1991).*Phys. Rev.* **A43**, 1134
21. VICSEK,T. (1992). *Fractal Growth Phenomena* (World Scientific, Singapore.
22. VICSEK, M. and VICSEK, T. (1992). *Fractal Growth, Demonstration of Fractal Aggregation Models*. World Scientific, Singapore.
23. VICSEK, T. and FAMILY, F. (1984). *Phys. Rev. Lett.* **52**, 1669
24. T. VICSEK, M. SHLESINGER and M. MATSUSHITA (editors) (1994). *Fractals in Natural Sciences*, World Scientific, Singapore, New Jersey.
25. WILKINSON, D. and WILLEMSEN,J. (1983). *J.Phys.* , **A16**, 3365
26. WITTEN,T.A. and SANDER,L.M. (1981). *Phys. Rev. Lett.*, **47**, 1400