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Random successive growth model for pattern formation

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In order to explain patterns formed during crystallization of metal-semiconductor films and during bacterial colony growth, a random successive growth model where no long-range diffusion is necessary has been proposed. In this model the growth is controlled by two local conditions: growth probability of the neighboring sites around the cluster and occupation ratio of the sites of the cluster inside a small circle with the potential growth site as its center. Varying these conditions, fractal, dense-branching, and compact growth patterns have been obtained.

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There are various patterns such as fractal growth, dense-branching morphology (DBM), compact growth, dendritic growth, and chiral growth [1] in the field of materials science and life science. Understanding the formation of these patterns from a nonequilibrium environment and the transition between different growth morphologies has long been a challenge [2]. In the last decade, it was widely studied in order to characterize the geometrical properties of patterns formed in electrochemical deposition [3], colloidal aggregation [4], dielectric breakdown [5], thin solid films [6], and bacterial colony growth [7]. Much effort has especially been devoted to establishing the relationship between cluster geometry and the growth mechanism. A number of models for pattern formation have been proposed to account for various experimental results. For example, the Eden model [8], in which each vacant surface site of a cluster can grow in the same probability, can generate a compact pattern with a rough surface. The model proposed by Meinhardt [9] can form a uniform (nonfractal) network of occupied lattice sites after combining nonlinear chemical reaction processes and diffusion. The prototype of the fractal growth model is the diffusion-limited aggregation (DLA) model introduced by Witten and Sander [10]. Since this work, many variants of the DLA model have been studied for various purposes. Erlebacher, Searson, and Sieradzki [11] divided the growth field of electrochemical deposition into two areas: a "space charge" region surrounding the aggregate in which particle motion is biased to branch tips, and a second region surrounding the first in which particle motion follows unbiased random walks. They found a smooth transition between DLA and DBM. Recently, Ben-Jacob and co-workers [12] proposed a communicating walkers (mesoscopic units of bacteria) model to simulate the growth of bacterial colonies. The model incorporates random walkers, representing aggregates of bacteria, which move in response to gradients of nutrient concentration and communicate with each other by means of chemotactic "feedback." The model can generate self-organization over a wide range of length scales. DLA and its variants depend on the Laplace equation which determines a long-range field, e.g., concentration field, electric potential field, or pressure field.

While DLA and other related models are important in understanding growth phenomena in nonequilibrium systems, it is difficult to simulate various patterns by long-range growth processes. So the recent model proposed by Ben-Jacob *et al.* incorporates local terms expressed in the δ functions at the walkers' sites into the diffusion equation to generate various patterns. In our opinion, the pattern formation in thin solid films or in bacterial colony growth processes might be mainly explained by a local mechanism rather than the long-range one.

A high-resolution electron microscopy observation [13] shows that the pattern formation of amorphous Ge crystallation in metal–a-Ge films is a random successive nucleation process. The fractal regions are composed of many Ge crystallites, while the Ge sublayer in matrix regions remains in the amorphous state. At the early stage of crystallization, Ge nuclei are formed at preferred sites located at the metal-Ge interface. The released heat (crystallization energy and strain energy) leads to a local temperature rise which stimulates new nuclei of the next generation appearing randomly in nearby regions. The above process repeats many times during annealing, resulting in the pattern formation. Based on the crystallization of amorphous Ge or Si films contacted with various metal films [13,14] we proposed a random successive nucleation model which depends only on local conditions [15]. In this model, long-range diffusion is not necessary since Ge atoms are uniformly distributed over the whole sublayer. The main purpose of the model is to simulate the experimental fractal-like structures, but the growth conditions remain more or less complicated.

In this paper a simpler and more universal model, the random successive growth model, is used to simulate not only fractal-like structures in thin metal-semiconductor films, but also various patterns of bacterial colony growth, since the colony growth is also based on the division of existing bacteria. In Fig. 1 the initial stage of growth on a 181×181 square lattice from a seed of five neighboring sites (small filled squares) is shown. The random successive growth proceeds generation by generation at the sites in the ring of increasing radius R with a width of one lattice unit

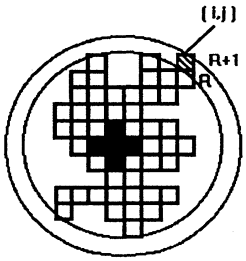

FIG. 1. The model of random successive growth on a square lattice with a seed of five neighboring sites, where R is the maximum radius if the cluster composed of filled and open squares and (i,j) is the potential growth site.

after the maximum radius of the cluster composed of all the filled and open squares has reached R. The lattice sites inside the circle of radius of $(R-1)$ have to stop their growth since the previous growth has caused part of the stored energy to be released in bilayer films. In the case of colony growth, the stop of growth inside the circle of R may be caused by the consumption of nutrients and accumulation of harmful metabolites in the fjords. All sites in the ring R are randomly chosen as well as the sites of the next ring of radius $R+1$. The cluster can grow (the filled square in Fig. 1 can transform to an open square) if the following two conditions are fulfilled simultaneously.

(i) One of the nearest vacant sites, e.g., the filled site (i,j) (the nearest-neighbor option), or one of the nearest and next-nearest vacant sites (the next-nearest-neighbor option) around the cluster may become a site of the cluster (open square) if a random number between 0 and 1 produced by computer is smaller than a given growth probability p.

(ii) Inside the circle of radius r (≥ 1) centered on the chosen site (i,j), the number of cluster sites must be less than a given integer n which is smaller than the total number of lattice sites inside the circle, i.e., the growth must take place in a region not so crowded with cluster sites. In other words, the growth is prevented by the crowding condition. The ratio $n/\mathcal{I}(\pi r^2)$ can be defined as an occupation ratio c inside the circle, where $\mathcal{I}$ means truncating the following expression to an integer. The fractal dimension was calculated by a sandbox method using circles of increasing radius which is limited inside the radius of gyration of the pattern obtained.

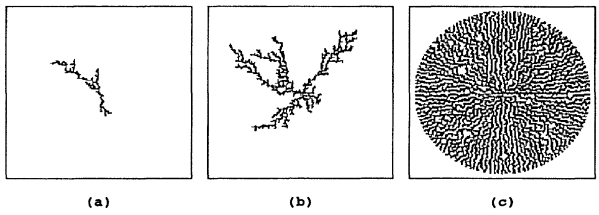

FIG. 2. The simulation results obtained from the nearest-neighbor option when the crowding condition is $r=3$ and $n=7$ (seven allowable occupied sites inside a circle of radius of 3) with various growth probabilities: (a) $p=0.62$, (b) $p=0.68$, and (c) $p=1.0$.

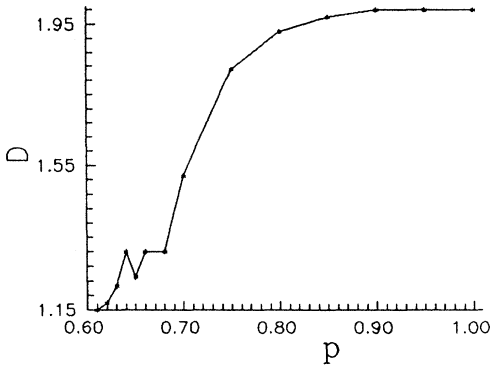

FIG. 3. The dependence of fractal dimension on the growth probability p in the case of $r=3$ and $n=7$.

Figures 2(a)–2(c) show the results obtained from the nearest-neighbor option in the cases of $r=3$ and $n=7$ where $p=0.62$, 0.68, and 1.0, respectively. It can be seen from the figure that the growth probability p can influence the pattern formation significantly. When p is not large enough, e.g., $p\leq 0.62$, the pattern will interrupt somewhere, but the fractal dimension can be obtained inside the radius of gyration of the pattern. The pattern can continue growing when p is equal to or larger than 0.68. The pattern becomes a dense-branching one when p is large enough $(p\geq 0.9)$. Figure 3 shows the dependence of fractal dimension D on growth probability p in the case of $r=3$ and $n=7$, where D increases rapidly in the range of p from 0.6 to 0.8 and reaches 2.0 as $p\geq 0.9$.

If the crowding condition can be ignored, i.e., the ratio $c=n/\mathcal{I}(\pi r^2)$ is 1, indicating that inside the circle around the site (i,j) the allowable number of the cluster sites n can reach the total number of lattice sites, then our model is similar to the percolation model in the sense of forming an infinite growth pattern. The threshold value of site percolation on a square lattice is 0.593, which is significantly smaller than the value 0.68 in our case. The ratio c used to obtain the results in Fig. 2 is 0.24, from which one finds that the crowding condition will prevent the growth process when the growth probability is less than 0.68. The patterns obtained in the case of $p=0.62$–0.68 are similar to the experimental fractal-like structures formed in the Pd–a-Si [14] and Au–a-Ge [13] bilayer films.

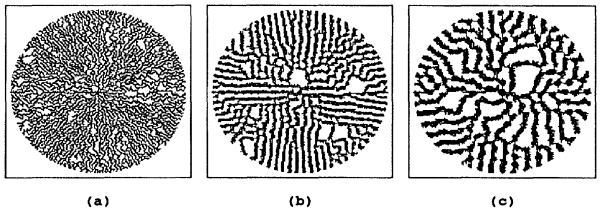

FIG. 4. The simulation results in the case of growth probability $p=1$ with the same occupation ratio inside the circle of various radii. (a) $r=2$, $n=3$; (b) $r=5$, $n=19$; and (c) $r=8$, $n=46$, respectively.

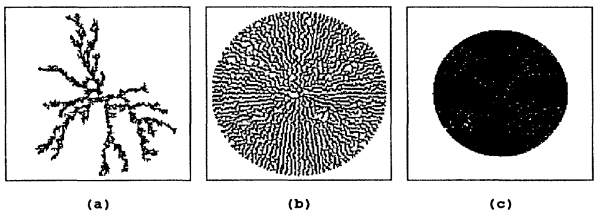

FIG. 5. The patterns formed by the random successive growth model, where (a) $p=0.62$, $c=0.25$, (b) $p=1.0$, $c=0.25$, (c) $p=1.0$, $c=1.0$, which are similar to the patterns of bacterial colony growth in Ref. [7].

Figures 4(a)–4(c) show the influence of the crowding condition, where $r=2$, 5, and 8, and $n=3$, 19, and 46, respectively, and the occupation ratio is nearly the same in patterns in the case of the growth probability $p=1$. It is clear that the larger the radius r, the thicker the branches of the patterns. The influence of range r is similar when the growth probability is much less than 1.

In order to simulate various patterns of bacterial colony growth (fractal-like, dense branching, and compact morphologies as shown in Ref. [7]), the growth probability p and the occupation ratio c have changed. In Figs. 5(a)–5(c) patterns are obtained where $p=0.62$, $c=0.25$ (a), $p=1.0$, $c=0.25$ (b), and $p=1.0$, $c=1.0$ (c), respectively. The fractal dimension of Fig. 5(a) is 1.65 as measured by the sandbox method and the dimensions of Figs. 5(b) and 5(c) are obviously close to 2. It can be seen that the patterns obtained by the simulation and the pattern generated by the colony growth (Figs. 1(a), 5, and 8 of Ref. [7]) are quite similar.

Figure 6 shows the pattern obtained by the next-nearest option where the growth site (Fig. 1) may also be the next-nearest-neighboring site around the cluster site, and the growth probability is 0.37 in the case of a constant crowding condition $r=3$ and $n=7$ ($c=0.24$). Comparing the pattern with the patterns generated by the nearest option in Fig. 2 we can see that many next-nearest-neighboring sites have become cluster sites so that the threshold value of continuing growth is about 0.37, which is much smaller than that of the nearest option, 0.68. It is also found that the dependence of the pattern morphology on the growth probability p is more sensitive than that of the nearest option.

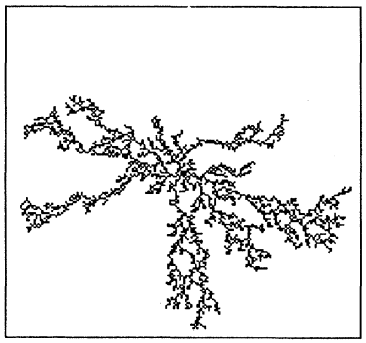

FIG. 6. The result obtained from the next-nearest-neighbor option in the case of $r=3$, $n=7$ with growth probability $p=0.37$.

The various patterns formed in the colony growth have been mainly explained by the diffusion-controlled growth mechanism [1,7,12]. But the results obtained by the random successive growth model suggest that a local model not controlled by the long-range [$(2-3)R$)] process can also generate various patterns. This model is originally based on the experimental results of crystallization of metal-semiconductor bilayer films, but it can also be applied to explain the colony growth on agar plates. In either of these two cases, atoms of amorphous semiconductor or the nutrient particles exist everywhere over the whole area. Indeed the fractal formation is accompanied by the interdiffusion between metal and semiconductor atoms in the bilayer films, but its range is limited in an order of the spacing between branches as shown by transmission electron microscopy and x-ray energy dispersive spectroscopy [13,16], i.e., in an order of r (much smaller than R). In the case of colony growth, the range of the interdiffusion between nutrients and harmful metabolites may be similar. So, the pattern formation can be controlled by local conditions rather than by the long-range field of the DLA model and the other related models.

The fractal-like structure is obtained when the growth probability p is less than some certain value. In the case of large growth probability, a small occupation ratio will generate a dense-branching pattern and a large one a compact morphology. So the various patterns of colony growth determined by the concentration of nutrients and moisture can be naturally explained by varying the growth probability and the occupation ratio in our model as shown in Fig. 6, since a higher concentration of nutrients may be regarded as a larger allowable occupation ratio and the moisture (later expressed inversely as the agar concentration) in colony growth [7] may increase the growth probability. Then as the growth probability and the allowable occupation ratio increase, the morphology changes from a fractal-like pattern to a dense-branching or round pattern, in agreement with the experimental results. It is not necessary to explain the various patterns by different models, e.g., the fractal-like morphology by the DLA model and the compact one by the Eden model as suggested by Fujikawa and Matsushita [7]. It is also not necessary to combine the random walk in the outer region and migration in the inner region to obtain the transition between fractals and dense-branching morphology as proposed by Erlebacher *et al.* [11].

The DLA model and our model can be considered as two extreme cases: in the former the limiting factor is the long-range diffusion, and in the latter the limiting factor is the local growth condition. The situation is similar to the reactive diffusion, where two extreme cases are treated as a diffusion-limited process through a reacted layer and reaction-limited process at the interface between the substrate and reacted layer. The communicating walkers model has combined the local condition into the diffusion equation so it is situated in the intermediate case. When the nutrient is sufficient enough, the diffusion process can be neglected and a compact pattern based on the local reproduction of bacteria at the envelope is formed.

Since the random successive growth model is simple, it can be easily modified in several ways. First, the width of the

ring can be changed from one lattice unit to several units. In this case the surface of a compact pattern will become rough when the growth probability is large, and the model will be similar to the Eden model. Second, the distance between the potential growth site (filled site) and the nearest cluster site may be larger and one step of the growth may also be expanded to 5, 9, or more neighboring sites simultaneously. Then the branches on the fractal-like structure will be thicker, similar to the morphologies obtained from crystallization at higher annealing temperature and from the colony growth at higher nutrient concentration. Finally, the growth chance for every site is only once, i.e., its growth probability attenuates suddenly from p to zero in the present simulation. Of course, the probability p may be gradually decreased generation by generation. However, we believe that although the modifications will change the morphology in some detail, the main results will remain unchanged.

In summary, we have performed a computer simulation for pattern formation in metal-semiconductor films and in bacterial colony growth. Our simulations incorporate two local conditions: growth probability of the neighboring sites around the cluster and occupation ratio of the sites of the cluster inside a small circle with the potential growth site as its center. Varying growth conditions, a smooth transition from fractals to DBM and compact growth has been obtained. We have shown that the growth mechanism revealed by this model applies to pattern formation. A better understanding of this mechanism, for example, why the transition takes place from fractals to DBM, and from DBM to compact growth at certain growth conditions, requires further theoretical work.

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