Pattern formation on nonuniform surfaces by correlated random sequential absorptions

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The pattern formation on nonuniform surfaces by correlated-random sequential absorption (CRSA) process has been investigated by computer simulations. The nonuniform surfaces are represented by percolation clusters with probabilities p_s and p_s stands for the nonuniform degree of surfaces. The interactions between the particles and the defects in surfaces are involved by introducing a sticking coefficient s. When $s \to 0$, the CRSA process is controlled by the absorption of surfaces and the correlation between particles. With the correlation increasing from a weak limit to a strong one, the cluster consisting of absorbed particles changes from the dispersed pattern of site percolation to correlated percolation, and then to Leath percolation clusters. When $s \to 1$ and $p_s \to p_c$, the CRSA process is dominated by the absorption of the defects, where p_c is the threshold of percolations. The patterns appear randomly dispersed in spite of the correlation. With the decrease of s and increase of p_s, the interaction controlling the CRSA process changes from the absorption of defects to that of surface and the correlation between particles gradually. For the system $s \to 0$, the transition correlation exponent $\alpha_c = d_s$, where d_s is the fractal dimension of the percolation surfaces.

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I. INTRODUCTION

Nonequilibrium growth often gives rise to fractal structure, which are statistically self-similar over a range of length scales. Fractal growth phenomena have attracted considerable attention during the last two decades [1–3]. There are two basic models of fractal aggregation: particle-cluster aggregation (PCA) [4,5], cluster-cluster aggregation (CCA) [6,7]. In PCA model, the particles are deposited one by one, so it is suitable in the case of very low flux for monolayer growth [2,4,8]. The model describes a great variety of aggregation processes such as cluster formation in electrodeposition, molecular-beam epitaxy, and growth of bacterial colonies [3,9]. If the particles are put into the system all at once, the aggregation process can be described by CCA model, e.g., colloid aggregation [1,3,6,7].

In many other systems, particles become immobile once they are deposited onto the surface. These systems can be described by the random-sequential-adsorption (RSA) processes [10]. A variety of physical, chemical, and biological problems can be modeled by RSA processes [11]. For example, chemisorption on single crystal surfaces, at low temperature where surface diffusion is limited, provides a natural application of RSA processes. Other examples include deposition of macromolecules and microscopic particles, reactions on one-dimensional (1D) polymer chains, car parking, etc. [11]. Except for the shape size and overlapping of particles, RSA is similar to the process of producing site percolation [11–14]. The geometric and kinetic characteristics of RSA process in uniform space are fairly well known [10–12]. In RSA process, the correlation between deposited particles, the nonuniformity of space, and the interaction between the defects in surfaces and particles, have not been paid much attention [10–13]. In the process of urban growth, there is the correlation between settlers, i.e., later settlers prefer to settling down near to the former ones [15]. And in the process of epidemic spreading, the individuals become correlated due to the spreading method of diseases [16–18]. As for the cluster formation on surfaces, many actual surfaces are not so uniform and flat due to the local oxidation, pollution, and other factors. Therefore, the study on the correlated RSA process on uniform surfaces is important. The nonuniform surfaces can be described by percolations [14,19,20]. The probability p_s by which the percolation is created, stands for the nonuniform degree of surfaces.

In this paper, we present the model of correlated-random sequential adsorption (CRSA) on nonuniform surfaces. Our results will be helpful for understanding the influences of spatial nonuniformity and the correlation between particles on the RSA processes.

II. MODEL AND SIMULATIONS

The simulation algorithm is composed of two parts: producing percolation surfaces and producing clusters by CRSA on the percolation surfaces (spaces).

The percolation surfaces were produced by Leath method, which can generate a single percolation cluster and has been introduced in Refs. [14,21] in detail. There is a critical probability p_c (for 2D square lattice, $p_c \sim 0.593$) [14,21]. When $p_s < p_c$, the Leath percolation cluster can grow to an infinite one. To produce an infinite surface, the probability p_s is taken values in the range $p_c \lesssim p_s \leqslant 1$, which describes the nonuniform degree of surfaces. In percolation surfaces, *the occupied sites and blocked ones represent the normal sites and defects in surfaces*, respectively.

Going a further step, we generate clusters on nonuniform surfaces by CRSA processes as follows. At the initial time, there is one seed on the percolation surfaces. Then particles are randomly deposited on the surface one at a time. *The particles can only be deposited onto the normal sites in surfaces*. The adsorbed probability for a deposited particle due

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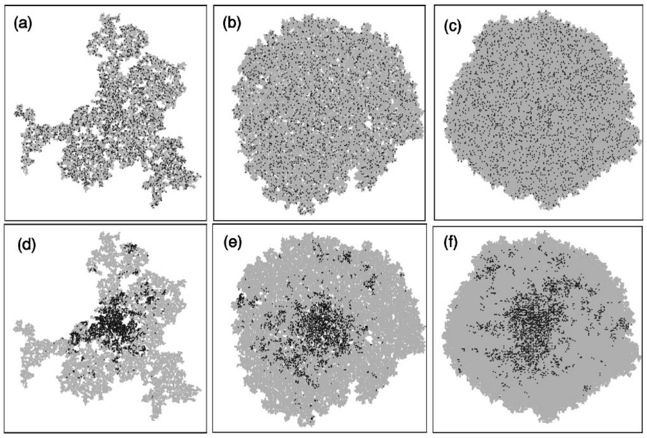

FIG. 1. In the case where the defects are inert, i.e., $s=0$, the distributions of absorbed particles on percolation surfaces with variant probabilities p_s and correlation exponents α. (a) $p_s=0.593$ and $\alpha=1.5$; (b) $p_s=0.7$, and $\alpha=1.5$; (c) $p_s=1$ and $\alpha=1.5$; (d) $p_s=0.593$ and $\alpha=3.0$; (e) $p_s=0.7$ and $\alpha=3.0$; and (f) $p_s=1$ and $\alpha=3.0$.

to the ith already adsorbed particle is given by the following form [15–18,22]:

$$p_i=1/r_i^{\alpha}, \tag{1}$$

where α is the correlation exponent ($\alpha\geqslant 0$) and r_i stands for the distance from the deposited particle to ith adsorbed particle. For some systems, the defects in surfaces can also become traps for particles [8,23]. So the adsorbed probability for a particle due to the kth defect can be written as [8,23,24]

$$p_k'=s, \tag{2}$$

where s $(0\leqslant s\leqslant 1)$ is the sticking coefficient and stands for the ability of defect absorbing particles, just like that in the model of reaction-limited aggregation [25,26]. Furthermore, considering the effects of all the already adsorbed particle and defects on the deposited particle, the total adsorbed probability can be given by

$$\mathrm{P}=\sum_{i=1}^{N} p_i+\sum_{k=1}^{N'} p_k', \tag{3}$$

where N and N' are the current numbers of absorbed particles and defects, respectively. Deposit a particle, calculate its adsorbed probability P, and the particle is absorbed with the probability P. This process is repeated until the expected growth scale is obtained.

III. RESULTS AND DISCUSSION

Numerical simulations are performed on a finite square lattice by Monte Carlo methods. The length of square particles is chosen to be the unit of length. The occupied fraction is given by $\phi=N/N_0$, where N and N_0 are the particle number of absorbed particles and that of normal sites in percolation surfaces.

A. Patterns on nonuniform surfaces formed by CRSA process

We simulated the clusters by CRSA processes on percolation surfaces with variant occupied probabilities p_s and sticking coefficients s of defects for a set of correlation exponents α.

First, we investigate the case where the defects in surfaces are inert $(s=0)$, i.e., the defects cannot absorb particles. In the case of the weak correlation (α is small), i.e., there is only a weak interaction between deposited particles. All adsorbed particles are dispersed randomly on the percolation surfaces [see Figs. 1(a–c)]. It is the random pattern formed by RSA processes [11], except that the dispersed distribution is limited by percolation spaces. Due to the nonuniformity of surfaces, the patterns in percolations with small p_s appear sparser than those in ones with large p_s. This is just the site percolation in percolation space, and the occupied fraction ϕ corresponds to the occupied probability in producing site percolation [14]. As α increases, the correlation between particles gets strong, i.e., former absorbed particles have influence on the adsorption of later deposited particles. Particles prefer to settle near the already absorbed ones. Thus, absorbed particles concentrate in some areas and form dense pattern locally on percolation surfaces [as shown in Figs. 1(d–f)]. The pattern is just the correlated site percolation except for the percolation space [22], which is similar to the pattern of urban growth [15]. If α is large enough, the correlation becomes very strong, i.e., the absorption strongly depends on the former particles. In this case, we get $P=1$ (when $r=1$) and 0 (when $r>1$) from Eqs. (1) and (3). It means that the deposited particles can only be absorbed on the nearest sites to the cluster composed of the former absorbed particles. This is just the growth rule of the Eden cluster [28]. Because the growth spaces are percolation clusters, the patterns consisting of absorbed particles are Leath percolation clusters with variant p_s. The cluster is the Eden "pie" for $p_s=1$, or Leath percolation for $p_s<1$ [14,28].

Now, we study the effect of the absorption between the defects and deposited particles on the pattern produced by CRSA processes. In this case, the defects are active $(s\neq 0)$, the interaction between defects and particles should be considered. In the case of small α, the correlation between particles is weak, but the absorption of surfaces is strong. s has little influence on the pattern and particles still keep randomly dispersed on percolation surfaces.

In the following, we discuss the case of strong correlation in details. In the case of large α, particles prefer to aggregate due to the strong correlation between particles. But when p_s is small, there exist many defects in surfaces and the defects can absorb the particles with probability s. So the patterns change a lot, especially for $s\rightarrow 1$. As $s\rightarrow 1$ and $p_s\rightarrow p_c$, the pattern keeps random dispersed in spite of the change of α. The growth process is dominated by the absorption of the defects which are distributed dispersedly in surfaces. As s decreases to 0, the patterns change to some sparse islands [shown in Figs. 2(a) and 2(b)]. If the correlation becomes very strong, the islands will appear very dense and the number of islands decreases with the increase of α and the decrease of s [see Figs. 2(d) and 2(e) comparing with Figs. 2(a)

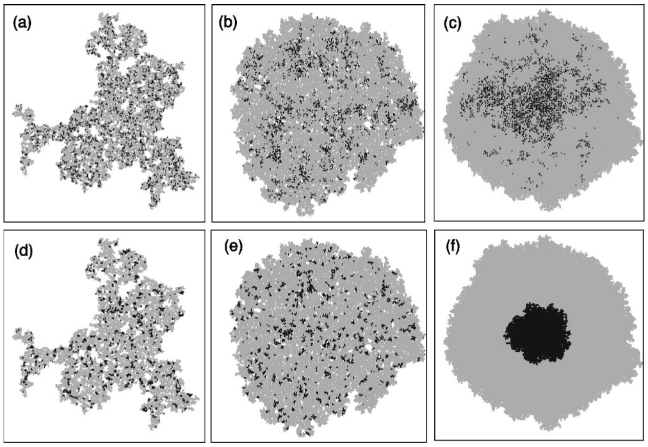

FIG. 2. In the case that sticking coefficient of defects $s=0.01$, the distributions of absorbed particles on percolation surfaces with variant probabilities p_s and correlation exponents α. (a) $p_s=0.593$ and $\alpha=3$; (b) $p_s=0.7$ and $\alpha=3$; (c) $p_s=1$ and $\alpha=3$; (d) $p_s=0.593$ and $\alpha=7$; (e) $p_s=0.7$ and $\alpha=7$; (f) $p_s=1$ and $\alpha=7$.

and 2(b)]. When $p_s=1$, the surface reduces to Eden ''pie'' which is similar to a uniform and flat 2D space. There are very few defects in Eden ''pie,'' so s has little influence on the pattern, which can be seen in Fig. 1(f) and Fig. 2(c). If α is large enough, the pattern will become the Eden ''pie'' [shown in Fig. 2(f)].

From the above, we generalize a conclusion. For $s=0$ (inert defects), the influence of surfaces on the pattern is not much. The nonuniform spaces only make the patterns become sparser. Particles are distributed randomly in percolation space, which is original RSA in percolations and corresponds to the weak limit of the present model. For the case that the adsorption between particles is very strong, particles prefer to aggregate and form dense pattern of single cluster, which reduces to Leath percolation and corresponds to the strong limit of the model. For $s\neq 0$ (active defects), due to the adsorption of defects, the influence of surfaces on the pattern is very much. When p_s is small and s is large, the process is controlled by the adsorption of defects and the correlation has little effect on the pattern. Then the pattern appears random and dispersed in spite of the correlation. When $s\rightarrow 0$, the process is dominated by the adsorption of surface and the correlation between particles. With α increasing, there exists the transition from the random pattern to dense one.

B. Fractal dimension of the clusters

The geometric property can be expressed by fractal dimension D_f. The fractal dimension of the cluster consisting of adsorbed particles can be calculated by the box-counting method [3]. It has been found, for a low occupied fraction, apparent fractal behavior was observed between physically relevant cutoffs in a system of random sets in a box [27]. The lower cutoff r_0 is presented by the length of particles. The upper cutoff r_1 is given by the average gap between adjacent particles [27]. The calculations below were completed by this method. The results are plotted in Fig. 3.

The results for $s=0$ are shown in Fig. 3(a). As α increases, the fractal dimension D_f increases from small value up to 2 (for $p_s=1$) or 1.89 (for $p_s=0.593$) and the small values correspond to the occupied fraction ϕ and p_s. These values are just the expected results, stated below. When the correlation is weak, particles are dispersed randomly on percolation surfaces. These are the pattern of site percolations in percolation spaces. The fractal dimension D_f is small and related to the occupied fraction ϕ [14,27], as well as the percolation space. The larger ϕ and p_s are, the larger D_f becomes. When the correlation is strong enough, the particles construct Leath percolation cluster ($p_s<1$) or Eden one ($p_s=1$). The pattern is dense growth morphology except that it is limited by the space. Thus, if $p_s=1$, $D_f=2$ [28]. And if $p_s=0.593$, the Leath percolation is at the critical threshold and with the fractal dimension of 91/48 [14].

Figure 3(b) plots the results for $s\neq 0$. It can be seen s has little influence on the D_f for the case that $p_s=1$. But for $p_s\sim p_c$, a little increase of s from zero can result in the great decrease of D_f. In the case of large s and small p_s, D_f keeps small values in spite of the change of α. But for $s\rightarrow 0$, D_f increases with the increase of α. These results can be understood as follows. In the case of large s and small p_s, there exist many defects in surfaces and their absorbing ability is very strong. The process is controlled by the adsorption of defects, so the correlation has little influence on D_f. Due to the dispersed distribution of defects, absorbed particles are

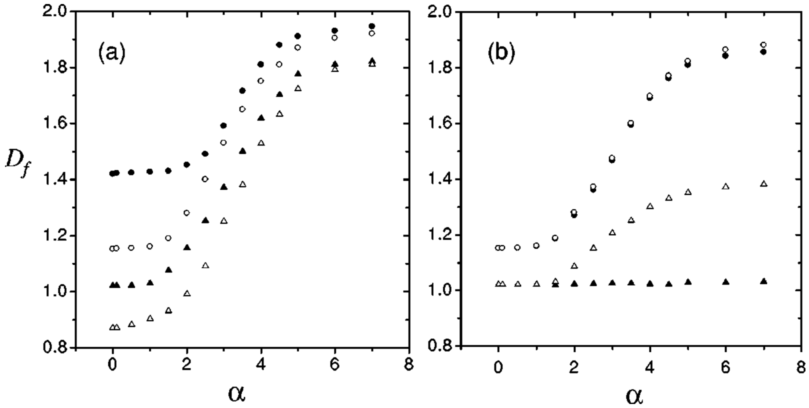

FIG. 3. The fractal dimensions D_f of the clusters consisting of absorbed particles as functions of the correlation exponent α for variant percolation surfaces. (a) $s=0$, $p_s=1$, and $\phi=0.2$ (full circle); $p_s=1$ and $\phi=0.1$ (open circle); $p_s=0.593$ and $\phi=0.1$ (full triangle); $p_s=0.593$ and $\phi=0.02$ (open triangle). (b) $\phi=0.1$, $p_s=1$, and $s=1$ (full circle); $p_s=1$ and $s=0.01$ (open circle); $p_s=0.593$ and $s=1$ (full triangle); $p_s=0.593$ and $s=0.01$ (open triangle).

distributed randomly also. Thus, D_f keeps small values for variant α. But for the case $s\rightarrow 0$, the absorbing ability of the defects is very weak. The absorption of defects has little effect on the pattern. With the increment of α, D_f increases from low values to high ones.

It can be seen from Figs. 1 and 2, there exists the transition from the random dispersed pattern to compact one, which is also reflected by the sharp step in $D_f-\alpha$ curves in Fig. 3. To understand the behavior of D_f with α varying, an analysis is presented on the distribution of new absorbed particles. We consider the absorbed probability P_l of particles at the sites whose distance from the nearest already absorbed particle are larger than a certain value l. Then P_l can be given by

$$\mathrm{P}_l=\sum_{r=l}^{\infty} p(r) \Bigg/ \sum_{r=1}^{\infty} p(r). \tag{4}$$

We use integral instead of summation approximately. Noting that the integral is performed in percolation spaces, Eq. (4) becomes $\mathrm{P}_l=(r^{d_s-\alpha}|_l^{\infty})/(r^{d_s-\alpha}|_1^{\infty})$, where d_s are the fractal dimensions of percolation spaces. As a rough estimate, we obtain that $\mathrm{P}_l\approx 1$ for the range $\alpha<d_s$, $\mathrm{P}_l\approx l^{d_s-\alpha}$ for the range $\alpha>d_s$; and in the case $\alpha=d_s$, $\mathrm{P}_l=\lim_{\alpha\rightarrow d_s}(r^{d_s-\alpha}|_l^{\infty}/r^{d_s-\alpha}|_1^{\infty})=1$. $\mathrm{P}_l=1$ $(\alpha\leqslant d_s)$ corresponds to that absorbed particles appear everywhere randomly and $\mathrm{P}_l=l^{d_s-\alpha}$ $(\alpha>d_s)$ means the probability with which particles are absorbed reduces exponentially as the distance increases. In consequence, $\alpha_c=d_s$ is the transition point from a random dispersed distribution to a compact one of particles. Since d_s is the fractal dimension of the percolation spaces with probability p_s and the reduction of p_s brings about the decrease of d_s, we can also get the result that α_c decreases with the reduction of p_w. Now we turn to the discussion about the sharp step appearing at $\alpha\approx d_s$ in $D_f-\alpha$ curve of Fig. 3. As mentioned before, α_c decreases with the reduction of d_s. When $\alpha<d_s$, the system belongs to the universality class of random graph with a certain fractal dimension depending on p_s and ϕ. When once $\alpha>d_s$, the particles concentrate locally, and the fractal dimension increases.

IV. CONCLUSION

In this paper, a model of correlated-random sequential absorption on nonuniform surfaces was introduced and investigated by Monte Carlo simulations. Our model gave a variety of patterns including site percolation, correlated site percolation, Leath percolation, Eden cluster, sparse islands, and dense islands for different cases. These results can help us to describe and understand a large class of absorbed and clustering phenomena in nonuniform spaces, such as population distribution in actual cities, cluster formation on nonuniform surfaces, epidemic spreading in real worlds, and so on.

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[1] P. Meakin, *Fractals, Scaling and Growth Far From Equilibrium* (Cambridge University Press, Cambridge, 1998).
[2] A.-L. Barabasi and H. E. Stanley, *Fractal Concepts in Surface Growth* (Cambridge University Press, Cambridge, 1995).
[3] T. Vicsek, *Fractal Growth Phenomena* (World Scientific, Singapore, 1992).
[4] T.A. Witten *et al.*, Phys. Rev. Lett. **47**, 1400 (1981).
[5] Z.J. Tan, X.W. Zou, W.B. Zhang, and Z.Z. Jin, Phys. Rev. E **60**, 6202 (1999); Phys. Lett. A **268**, 112 (2000).
[6] P. Meakin, Phys. Rev. Lett. **51**, 1119 (1983); M. Kolb, R. Botet, and R. Jullien, *ibid.* **51**, 1123 (1983).
[7] Z.J. Tan *et al.*, Phys. Rev. E **62**, 734 (2000).
[8] P. Jensen, Rev. Mod. Phys. **71**, 1695 (1999).
[9] Y. Kozlovsky *et al.*, Phys. Rev. E **59**, 7025 (1999); E. Ben-Jacob *et al.*, Nature (London) **368**, 46 (1994).
[10] M.C. Bartelt *et al.*, Int. J. Mod. Phys. B **5**, 2883 (1991).
[11] J.W. Evans, Rev. Mod. Phys. **65**, 1281 (1993).
[12] N.V. Brilliantov, Y.A. Andrienko, P.L. Krapivsky, and J. Kurths, Phys. Rev. Lett. **76**, 4058 (1996).
[13] G. Kondrat and A. Pekalski, Phys. Rev. E **63**, 051108 (2001).
[14] *Fractals and Disordered Systems*, edited by A. Bunde and S. Havlin (Springer, Berlin, 1991).
[15] H.A. Makse *et al.*, Phys. Rev. E **58**, 7054 (1998); H.A. Makse *et al.*, Nature (London) **377**, 608 (1995).
[16] H.J. Herrmann, Phys. Rep. **136**, 153 (1986).
[17] A. Bunde *et al.*, Phys. Rev. Lett. **55**, 653 (1985).
[18] Z.J. Tan *et al.*, Phys. Rev. E **62**, 8409 (2000).
[19] A. Ordemann, M. Porto, H.E. Roman, and S. Havlin, Phys. Rev. E **63**, R020104 (2001); A. Ordemann, M. Porto, H.E. Roman, S. Havlin, and A. Bunde, *ibid.* **61**, 6858 (2000).
[20] Z.J. Tan *et al.*, J. Phys. Soc. Jpn. **70**, 3251 (2001).
[21] P.L. Leath, Phys. Rev. B **14**, 5046 (1976).
[22] Z.J. Tan *et al.*, Phys. Lett. A **282**, 121 (2001).
[23] H. Larralde, P. Jensen, M. Meunier, and A. Pimpinelli, in *Proceedings of the Materials Research Society, Fall Meeting*, edited by R.C. Cammarata, E.H. Chason, T.L. Einstein, and E.D. Williams (Materials Research Society, Boston, 1996).
[24] In the model in Ref. [23] for island formation on surface, if a particle hits a defect, it will be trapped forever. In the present model, the particle becomes immobile with probability *s*.
[25] P. Meakin, Phys. Rev. A **27**, 1495 (1983); see also in Refs. [1,3]
[26] T.-C. Chang *et al.*, Phys. Rev. Lett. **83**, 1191 (1999); B.G. Liu *et al.*, *ibid.* **83**, 1195 (1999).
[27] D. Hamburger *et al.*, Phys. Rev. E **53**, 3342 (1996).
[28] M. Eden, in *Proceedings of the Berkeley Symposium on Mathematical Statistics and Probability*, edited by J. Neyman (University of California Press, Berkeley, 1961), pp. 223–239; see also in Refs. [1,3].