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Diffusion-limited aggregates grown on nonuniform substrates

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HIGHLIGHTS

- Patterns of DLA grown on nonuniform substrates are studied.
- Incipient percolation clusters of k-mers are used as nonuniform substrates.
- At the p_c, the DLA are asymmetrical and the branches are relatively few.
- The fractal dimension of the aggregates increases as p increases.
- This behavior is discussed in the framework of existing theoretical approaches.

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ABSTRACT

In the present paper, patterns of diffusion-limited aggregation (DLA) grown on nonuniform substrates are investigated by means of Monte Carlo simulations. We consider a nonuniform substrate as the largest percolation cluster of dropped particles with different structures and forms that occupy more than a single site on the lattice. The aggregates are grown on such clusters, in the range the concentration, p, from the percolation threshold, p_c up to the jamming coverage, p_j. At the percolation threshold, the aggregates are asymmetrical and the branches are relatively few. However, for larger values of p, the patterns change gradually to a pure DLA. Tiny qualitative differences in this behavior are observed for different k sizes. Correspondingly, the fractal dimension of the aggregates increases as p raises in the same range $p_c \leq p \leq p_j$. This behavior is analyzed and discussed in the framework of the existing theoretical approaches.

1. Introduction

Fractal growth and aggregation phenomena have attracted considerable interest in the surface science community not only because of its importance for experimentalists but also by the basic theoretical background behind it [1–11]. In particular, it is interesting to notice that the structure of the aggregates strongly depends on the dynamics of the growth process as well as the topology of the substrate where they are grown [12]. In fact, great efforts have been made to develop theories and experiments capable of predicting the connection between cluster geometry and aggregation processes. Thus, exact analytical calculation describing the growth process has been developed only for simple cases. However, analytical expressions cannot be derived in more realistic cases and Monte Carlo simulations have proven to be an adequate and powerful tool to study the problem.

T.A. Witten Jr. and L.M. Sander [1] have built up one of the most studied models of surface grown: the diffusion-limited aggregation (DLA). The interest in this model is based on the essential role that the phenomenon plays in many

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experimental situations, such as electrodeposition, fluid–fluid displacement (viscous fingering), dielectric breakdown, chemical dissolution, just to name a few. As a consequence, an increasing interest has also been devoted to enhance our understanding of the theoretical basis of growing processes [1,5–10,13,14].

Most of the contributions dealing with DLA consider the fractal aggregate growing over a homogeneous surface. In contrast, considerable less attention has been paid to the study of the influence of heterogeneous substrates on the formation of DLA clusters [12,15]. It has been an old idea to consider the heterogeneous surface where DLA are grown to be the percolation cluster [16–18]. This line of thinking has been motivated by the fact that percolation clusters are good models for random porous media and DLAs have been connected to miscible displacement of one fluid by another in such media [19]. In fact, the growth of DLA on percolation clusters has been studied by numerical simulations in several seminal contributions [19–21]. In addition, theoretical predictions based on mean fields has also been established. In both cases, the percolating species forming the percolation cluster is allowed to occupy only a single site of the lattice. Recently, the influence of surface heterogeneities on the formation of DLA has been studied as aggregates are formed on, (a) one of the simplest disordered surfaces, the patchwise heterogeneous surface [22,23] and (b) Leath percolation substrates [24].

On the other hand, the pure site and the pure bond percolation of polyatomic species have been studied by using Monte Carlo simulations [25–27]. In both cases, the dependency of the percolation threshold with the size of the element deposited was discussed. In Ref. [26], it was established that the percolation threshold exhibits an exponentially decreasing behavior when it is plotted as a function of the size of the percolating species. However, very recently, Tarasevich et al. [27] have shown a nonmonotonic behavior of the percolation threshold with the size of linear polyatomic species. Nevertheless, the problem belongs to the random percolation universality class regardless the size of the percolating object.

The main aim of the present paper is to study the growing process of diffusion-limited aggregation on substrates formed by percolation clusters of particles which occupy more than one single site. These results will be helpful in understanding the formation of clusters on nonuniform substrates in thin-film-growth processes, such as vapor deposition, molecular-beam epitaxy, and so on.

In detail, the paper is organized as follows. In Section 2 the description of the substrate used for growing the DLA will be presented. The numerical procedure to produce a pattern of DLA is introduced in Section 3, along with the definitions of the critical exponents. The results of the numerical analysis are shown in Section 4, where the connections between the fractal dimension of the DLA and the critical parameters of the substrate are discussed in terms of well known relationships [28]. Conclusions are drawn in Section 5.

2. The nonuniform substrates

Let us consider a periodic square lattice of linear size L on which k-mers (a k-mer is an object composed of k identical particles each one occupying one site of the lattice) are deposited at random. For different k-mers, one must take care that the ratio L/k remains constant in order to prevent spurious finite size effects. The procedure is as follows. A k-tuple of nearest neighbor sites is randomly selected; if it is vacant, the k-mer is then adsorbed on those sites. Otherwise, the attempt is rejected. In any case, the procedure is iterated until N k-mers are irreversibly adsorbed and the desired concentration (given by $p = (kN)/L^2$) is reached. Notice that due to the already randomly adsorbed particles blocking the area, the limiting concentration or *jamming coverage*, p_j is less than that corresponding to the close packing ($p_j < 1$). In other words, p_j is defined as the maximum value of the concentration where there are no gaps available for deposition of particles of size k. The jamming coverage depends on k and exponentially converges to an asymptotic value as $k \to \infty$ [29,30].

The central idea of the percolation theory is based in finding the minimum concentration p for which a cluster (a group of occupied sites in such a way that each site has at least one occupied nearest neighbor site) extends from one side to the opposite one of the system. This particular value of the concentration rate is named *critical concentration* or *percolation threshold* and determines a phase transition in the system. In the random percolation model, a single site is occupied with probability p. For a precise value of p_c, the percolation threshold of sites, at least one spanning cluster connects the borders of the system (indeed, there exists a finite probability of finding n (> 1) spanning clusters [31–34]). In that case, a second order phase transition appears at p_c which is characterized by well defined critical exponents.

In Ref. [27], has been reported how the percolation threshold (linear k-mers) depends on k. For small values of k, the curve rapidly decreases for values of k less than 16 while this behavior changes for larger values. This nonmonotonic behavior of the percolation threshold as a function of the segment size can be associated to the following function:

$$p_c(k) = a_0/k^{\alpha_0} + b \log_{10} k + c \tag{1}$$

being $a_0 = 0.36 \pm 0.02$, $\alpha_0 = 0.81 \pm 0.12$, $b = 0.08 \pm 0.01 c = 0.33 \pm 0.02$.

As it was mentioned, the largest cluster in the range $p_c \leq p \leq p_j$ is isolated and used as a substrate for the growing process. Then, it is important to know that the main features of such a substrate are reflected in the behavior of critical exponents. For this purpose, the fractal dimensions d_f^c and d_w^c defined below are studied.

The fractal dimension, d_f^c, of the largest cluster in the lattice can be defined as [35]:

$$s \propto \rho^{d_f^c} \tag{2}$$

where s represents the number of elements belonging to the studied cluster deposited in a circle of radio ρ.