Analytical model for a cooperative ballistic deposition in one dimension

M. Kamrul Hassan,[1,2] Niels Wessel,[1] and Jürgen Kurths[1]
[1]*Department of Physics, University of Potsdam, Postfach 601553, D-14415 Potsdam, Germany*
[2]*Theoretical Physics Division, Department of Physics, University of Dhaka, Dhaka 1000, Bangladesh*
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We formulate a model for a cooperative ballistic deposition (CBD) process whereby the incoming particles are correlated with those already adsorbed via attractive force. The strength of the correlation is controlled by a tunable parameter a that interpolates the classical car parking problem at $a=0$, the ballistic deposition at $a=1$, and the CBD model at $a>1$. The effects of the correlation in the CBD model are as follows. The jamming coverage $q(a)$ increases with the strength of attraction a due to an ever-increasing tendency of cluster formation. The system almost reaches the closest packing structure as $a\rightarrow\infty$ but never forms a percolating cluster, which is typical of one-dimensional systems. In the large a regime, the mean cluster size k increases as $a^{1/2}$. Furthermore, the asymptotic approach towards the closest packing is purely algebraic both with a as $q(\infty)-q(a)\sim a^{-1/2}$ and with k as $q(\infty)-q(k)\sim k^{-1}$, where $q(\infty)\simeq 1$.

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I. INTRODUCTION

The kinetics of a monolayer growth by the deposition of macromolecules and colloidal particles onto solid substrates has been the subject of extensive research for the recent years (see Refs. [1–3] for extensive review). The reason is well justified because its importance and significance cover many seemingly unrelated topic in physics, chemistry, biology, and other branches of science and technology. From a theoretical point of view, the random sequential adsorption (RSA) of a monodisperse particle is one of the simplest models that can describe deposition phenomena [4]. In this process, particles are deposited randomly, one at each time step, with the strong restriction that overlapping is forbidden. This can be described by the following algorithm. (i) At each time step, a random position is chosen from the whole substrate and is assigned to the center of the particle picked for deposition. (ii) If the incoming particle collides with a previously adsorbed one, the trial attempt is rejected; otherwise it is adsorbed irreversibly. (iii) In either case, the time is increased by one unit and the steps (i) and (ii) are repeated until the system reaches a state when particles can no longer be adsorbed. Alternatively, a recursive algorithm can be used if one is not interested in the kinetic aspects of the process. The rules for the recursive algorithm are the same as that for the sequential deposition, except the rule (i) which is replaced by the following rule. The number of particles that make an attempt for adsorption at the nth step are 2^n.

One of the virtues of the RSA model is that like many statistical physics problems, it is exactly solvable in one dimension in both its continuum and lattice versions for some specific cases. The continuum version of the model in one dimension (1D) is popularly known as the *random car parking* problem and has attracted much attention. Despite its inherent simplicity, it still captures the essential generic features of the process and has proved to describe successfully the behavior of many experimental systems, namely, the adsorption of proteins, latex, and colloidal particles [5–7]. Nevertheless, there have been continuous research efforts to include various important physical features to make it more realistic and thus covering a wider range of real life situations [8–10]. Along this road, a good deal of progress has already been achieved and yet we are far away from a complete theory. In recent years it has received extra momentum and the number of papers published is a clear testimony to this [3,11–15].

The most distinctive feature of the RSA model is its outright rejection of the particles that fall on an already adsorbed one. This outright rejection criterion has partially been lifted by the ballistic deposition (BD) model proposed by Talbot and Ricci [16,17], which is best explained in terms of the deposition of disks of diameter m instead of a line segment [16,17]. It is worth mentioning that the landing point in the BD model is chosen randomly over the entire line exactly in the same way as is done for the classical RSA model. The only difference between the two models lies in the fact that in the BD model whenever an incoming disk overlaps an already adsorbed one, it is allowed to roll over the latter disk following the path of the steepest descent, whereas in the RSA model this is rejected. In allowing such rolling mechanism, the disk can either touch the the global minimum (adsorbing plane) or it may find itself trapped in the local minimum formed by two or more connected disks. In the former case, the disk is irreversibly attached to the one it rolled over leaving no gaps in between, while in the latter case the trial attempt is rejected. Both, in the simple RSA and in the BD model, only a short-range hard-core repulsion via the excluded volume effects is taken into account. All forms of long-range interactions between the particles in the adsorbed and adsorbing phases are ignored. There are some fragmented attempts, though, to include some specific forms of interactions such as the electrostatic, dipolar, and the hydrodynamic interactions [18–20].

In this paper, we consider a model that includes the attractive force between the particles in the adsorbing and the adsorbed phase mimicking the long-range interaction. In order to increase the flexibility of the model, we introduce a parameter a that can tune the strength of the attractive forces. This would certainly facilitate the study of the general effect of the long-range interactions in the whole process. We for-

mulate the model in such way that we still can recover the simple RSA results by setting $a=0$, the BD results by setting $a=1$, and the mixture of the two (RSA and BD) for $0<a<1$ when a describes the probability with which the particle that falls onto an already adsorbed one decides to roll over it. On the other hand, for $a>1$, we show that the model describes the cooperative ballistic deposition (CBD) instead of describing the generalized BD model as studied in Ref. [21]. The strength of the correlation in the CBD model is determined by the strength of the attractive force. Note, though, that once a particle collides with an already adsorbed one, it follows the rule of the simple BD and hence we call it the cooperative BD model. Thus, as the strength of interaction increases, we expect an increasing rate of successful adsorption via the roll-over mechanism. This results in a decrease in the density of gaps and in a higher coverage.

Interestingly, the rate equation that governs the dynamics of the CBD model appears to be similar to that for the generalized BD model studied by Viot *et al.* [21]. However, the physical interpretation of a, in the CBD model, is completely different from that offered by Viot *et al.*, where it is taken as the ratio of probabilities corresponding to two mutually exclusive events. In fact, we show that one cannot define a as the ratio of two such probabilities. In addition, we give an exact analytical solution to the CBD model considering the sequential deposition, that is, at each time step only one trial attempt is made for deposition. To further support our theory and to gain insightful information on this work, we then use the recursive algorithm to solve the present problem numerically where one trial attempt is made for every gap, i.e., the number of particles that make attempt to be adsorbed at the nth step are 2^n. This proved fine for all the aspects of the problem such as the jamming coverage, number density of the gaps, and their relations with the strength of attractive force except for the kinetic aspects of the problem. Interestingly, the two processes, namely, the sequential deposition and the recursive algorithm, helped to understand the problem better than it would have been otherwise.

The rest of the paper is organized as follows. First, we give a general explanation of the model and a means to translate it into a simple and well-known BD model. Our ideas are backed up and well supported by direct numerical simulations, which led to a better understanding of the physical nature of the system. Second, we present extensive results showing the asymptotic approach of the coverage towards the jamming limit in terms of the various parameters involved in the processes.

II. COOPERATIVE BALLISTIC DEPOSITION MODEL

We consider a system that consists of a reservoir of particles having diameter m lying in the immediate vicinity of a 1D substrate. The adsorbing particles we may consider to be in the gas or in the fluid phase and arrive in the adsorbing plane through the Brownian motion. As soon as a particle comes into contact with a gap large enough to accommodate it, it is then adsorbed immediately and irreversibly. On the other hand, the incoming particle that touches an already adsorbed one, is allowed to follow the BD rules to form a monolayer. However, for clarity reason, we postpone discussions on how we take into account the attractive interaction. Perhaps it is worth mentioning that any trial attempt that results in the rejection in anyway may be considered to be bounced back to the bulk and may lose the memory of its history. Therefore, it may mimic the deposition via the Brownian motion.

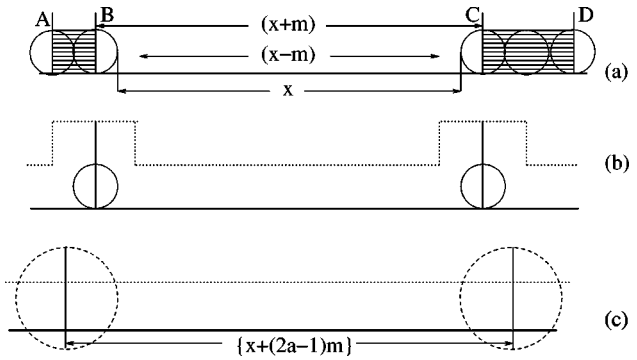

FIG. 1. Schematic illustration of the model in different situations. The shaded regions between AB and CD in (a) have local minimums and play no other role except for kinetic reasons. We therefore eliminate all such shaded regions so that we have a system where all the gaps are separated from their neighbors by only one disk as shown in (b). The dotted lines in (b) represent the probability distribution in different regions of a given gap. The steepest descent path in (c) is artificially increased by using the idea of a virtual disk at the expense of lowering the height of probability distribution so that every point of $x+(2a-1)m$ is now equally likely to be chosen by the trial attempt.

The simplicity of the 1D problem lies in the fact that every successful deposition of a particle on a given gap divides it into smaller gaps having the same geometry as the parent gap. It is this *shielding property*, found only in 1D, that we shall use to gain further insight into the problem and to tackle it analytically. For the sake of simplicity, we assume that the daughter gaps are uncorrelated, irrespective of the island size separating the gaps from their neighbors, so that we can treat each gap as an independent entity. We further assume that each roll-over motion is completed prior to the next trial attempt for deposition.

At this point, it is useful to discuss first the classical RSA and the simple BD model before introducing the cooperative BD model. In the classical RSA, an incoming particle of size m is adsorbed successfully in a gap of size x, if the center of the incoming particle arrives in any place but $m/2$ away from either ends of a gap. This means that only $(x-m)$ of a given gap of size, say x, is accessible for adsorption, which we have illustrated in Fig. 1(a). In the BD model, on the other hand, those particles that fall on an already adsorbed one, may reach the substrate via the roll-over motion. The deposition via rolling is successful if the center of the incoming particle falls within a distance of $m/2$ on either side of both ends, otherwise it is rejected. It is then adsorbed on the respective edge creating a new gap of size $(x-m)$. That is, for a given gap x, the total position accessible to a new arrival is $(x+m)$, which is shown in Fig. 1(a). That is, any point of size $x+m$ can be occupied with an equal probability. Note

that any particle dropping at any point within the shaded regions AB or CD are, in fact, trapped due to the local minimum. These particles will never reach the global minimum or the adsorbing substrate and hence they may be considered to be rejected. For the sequential deposition, when the outcome by a trial attempts results in rejection, the time step is increased by one unit [11]. However, if we are not interested in the temporal or the kinetic aspect of the system, we can safely delete the shaded regions as if these did not exist, since they play no role in determining the jamming coverage or the number density of gaps. In such a case, we may assume that the neighboring gaps are separated by only one disk as shown in Fig. 1(b). We can thus define each gap as an independent isolated interval bordered on either end by a semidisk so that if we connect the two remote ends they would then form a ring with one particle at the joint.

We are now in a position to introduce the long-range attractive force among the incoming particles and the particles in the adsorbed phase. We may assume that the particles are still in the gas or in the liquid phase and that they arrive on the substrate through the Brownian motion. However, in the present problem, each adsorbed particle attracts the incoming particles towards its center. The question is how can we incorporate such attractive force in a tractable way? First, we need to understand the effects of such an attractive force. The most significant one is that each adsorbed particle will tend to attract the incoming particle towards it. This would immediately break the nature of the uniform probability density of the problem as is the case for the BD model since the incoming particles are more likely to land on an already adsorbed one than on a gap. For the sake of simplicity, we consider a square-well-type attractive force of width $2m$ around the center of each adsorbed particle. We therefore have two distinct probability distribution (PD) regions as indicated in Fig. 1(b). First, the *force-free region* $(x-m)$, where the strength of the attractive force is strictly zero. Second, the *force field region* of width $2m$ about the center of each adsorbed particle, where the adsorbed particle exerts an attractive force on the incoming particles. As a result, any point in this region is more likely to be selected than the points in the force-free region, which is indicated by the dotted lines in Fig. 1(b). The flat PD implies that all the points within the respective regions have the same *a priori probability*.

It is worth mentioning that the simple BD model refers to the case where both regions have the same height in PD, which is flat in nature and hence the whole substrate represents the zero force field or the zero attractive force. Now, as soon as we switch on the attractive force, the height of the PD around each adsorbed particle of width $2m$ will increase to a degree depending on the strength of the attractive force. We then attempt to translate such a problem into a simple BD model, which is the key to the analytical solvability of the problem. This can be done in the following way. Note that we can lower the height of the PD by increasing its width but keeping the total area unchanged. In this way, we can make the whole system have the same height of the PD and treat it like the simple BD model where an incoming particle can land anywhere with an equal probability including the disks that are already occupied as represented in Fig. 1(c). In other words, we can describe it as follows. Prior to selecting a position for an incoming particle, we replace the size of the deposited particles used for defining the gaps by a virtual disk of diameter $R=(2a-1)m$, without altering the gap size. In this way, we hypothetically enlarge the cross section of collision of an incoming particle with an already adsorbed one. Notice the role of the attractive force for which there will be an extra number of incoming particles to fall on an already adsorbed particle. The increased cross section $2m(a-1)>0$ provided $a>1$ would count exactly the same number of additional collisions as it would have been by the potential in question. Once the position for the next trial deposition is chosen, we can immediately return to the system with disks having diameter m and proceed according to the simple BD rules as depicted in Fig. 2. What we have done is that we have artificially increased the probability with which an incoming particle may collide with an already adsorbed disk, thus enhancing the probability of adsorption via the rolling mechanism and mimicking the effect of the attractive force. One can thus expect an enhanced adsorption probability near the two extreme ends of each gap as the virtual disk size increases and, in the limit $R\to\infty$, we can only expect the adsorption via only the rolling mechanism except in the very early stage where the virtual diameter and the gap size may be of the same order in size.

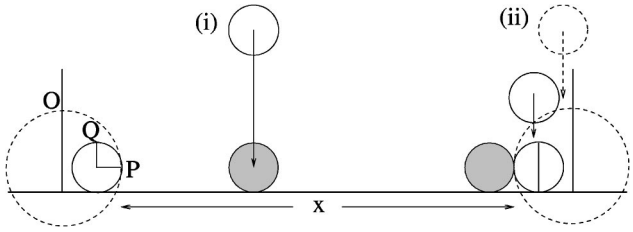

FIG. 2. Schematic illustration of the rules for the cooperative ballistic deposition model. The incoming disk is adsorbed directly as shown in (i) if it is dropped within $(x-m)$. The deposition via the rolling mechanism is depicted in (ii). Here, any disk that falls on the steepest descent path of the virtual disk, OP, is assumed to be dropping on its corresponding equivalent point of the real particle QP from where it can successfully reach the global minimum.

III. ANALYTICAL SOLUTION OF THE SEQUENTIAL CBD MODEL

To address the CBD model analytically, we adopt the well-studied rate equation approach of the gap size distribution function or concentration $c(x,t)$. The quantity $c(x,t)dx$ is defined as the number of gaps at time t in the size range between x and $x+dx$. Within this rate equation approach, it is beyond our scope to add the particle-particle interaction of any type directly into the rate equation as it is based on the master equation, which is typical of nonequilibrium statistical physics. We therefore use the possible effect of such interaction and scale it in terms of the size of the depositing particles. The most immediate consequence of the attractive interaction as we already mentioned is that the incoming particles will be more likely to fall on an already adsorbed one than on any gap. Let us assume now that the depositing

particle, which is adsorbed successfully on the substrate, instantaneously grows to a disk of a larger size having diameter $(2a-1)m$. This happens without occupying any space of the substrate. That is, the extra spaces $2(a-1)m$ required for the growth of the deposited particles are assumed to be hypothetical as they do not destroy the size of the gap created by each deposition event. The whole thing happens in such a way, as if, only the incoming particles see such growth and hence plays no role in calculating the coverage and the number density of the gaps. The larger they grow, the larger is the steepest descent path, which will essentially lead to a higher number of particles falling on the particles those are already adsorbed. The excess size $2(a-1)m$ thus takes care of the attractive force in an appropriate manner. The kinetics of adsorption of the monodisperse particles can then be described by the following set of rate equations:

$$\frac{\partial c(x,t)}{\partial t} = -(x-m)c(x,t) + 2\int_{x+m}^{\infty} c(y,t)dy + 2am\{c(x+m,t)-c(x,t)\} \tag{1}$$

for $x \geqslant m$ and

$$\frac{\partial c(x,t)}{\partial t} = 2\int_{x+m}^{\infty} c(y,t)dy + 2amc(x+m,t) \tag{2}$$

for $x<m$. The above rate equations are mean field in nature as the fluctuations and correlations are ignored. The rate equation approach is based on the assumption that the creation and annihilation of gaps are independent of the size of the neighboring gaps. The first two terms of Eq. (1) and the first term of Eq. (2) are the same as that of the simple RSA process and thus describe the creation and destruction of a gap of size x due to the direct adsorption of size m on size $y \geqslant x+m$ or on size x, respectively. The remaining terms in both equations also describe the creation and destruction of gaps but via a rolling mechanism following the steepest descent path allowing to travel the maximum linear distance am, where a is a dimensionless constant number that we can tune. The factor 2 in the integral terms accounts for the fact that any of the two new gaps created upon a direct deposition on the gap size $y \geqslant x+m$ can be of size x; whereas the same factor in the remaining terms takes into account that a gap of size x can be created or destroyed from either end by adsorption. In order to understand the role of a, it is convenient to rewrite Eq. (1) as

$$\frac{\partial c(x,t)}{\partial t} = -\{x+(2a-1)m\}c(x,t) + 2\int_{x+m}^{\infty} c(y,t)dy + 2amc(x+m,t). \tag{3}$$

The $\{x+(2a-1)m\}$ term in the above equation is the key to understand the role of a. Note that by setting $a=0$ we recover the classical RSA case where $(x-m)$ of a given gap x is accessible for adsorption, which is consistent with our discussion in the preceding section. Similarly, $a=1$ describes the simple BD model where the total positions accessible to a new arrival are $(x+m)$ which is again consistent (see Fig. 1). However, for $a>1$ $\{x+(2a-1)m\}$ means that a given gap of size x is bounded by at least two semidisks of diameter $R=(2a-1)m$ while the adsorbing particles are of size $m<R$. That is, any particle that falls within a distance am from either end of $\{x+(2a-1)m\}$ effectively will collide with the virtual disk. Every point of the steepest descent of the virtual path OP in Fig. 2 has its corresponding equivalent point on the real path QP. Therefore, an incoming particle falling on the virtual path is assumed as if it were falling on the exact equivalent position of the real path and vice versa.

To solve Eq. (1) we seek a trial solution of the following form

$$c(x,t) = A(t)e^{-(x-m)t}, \tag{4}$$

where $A(t)$ is still an undetermined quantity fixed by the initial condition. Let us assume a monodisperse initial condition $c(x,0)=\delta(x-L)/L$, so that we have

$$\lim_{L\to\infty}\int_0^L c(x,0)dx = 0, \quad \lim_{t\to 0}\int_0^{\infty} xc(x,t)dx = 1. \tag{5}$$

Substituting the trial solution into Eq. (1), we obtain the following differential equation for $A(t)$:

$$\frac{d\ln A(t)}{dt} = \frac{2e^{-mt}}{t} + 2ae^{-mt}. \tag{6}$$

Solving it, satisfying the initial conditions, we get

$$A(t) = t^2 F(a,mt), \tag{7}$$

where the auxiliary function $F(a,mt)$ is defined as

$$F(a,mt) = \exp\left[-2\int_0^{mt}\frac{1-e^{-u}}{u}du + 2a(1-mt-e^{-mt})\right]. \tag{8}$$

To obtain $c(x,t)$ for $x<m$, we substitute the solution of Eq. (1) into Eq. (2) and then upon a direct integration, we get

$$c(x,t) = 2\int_0^t u(1+amu)F(a,mu)e^{-xu}du. \tag{9}$$

The solutions $c(x,t)$ can provide a complete analytical description of the process including its kinetic aspect. All we need now is to find useful ways of using these solutions for computing various physical quantities of interest such as the jamming coverage, the mean number density, the mean cluster size, etc.

IV. NUMERICAL SIMULATION OF THE RECURSIVE CBD MODEL

To test the physical description of our CBD model, we have simulated it on a computer. One obvious constraint of the simulation is, of course, the finite-size effect. However, for a sufficiently large substrate in comparison to the depositing particles, the finite-size effect can be made sufficiently

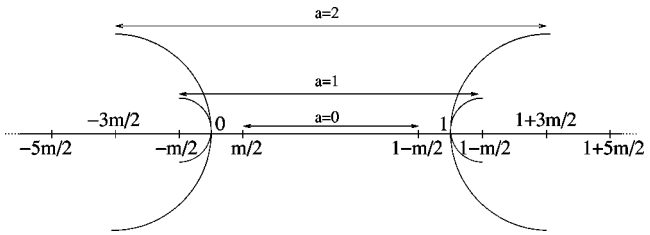

FIG. 3. Schematic illustration of the recursive simulation scheme of the CBD model.

small. To simplify the simulation, we use the approximation mentioned in the preceding section that the gaps are uncorrelated and can be treated independently. Furthermore, to simplify the problem we use a recursive algorithm. The simplicity of the problem is achieved at the expense of the kinetic aspect of the system. In brief, the description of the recursive scheme of the simulation is as follows. Let us assume that the initial gap of unit interval $[0,1]$ is bordered on both ends by semidisks of radius $(2a-1)m/2$ as shown in Fig. 3. We then generate a random number n from the interval $[-(2a-1)m/2,1+(2a-1)m/2]$ to assign to the center of the incoming particle. The incoming particle is then adsorbed directly, creating two new smaller intervals if $n \in [m/2,1-m/2]$. Otherwise, the disk is adsorbed on one of the edges of $[0,1]$ creating only one new gap of interval $[m,1]$ if $n \in [-(2a-1)m/2,m/2]$ or of interval $[0,1-m]$ if $n \in [1-m/2,1+(2a-1)m/2]$, respectively. In the case when the disk is adsorbed in one of the two edges, we increase the counter that provides the information on the mean cluster size of the system. We then continue the process assuming each new gap is again bordered by virtual semidisks and treat them in the same way as the first step until we have no more gaps of size $\geqslant m$. The number of particles thus deposited at the nth step is simply 2^n. Finally, we add all the gaps of size less than m and, using this, we can immediately calculate the jamming coverage and the mean cluster size. We have performed the simulation with substrate size $\sim 10^6 m - 10^8 m$ within the interval $[0,1]$ and found an excellent match with the corresponding analytical results up to several digits. We also noticed that increasing the substrate size by decreasing the m value only contributes to a higher order precision as expected. The simulation results are averaged over 500 different realizations and are shown by the symbols $(\times)$ along with the theoretical results shown by solid lines in various plots where appropriate.

V. RESULTS AND ANALYSIS

It is worth mentioning here that only the adsorbed particles are enlarged keeping the size of the gaps unchanged. The fraction of the line covered by the adsorbing particles or the coverage $\theta(a,t)$ at different instants of time can therefore be defined as

$$\theta(a,t) = 1 - \int_0^\infty x c(x,t) dx. \tag{10}$$

The number density of gaps, on the other hand, is defined by the following relation:

$$N(a,t) = \int_0^\infty c(x,t) dx. \tag{11}$$

However, we find it more convenient to handle their rate equation rather than their definition itself. The kinetic equation for the coverage is

$$\frac{d\theta(a,t)}{dt} = m \int_m^\infty \{x + m(2a-1)\} c(x,t) dx = \Phi(a,t). \tag{12}$$

Here, quantity $\Phi(a,t)$ is the fraction of the substrate accessible to a new particle at a given time t. The kinetic equation for the number density, on the other hand, is

$$\frac{dN(a,t)}{dt} = \int_m^\infty (x-m) c(x,t) dx. \tag{13}$$

The above two equations can be combined together to obtain

$$\frac{d\theta(a,t)}{dt} = m \frac{dN(t)}{dt} + 2am \int_m^\infty c(x,t) dx. \tag{14}$$

Note that, in the simple RSA, one gap corresponds to one particle and thus we have $\theta(0,t) = mN(0,t)$ reflecting the fact that the average particle size is the same as the size of the adsorbing particles. However, in the present case, for $a > 0$, the second term of the above equation describes the cluster formation. That is, the first term on the right-hand side of Eq. (13) takes into account the direct deposition while the effect of the rolling mechanism is described by the second term. Using the solution for the appropriate boundary in Eq. (12) yields

$$\theta(a,t) = \int_0^{mt} F(a,u)(1+2au) du. \tag{15}$$

This can provide all the information about kinetic aspects of the process, namely, how the coverage evolves in time. One of the characteristics of the deposition process is that the system reaches a state of deadlock in a finite time when particles can no longer be adsorbed. This is typically known as the jamming limit and the exact critical time to reach such a state should depend on a. However, to date there do not exist any theoretical means to pin down the exact critical time for reaching the jamming limit. Nevertheless, we can safely calculate the coverage in the jamming limit as

$$q(a) = \lim_{t \to \infty} \theta(a,t). \tag{16}$$

The jamming coverage has been of special interest in the study of the deposition phenomena as it can uniquely characterize the structure of the resulting monolayer. From the exact expression for the coverage, Eq. (15), it has been of interest to know how the coverage $\theta(a,t)$ approaches the corresponding jamming limit $q(a)$. We find that beyond the

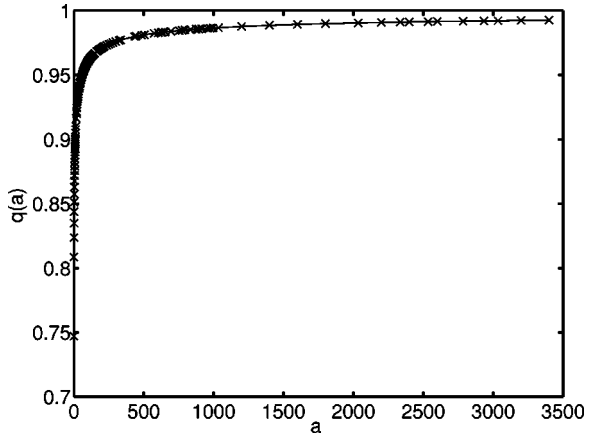

FIG. 4. Jamming coverage as a function of a that measures the strength of attraction: $q(a)$ vs a.

transient behavior, the system reaches its asymptotic coverage, namely, the jamming limit exponentially, with a decay constant $2a$, multiplied by an algebraic prefactor t^{-1}

$$q(a)-\theta(a,t)\sim t^{-1}e^{-2at}, \tag{17}$$

which was also reported in Ref. [21]. Obviously, for the classical RSA ($a=0$), we recover the power-law behavior, which is also known as Feder's law [22]. Here for $a>0$, the exponential approach towards the jamming limit reflects the fact that the increasing number of particles that land on an already adsorbed particles are successfully accommodated via rolling. Another interesting point to check is how the jamming limit varies as we increase the strength of interaction a. In other words, we want to see how the jamming limit changes as we increase the degree of correlation between the particles in the adsorbed and adsorbing phases. Figure 4 shows a sharp rise in the jamming coverage at low a and a slow rise towards the closest packing in the large a regime. In an attempt to quantify the slow regime we plot $\ln[q(\infty)-q(a)]$ against $\ln(a)$ in Fig. 5 and find that the jamming

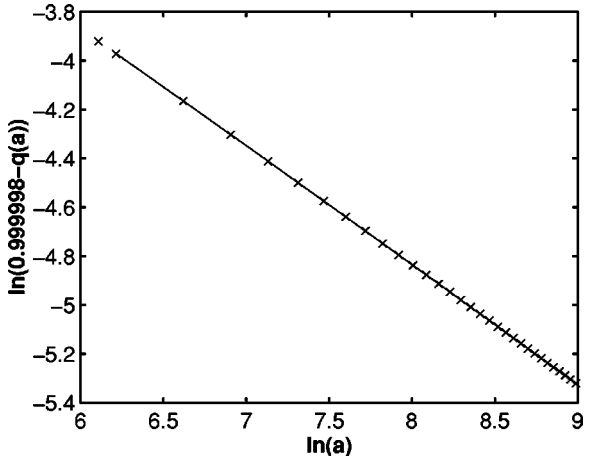

FIG. 5. The linear fits of $\ln[q(\infty)-q(a)]$ vs $\ln(a)$ having slope $-1/2$ in the large a regime reveals that the convergence of the jamming coverage towards the closest packing is power law in nature.

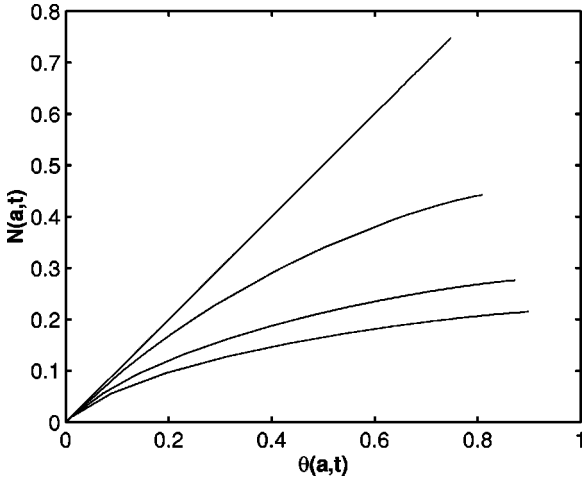

FIG. 6. Time dependence of the number density as a function of the coverage. The curves from top to bottom represents $N(a,t)$ vs $\theta(a,t)$ for $a=0$, 1, 5, and 10.

coverage converges towards the closest packing obeying a power law

$$q(\infty)-q(a)\sim a^{-1/2}, \tag{18}$$

where $q(\infty)\approx 1$. It is important to note here that the system never reaches a complete closest packing ($q=1$) even for $a\to\infty$. This is due to the fact that the substrate size too is of the same order as that of the size of the virtual disk, hence there is always a nonzero probability for a direct deposition at least in the early stage. We attempted to check it in the computer choosing both the virtual diameter and the initial substrate size to be of the same order and large enough to minimize the finite-size effect. This is exactly the case described by the analytical model as we let $a\to\infty$. In doing so we never find a cluster covering the whole substrate. We checked it over and over again by increasing the substrate size and the virtual disk size up to $\sim 10^{10}m$. Nevertheless, neither the analytical solution nor the simulation could give us an exact estimate for $q(\infty)$.

We now intend to obtain an exact expression for the number density by substituting the solution $c(x,t)$ for $x>m$ into Eq. (13), which yields

$$N(a,t)=\frac{\int_0^{mt}F(a,u)du}{m}. \tag{19}$$

The above relation for the number density immediately implies that it depends on the size of the adsorbing particles. Here it is worth mentioning that the mean number density for the classical RSA ($a=0$) is simply the coverage divided by the size of the adsorbing particles. In this case, the mean number density increases linearly with the coverage as time proceeds, having a slope m during the process (see Fig. 6). However, as soon as a particle that fell on a previously adsorbed one can roll over the latter, the linear relation between $N(a,t)$ and $\theta(a,t)$ is immediately ceased and it is replaced by a nonlinear relation (see Fig. 6). Therefore, the mere

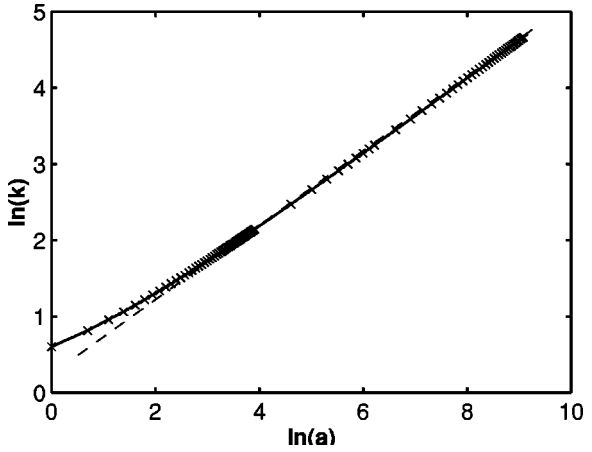

FIG. 7. The linear fits of the plot of ln(k) vs ln(a) in the large a regime with slope 1/2 show that the mean cluster size increases as $\sim a^{1/2}$.

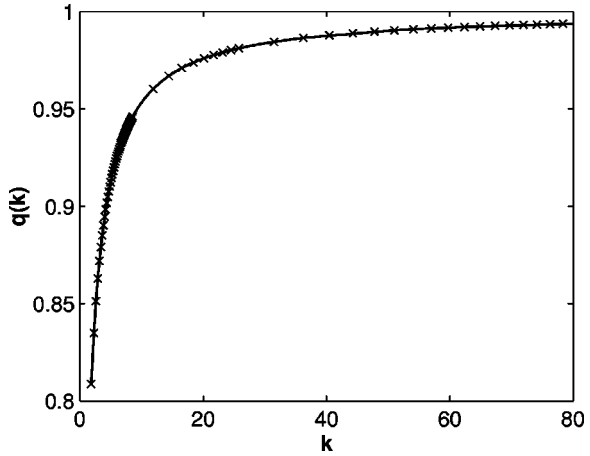

FIG. 8. The jamming coverage as a function of the mean cluster size k.

knowledge of one of the two is not sufficient to obtain the other. This is due to the fact that the mean cluster size is different from the size of the adsorbing particle as the system keeps producing connected clusters of different sizes depending on the value of a. Figure 6 shows that for $a>0$ the number density grows linearly at a very low coverage. This is due to the fact that at an initial stage the incoming particles hardly encounter any preoccupied species and therefore there exists almost no cluster. However, as the substrate gets crowded, it is evident from Fig. 6 that the mean number density increases in a nonlinear manner and the strength of nonlinearity increases with increasing a. Therefore, to obtain the coverage (the number density) from the number density (coverage), we need to know the mean cluster size. The expression for the coverage $\theta(a,t)$ and the mean number density $N(a,t)$ at different instants of time can give us an estimate of how the mean cluster size is defined, i.e.,

$$s(t)=\frac{\theta(a,t)}{N(a,t)} \tag{20}$$

grows in time and with the strength of a. We find that for a given size of the adsorbing species, the mean cluster size in the jamming limit is

$$\lim_{t\to\infty}\frac{\theta(a,t)}{N(a,t)}=s=\frac{\int_0^\infty F(a,u)(1+2au)du}{\int_0^\infty F(a,u)du}m. \tag{21}$$

Obviously, like the mean number density, the mean cluster size should depend on the size of the adsorbing particle. However, the ratio between the two $k=s/m$ remains constant in the jamming limit and therefore it is useful to call it the universal mean cluster size. It is worth mentioning here that for the simple RSA we get $k=1$ and hence the mean cluster size is the same as that of the size of the adsorbing species. However, for $a>0$ we find that the universal mean cluster $k>1$ and k increases monotonically with increasing a. Furthermore, Fig. 7 reveals that in the large a regime the universal mean cluster size k increases with the strength of attractive force as

$$k\sim a^{1/2}. \tag{22}$$

The probability of adsorption of particles without overlapping with any preadsorbed particle for all a decreases with time; however, the strength of such decrease gets sharper and sharper as a increases. In the limit $a\to\infty$ the virtual disk sizes are of the same order as that of the substrate. Therefore, at a very early stage the adsorption of particles by direct deposition or the adsorption by the rolling mechanism have the same probability. Thus, the system never reaches a state of closest packing but of almost closest packing. The jamming coverage thus increases with increasing k (see Fig. 8). We find that like the $q(a)$ vs a, the approach of the jamming limit towards almost closest packing against k also follows a power-law form but with a different exponent. As shown in Fig. 9, the plot of $q(k(a\to\infty))-q(k)$ vs k in the logarithmic scale along both axes is well fitted by a straight line with slope 1 and hence

$$q(k(a\to\infty))-q(k)\sim k^{-1}, \tag{23}$$

where $q(k(a\to\infty))\approx 1$.

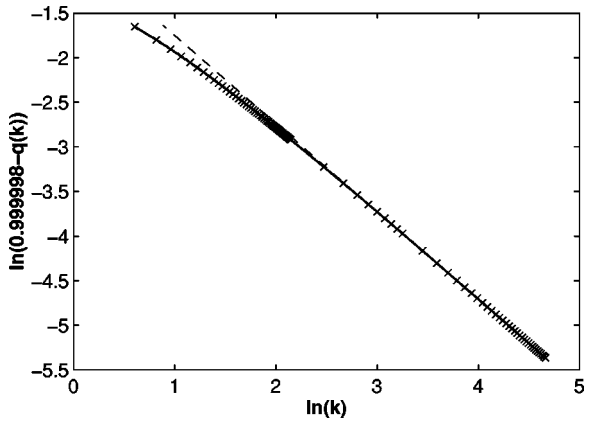

FIG. 9. The linear fits of ln[$q(\infty)-q(a)$] vs ln(k) in the large k regime show that the jamming coverage increases in a power-law form $\sim k^{-1}$.

VI. DISCUSSION

We first discuss various possible interpretations for different regions constituted by different values of a of the CBD model. It is interesting to note that when $a=1$, the size of the virtual and the real disk coincides and hence every point of the substrate bear the same probability of occupation by the incoming particle. In this case, particles that fall on an already adsorbed one may travel up to a linear distance of its own size m via the rolling motion following the steepest descent path. The particle can either be trapped in the local minimum or it can reach the global minimum. In the former case, it is rejected while in the latter case it is adsorbed irreversibly touching that it just rolled over, provided there is at least one gap to accommodate it. However, in either case the time is increased by one step. The case $a=1$ thus describes the well studied BD model. The situation for $0<a<1$ is also interesting as it describes a mixed process composed of the simple RSA and the BD process, which, in fact, should be termed as the generalized ballistic deposition processes. In this case the a parameter describes the probability with which the adsorbing particle decides to roll over that it falls on to. Such a rolling mechanism can be assumed to be due to the gravitational pull towards the adsorbing surface. The above two situations together, i.e., $0<a<1$, constitute the constituents of the generalized ballistic deposition processes. However, for $1<a<\infty$ the basic rules are still the same as for the simple BD model, but it also incorporates a square-well-type attractive interaction between the elements of the adsorbed phase and the incoming particles.

It is worth mentioning that Viot *et al.* made an attempt to generalize the ballistic deposition model and gave the following definition [21]. The disks having the same diameter are dropped uniformly and sequentially one at each time step. The disks can either reach the adsorbing plane or fall on an already adsorbed disk. In the former case, the trial attempt is retained with a probability $q=(1-p)$. In the latter case, on the other hand, the trial disk follows the path of the steepest descent over the disk it encountered. The disk is then adsorbed with probability p, provided the particle can reach the global minimum by the roll-over motion; otherwise it is trapped in an elevated position and is rejected. This is exactly the case defined in the mixed processes of both RSA and BD (i.e., the $0<a<1$ regime). However, Viot *et al.* then defined the a parameter as $a=p/q$. There is a clear mismatch between the definition of the model in words and the definition of a. We would like to point out anyway, that one cannot define $a=p/q$ due to the following reasons. We can rewrite Eqs. (1) and (2) for $a=p/q$ upon multiplying both equations by $(1-p)$.

$$q\frac{\partial c(x,t)}{\partial t}=q\left\{-(x-m)c(x,t)+2\int_{x+m}^{\infty}c(y,t)dy\right\}+2pm\{c(x+m,t)-c(x,t)\} \tag{24}$$

for $x\geqslant m$ and

$$q\frac{\partial c(x,t)}{\partial t}=2q\int_{x+m}^{\infty}c(y,t)dy+2pmc(x+m,t) \tag{25}$$

for $x<m$. If we then set $q=0$ in these equations, we find that the remaining terms alone are incapable of describing any meaningful physical process. The point to emphasize here is that the roll-over mechanism comes after the trial attempt for deposition is made. That is, the trial attempt to deposit a particle is the primary event of the process, which can only be described by the following equation:

$$\frac{\partial c(x,t)}{\partial t}=-\theta(x-m)(x-m)c(x,t)+2\int_{x+m}^{\infty}c(y,t)dy, \tag{26}$$

where $\theta(s)$ is the Heaviside step function The roll-over mechanism can only follow thereafter. The terms describing the trial attempts cannot bear any probability. Therefore, the two events, the direct adsorption and the deposition via rolling, are not mutually exclusive events and hence one cannot define the parameter $a=p/(1-p)$.

The model we have presented in this paper is solved exactly by means of a kinetic equation approach based on sequential deposition. In order for further support, we solved it by the numerical simulations based on the recursive algorithm. The two approaches, sequential and recursive, turn out to be highly rewarding in the sense that these helped not only to confirm the validity of the mean-field approximation, but also to shed a deeper insight into the nature of the problem. The basic principle of the model is the same as that of the simple ballistic deposition process as we mentioned earlier. Nevertheless, we have extended the simple BD by adding a certain degree of correlation between the adsorbing particles and those already adsorbed. To increase the flexibility of the model, we allowed a parameter a that can tune the strength of the correlation which is induced by the attractive force. Instead of using the attractive force directly, we have shown a way of transforming it into a virtual situation, which is then just the simple BD model.

The most significant consequence of the presence of the attractive force is that it results in an increased packing fraction as well as the mean cluster size due to the formation of the higher order connected clusters. Moreover, the jamming coverage increases with the increasing degree of the strength of the attractive force. Similar results have also been recently reported by Pastor-Satorras and Rubi [13], who studied a model of correlated sequential adsorption by numerical simulation both in one and two dimensions. However, unlike the square-well-type PD studied here, they used a Gaussian and exponential-type PD around the center of each adsorbed particle. Nevertheless, despite the apparent differences in the detailed nature of the forces or in the PD, the qualitative behavior seems remarkably identical to what we have found in this paper. Pastor-Satorras and Rubi too observed the same trend of the increase of jamming coverage as well as the mean cluster size. In addition, they too reported the approach towards the closest packing in the limit where the correlation is maximal. This reveals that the qualitative effect of the attractive force is insensitive to the detailed nature of the attractive force. However, in addition to solving the model analytically, we were able to quantify the effect of the attractive force. To this end, we have shown that in the strong

force regime the convergence towards the closest packing against the strength of the attractive force follows a power-law relation $q(\infty)-q(a)\sim t^{-1/2}$. Perhaps, the emergence of such a power-law behavior implies a universal nature of the phenomena. It also includes the exponent, in the sense that it is independent of the detailed nature of the attractive force. However, at this point it is just a conjecture and we intend to investigate it in our future work.

VII. CONCLUSION

In this paper, we have presented an extension of the simple BD model by incorporating an attractive force between the elements of the adsorbed phase and the incoming particle. The most significant consequence of the presence of the attractive force is the increase in packing fraction or the jamming coverage of the resulting monolayer as we increase the strength of the attractive force a. This is manifested through the increase in the mean cluster size k, which increases as $\sim a^{1/2}$ in the strong force field regions. We have shown that the system exhibits a power-law approach of the jamming coverage towards the closest packing both in a and k but with different exponents $\sim a^{-1/2}$ and $\sim k^{-1}$, respectively, except for the weak field. It is important to note that although the jamming coverage increases with the degree of correlation *vis-a-vis* the mean cluster size, we can never create one single connected cluster spanning the whole substrate and giving the coverage $q=1$, which is indeed a typical character to all 1D problems. Nonetheless, it indicates the potential structural phase transition in higher dimensions that has been indeed observed in Ref. [13]. Finally, we found that for a given set of rules various aspects of the problem except the temporal behavior are independent of how many particles arrive on the substrate at each step. It reveals the versatility of the adsorption phenomena.

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