Exact solution of a generalized ballistic-deposition model

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We study a monolayer deposition model which generalizes both random sequential adsorption and ballistic deposition: hard disks are dropped vertically and randomly onto a line; they can adsorb on the line either by direct deposition or after rolling over preadsorbed disks. An exact solution is given for the gap distribution function and for the density of adsorbed disks. Properties of the system at or close to the jamming limit are discussed. This generalized ballistic-deposition process leads to formation of connected clusters of all sizes: the time evolution of the cluster distribution is obtained analytically.

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I. INTRODUCTION

The adsorption of large molecules, such as latexes and colloids, on solid surfaces is often an essentially irreversible process [1–5], i.e., desorption and surface diffusion are negligible on the time scale of observation. Therefore, a complete description of adsorption mechanisms must take into account the transport of the particles from bulk to the surface, the interaction between adsorbed particles and bulk particles, and, finally, the subsequent adsorption. A large variety of forces is generally involved in each step of these processes: dispersion, electrostatic, hydrodynamic, and even external fields such as gravity.

An early, but nontrivial, approach is the widely studied random-sequential-adsorption (RSA) process [6], which is defined by a sequence of random placements of hard, impenetrable particles onto a surface. The geometrical blocking effects and the irreversible nature of the process result in the following properties: the structure of the adsorbed configurations are distinct from those of the corresponding equilibrium configurations, except for low densities; in the final state of this process, near the "jamming limit," the kinetics follow an algebraic power law [7,8].

Recently, several models have been proposed that incorporate some features of the transport mechanisms [9–14]. When the adsorbing particles are denser than the solution, gravity forces lead to a significant drift toward the surface, and, in the limit of very strong field, it is reasonable to consider the motion as deterministic, i.e., the trajectories simply become straight lines. If the particles do not contact the surface first, they follow the path of steepest descent on the previously deposited particles. This process corresponds to the irreversible adsorption in the limit of a strong external field and is called "ballistic deposition" (BD).

Most of the previous studies of the BD process were concerned with the formation of multilayer deposits and their scaling properties [15]. Recently, Talbot and Ricci [16] proposed a soluble model of a BD process on an infinite line in which multilayer deposits are prevented. The main features of this model are (i) the jamming-limit density, $\rho_{\rm BD}(\infty)=0.808\cdots$, is larger than in RSA, $\rho_{\rm RSA}(\infty)=0.747\cdots$. These results were obtained by Solomon [17] and Rényi [18], respectively; (ii) the long-time behavior is essentially exponential; and (iii) "restructuring," that is, the possibility for a trial disk to move after an initial contact with a preadsorbed disk without being rejected, contributes to the formation of connected clusters. Later, Jullien and Meakin [19] performed a simulation study of the (2+1)-dimensional version of this model, i.e., the deposition of spheres on a plane and observed the same features: (i) a saturation coverage larger than RSA ($\Theta_{\rm BD}(\infty)=0.611$ and $\Theta_{\rm RSA}(\infty)=0.547$), (ii) an exponential approach to the jamming limit, and (iii) cluster formation. Using methods borrowed from liquid-state theories, Thompson and Glandt [20] obtained the density expansion up to third order of the pair-density function, $\rho^{(2)}(r,\rho)$, and the adsorption kinetics to the same order.

In this paper, we introduce a generalized version of this model, in which a tuning parameter a measures the efficiency of restructuring: for $a=0$, the RSA model is recovered, whereas $a=1$ corresponds to the simple BD model. In Sec. II, the analytical treatment of the model in 1+1 dimensions is developed, and we obtain the time-dependent kinetics and gap-distribution functions. In Sec. III, we focus on cluster formation, which is described with analytical expressions for the cluster densities.

II. THE DEPOSITION MODEL

We consider the deposition of hard disks of diameter σ onto an infinite adsorbing line. The disks are dropped uniformly at a constant rate k per unit length as in a RSA process. If, at time τ, the new disk does not encounter any preadsorbed disk, it adsorbs with a probability p. Otherwise, the trial disk rolls over the perimeter of a preadsorbed disk following the path of steepest descent. Suppose now that there is a gap of length l on the relevant side of the preadsorbed disk: if l is more than σ,

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the new disk can reach the line and is accepted with a probability p'; if l is less than σ, the disk cannot reach the line and is rejected (no multilayer formation is allowed in this model). Using dimensionless variables $t=\sigma k p \tau$, $h=l/\sigma$, $a=p'/p$, one can see that for $a=0$, the model corresponds to the car-parking problem, whereas, for $a=1$, we recover the ballistic-deposition model of Talbot and Ricci [16]. In the limit $a \to +\infty$, only deposition via rolling is permitted, which results in a close-packed configuration. The tuning parameter a can be interpreted as a measure of the efficiency of the restructuring due to the rolling mechanism. The interest of this generalized version of ballistic-deposition model is that it permits a nonnegligible fraction of those particles rolling over previously adsorbed particles to be rejected, which seems to be a reasonable physical assumption for monolayer formation.

The rolling mechanism leads to the formation of connected clusters of different sizes. Thus, in the generalized ballistic-deposition model, the gap distribution $G^T(h,t)$, which is the total density of gaps of length h at time t, is given by $G^T(h,t)=G(h,t)+G^s(t)\delta(h)$, where $G(h,t)$ denotes the regular part of the gap-distribution function and $G^s(t)\delta(h)$ is the singular part at contact corresponding to the finite contribution of all clusters. The time evolution of $G^T(h,t)$ is governed by a rate equation that takes into account destruction and creation of intervals of length h. Figure 1 illustrates the two mechanisms by which these intervals may be eliminated: by direct deposition, if the center of the new particle arrives in the inner interval of length $h-1$, and by rolling mechanism if the particle touches the right or left neighbor. Similarly, intervals of length h are created from intervals of length $h+1$ by rolling mechanism and from intervals of length h', with $h'>h+1$, by direct deposition. One obtains for the regular and singular components of the gap-distribution function

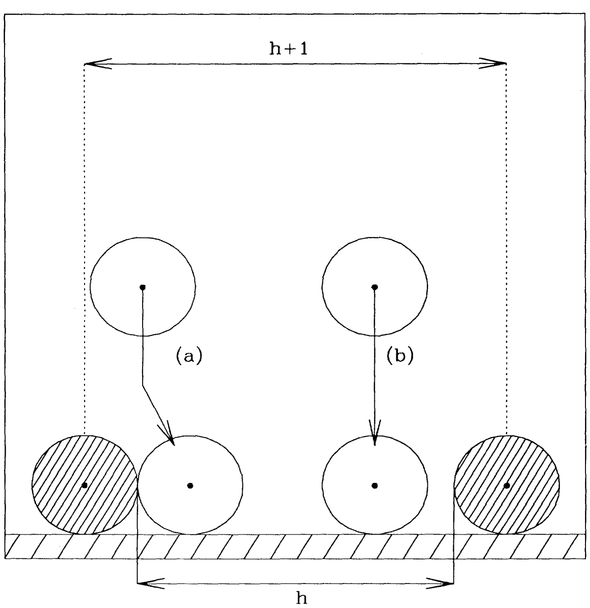

FIG. 1. Illustration of the generalized ballistic-deposition process. The insertion of a falling disk into a gap of length h happens either if the disk rolls over one of the two adjacent disks (a) or if the disk directly reaches the surface (b). The disk diameter is taken as the unit of length.

$$\frac{\partial G(h,t)}{\partial t}=-(h-1+2a)G(h,t)+2aG(h+1,t)+2\int_{h+1}^{\infty} dh' G(h',t), \quad h\geq 1 , \tag{1}$$

$$\frac{\partial G(h,t)}{\partial t}=2aG(h+1,t)+2\int_{h+1}^{\infty} dh' G(h',t), \quad h<1 , \tag{2}$$

$$\frac{dG^s(t)}{dt}=2a\int_1^{\infty} dh\, G(h,t) . \tag{3}$$

The total-number density of gaps, $n_G^T(t)$, is given by

$$n_G^T(t)=\int_0^{\infty} dh\, G^T(h,t) , \tag{4}$$

and by using the decomposition of $G^T(h,t)$ in a regular and singular part, it can be written as the sum of contributions from gaps of finite and zero length:

$$n_G^T(t)=n_G(t)+n_G^0(t)=\int_0^{\infty} dh\, G(h,t)+G^s(t) . \tag{5}$$

As one gap corresponds to one particle, the number density $\rho(t)=n_G^T(t)$. Hence, except in RSA [$a=0$ and $n_G^0(t)=0$], cluster formation induces the inequality $\rho(t)>n_G(t)$. Another route to expressing the density $\rho(t)$ is to calculate the fraction of the line that does not correspond to gaps:

$$\rho(t)=1-\int_0^{\infty} dh\, hG(h,t) . \tag{6}$$

Finally, $\rho(t)$ can be also deduced from the kinetic equation

$$\frac{d\rho}{dt}=\Phi(t)=\int_1^{\infty} dh\,(h-1+2a)G(h,t) , \tag{7}$$

where $\Phi(t)$ is the probability of inserting a new disk at time t.

Introducing the function $H_a(t)$ such that $G(h,t)=\exp[-(h-1+2a)t]H_a(t)$ for $h\geq 1$ and using Eq. (1), one gets the differential equation

$$\frac{d\ln H_a(t)}{dt}=2e^{-t}\left[a+\frac{1}{t}\right] , \tag{8}$$

the solution of which is

$$H_a(t)=H(t)e^{2a(1-e^{-t})} , \tag{9}$$

where $H(t)=t^2\exp[-2\int_0^t(1-e^{-u}/u)du]=\exp[-2\gamma+2\text{Ei}(-t)]$, where $\gamma=0.57721\ldots$ is the Euler constant and $\text{Ei}(-t)=\int_t^{\infty}dx\exp(-x)/x$ is the exponential integral.

The gap-distribution function is then given by

$$G(h,t)=e^{-(h-1)t}e^{2a(1-t-e^{-t})}H(t) , \tag{10}$$

for $h\geq 1$, whereas, for $h<1$, integrating Eq. (2) and then Eq. (3), one obtains

$$G(h,t)=\int_0^t dt'\frac{2}{t'}(1+at')e^{-ht'}e^{2a(1-t'-e^{-t'})}H(t')\ , \tag{11}$$

$$G^s(t)=2a\int_0^t dt'\frac{H(t')}{t'}e^{2a(1-t'-e^{-t'})}\ . \tag{12}$$

The short-time expansion of the gap distribution $G(h,t)$ is

$$G(h,t)=t^2+O(t^3) \tag{13}$$

and does not depend on a, because the initial deposition is always direct. Properties at the jamming limit ($t\to+\infty$) exhibit some differences compared to the RSA ($a=0$) case: the RSA gap distribution is given, when $h\to 0+$, by $G(h,\infty)\sim -2e^{-2\gamma}\ln(h)$, and it thus displays a logarithmic divergence at contact. For $a>0$, $G(h,\infty)\sim -2e^{-2\gamma}e^{2a}\ln(h+2a)$, and the regular gap-distribution function $G(h,\infty)$ remains finite at contact [there is, of course, a singular gap distribution which is proportional to $a\delta(h)$]. $G(h,\infty)$ is illustrated in Fig. 2 for different values of a.

The number density is obtained from the gap-distribution function $G(h,t)$ by using Eq. (6):

$$\rho(t)=\int_0^t dt'\frac{H(t')}{t'^2}(1+2at')e^{2a(1-t'-e^{-t'})}\ , \tag{14}$$

and from Eq. (5), the gap density $n_G(t)$ is given by

$$n_G(t)=\int_0^t dt'\frac{H(t')}{t'^2}e^{2a(1-t'-e^{-t'})}\ . \tag{15}$$

Figure 3 compares the number density $\rho(t)$ and gap density $n_G(t)$ for different values of a, and Fig. 4 displays their values at the jamming limit. One observes that the particle density increases with a while the number of *finite* length gaps decreases with a. As explained above, the increasing difference between $\rho(t)$ and $n_G(t)$ is due to the formation of clusters, which contributes to $\rho(t)$ and $n_G^0(t)$, but not to $n_G(t)$. From Eq. (14), one obtains the

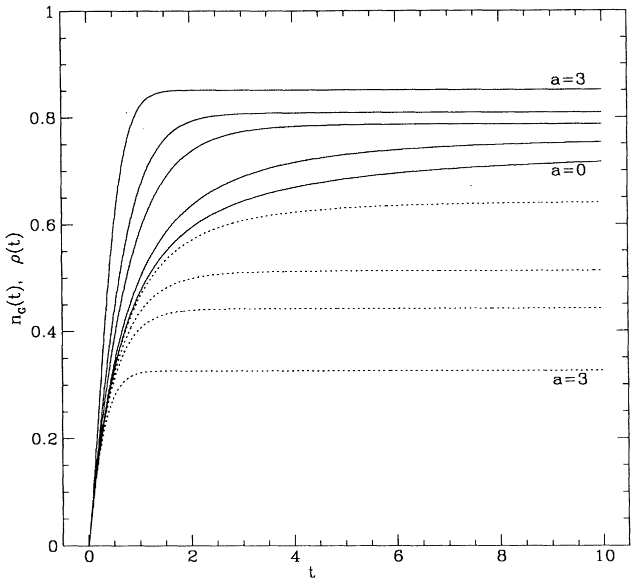

FIG. 3. Time dependence of the number density $\rho(t)$ and of the gap density $n_G(t)$ for various values of a. The middle curve ($a=0$) corresponds to the RSA process: no connected clusters can appear and then $n_G(t)=\rho(t)$. The curves from middle to top represent $\rho(t)$ and the dashed curves from middle to bottom represent $n_G(t)$ for $a=0.1,0.5,1.0,3.0$. $a=1$ corresponds to the ballistic deposition model of Talbot and Ricci.

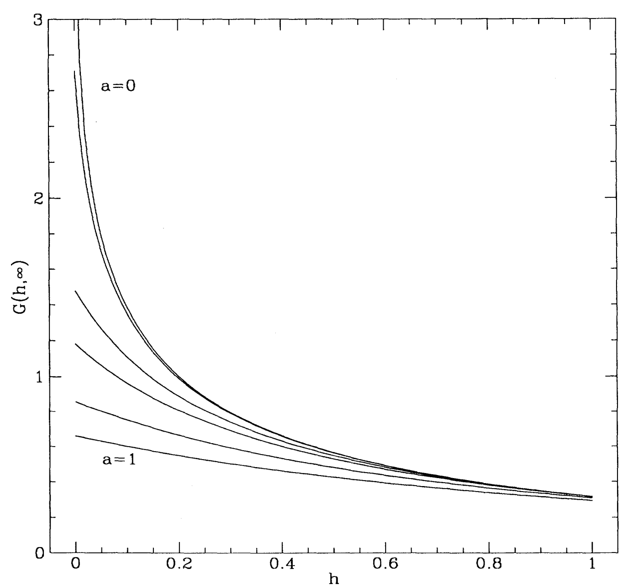

FIG. 2. Regular gap-distribution function at the jamming limit for various values of a. The curves from top to bottom correspond to the following sequence: $a=0,0.01,0.1,0.2,0.5,1.0$.

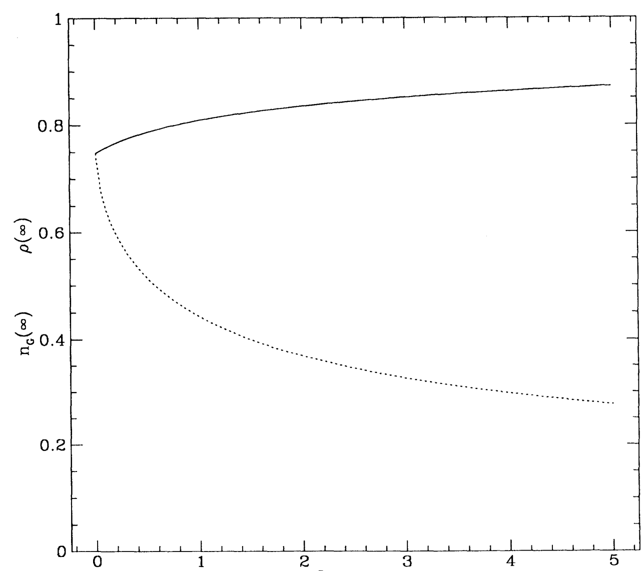

FIG. 4. Number density $\rho(\infty)$ (upper curve) and gap density $n_G(\infty)$ at the jamming limit versus a.

asymptotic behavior of the particle density when approaching the jamming limit, for $1 \ll t \ll 1/a$,

$$\rho(\infty)-\rho(t) \sim e^{2(a-\gamma)}\frac{1}{t} , \tag{16}$$

and for $t \gg 1/a$,

$$\rho(\infty)-\rho(t) \sim e^{2(a-\gamma)}\frac{e^{-2at}}{t} . \tag{17}$$

Thus, for any $a>0$, the approach to saturation is exponential, whereas the usual power law is recovered for RSA ($a=0$),

$$\rho(\infty)-\rho(t) \sim \frac{e^{-2\gamma}}{t} . \tag{18}$$

At short times, the density $\rho(t)$ behaves as

$$\rho(t)=t-(1-a)t^2+\tfrac{5}{6}(2a-1)t^3+\cdots \tag{19}$$

and the probability of inserting a new disk can be written as a density expansion:

$$\Phi=1-(1-a)\rho+(\tfrac{1}{2}-a-2a^2)\rho^2 + \left[\frac{2}{9}-\frac{2a}{3}+4a^3-\frac{2a^2}{3}\right]\rho^3+\cdots . \tag{20}$$

The preceding expansion can be interpreted as follows: starting with an empty line, the probability is equal to 1 due to direct deposition without exclusion effects. The linear term represents the possibility of rejecting particles that fall over isolated particles, i.e., over particles whose exclusion areas do not intersect themselves (low-density limit): for $a=1$, this term vanishes and this is also true in two dimensions [20]; the quadratic term takes into account corrections to the first-order exclusion effect for direct deposition as well as rejection of those particles that are prevented from reaching the line by rolling mechanism due to the presence of a nearby preadsorbed disk. This term is different from zero for $a=1$, contrary to the two-dimensional (2D) case for which the first nonzero term is the cubic one [20].

III. CLUSTER FORMATION

In the generalized ballistic model (for $a>0$), the rolling mechanism leads to formation of connected clusters of different sizes, contrary to the RSA case ($a=0$). Some preliminary information on the cluster distribution can be derived from the previous analysis. Let $\rho_s(t)$ denote the number density of connected clusters formed by exactly s disks. Then, the particle density is expressed as

$$\rho(t)=\sum_{s=1}^{+\infty} s\rho_s(t) , \tag{21}$$

and the gap density as

$$n_G(t)=\sum_{s=1}^{+\infty} \rho_s(t) . \tag{22}$$

The ratio $M(t)=\rho(t)/n_G(t)$ can then simply be interpreted as an average number of disks per cluster. Note that $n_G(t)=\rho(t)$ only for $a=0$, i.e., for the RSA process. For various values of a, the time dependence of $M(t)$ is shown in Fig. 5. As expected, the average number of disks per cluster increases with increasing efficiency of the rolling mechanism. In one dimension, the percolation threshold, corresponding to the formation of an infinite cluster, is only reached when a goes to infinity.

More detailed information on the densities of clusters $\rho_s(t)$ can also be obtained. However, the mere knowledge of the gap-distribution functions, $G(h,t)$, $G^s(t)$, does not permit the calculation of the cluster densities $\rho_s(t)$. To overcome this problem, we introduce higher-order *gap-cluster-gap* distribution functions, $G(h,h',s,t)$, defined as the number density of pairs of neighboring gaps of length h and h' that are separated by a cluster of exactly s disks. As for the single-gap-distribution functions in the preceding section, we can write rate equations in a closed form by analyzing destruction and creation of these "gap-cluster-gap" systems (see Fig. 6); If $h>1$ and $h'>1$, any gap-cluster-gap system labeled by (h,h',s) can be destroyed by direct deposition on the left or right interval or by rolling mechanism on the two sides of each interval; conversely, a gap-cluster-gap system (h,h',s) can come from a gap-cluster-gap (h'',h',s) or a gap-cluster-gap system (h,h''',s) by direct deposition if $h''>h+1$ or $h'''>h'+1$, or can result from gap-cluster-gap systems $(h+1,h',s)$, $(h,h'+1,s)$, $(h+1,h',s-1)$, $(h,h'+1,s-1)$ by rolling mechanism. Note that for the case $s=1$, the creation of a gap-cluster-gap system $(h,h',1)$ results only from a single gap of length $h+h'+1$. This gives the following equation for the time evolution of the "gap-cluster-gap distribution functions:"

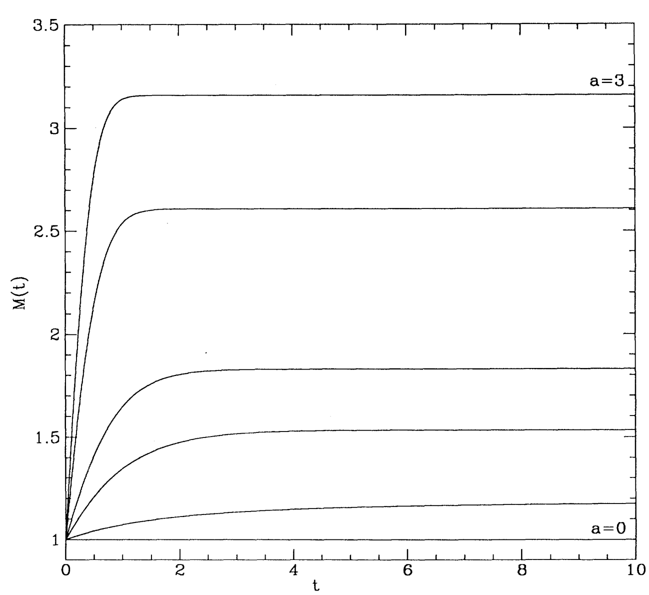

FIG. 5. Time dependence of the average number of disks per cluster for various values of a. The curves from bottom to top correspond to $a=0,0.1,0.5,1.0,3.0$.

$$\frac{\partial G(h,h',s,t)}{\partial t} = -[k(h)+k(h')]G(h,h',s,t)+\int_{h+1}^{\infty} dh''G(h'',h',s,t) + \int_{h'+1}^{\infty} dh''G(h,h'',s,t)+a[G(h+1,h',s,t)+G(h,h'+1,s,t)] + a(1-\delta_{s,1})[G(h+1,h',s-1,t)+G(h,h'+1,s-1,t)]+\delta_{s,1}G(h+h'+1,t) , \quad (23)$$

where $\delta_{s,1}$ is the Kronecker symbol, $G(h+h'+1,t)$ is the single-gap distribution for intervals of length $h+h'+1$, and $k(h)=h-1+2a$ if $h \geq 1$ and $k(h)=0$ if $h<1$.

The number density of clusters of size s can be obtained by integration over all gap-cluster-gap densities

$$\rho_s(t)=\int_0^{\infty}\int_0^{\infty} dh\, dh' G(h,h',s,t) . \quad (24)$$

The strategy for solving these coupled equations consists of considering first the situation $h>1$ and $h'>1$ with the substitution $G(h,h',s,t)=e^{-(h+h'-2+4a)t}H_a(s,t)$; the functions $H_a(s,t)$ must then satisfy

$$\frac{\partial H_a(s,t)}{\partial t} = 2e^{-t}\left[a+\frac{1}{t}\right]H_a(s,t) + 2ae^{-t}H_a(s-1,t), \quad s \geq 0 , \quad (25)$$

with the initial condition

$$H_a(0,t)=\frac{1}{2a}H_a(t)e^{(2a-1)t} , \quad (26)$$

where $H_a(t)$ is given by Eq. (9). Second, for $h<1$ and $h' \geq 1$ (respectively for $h \geq 1$ and $h'<1$), the form of the gap-cluster-gap distribution function is given by $G(h,h',s,t)=e^{-(h'-1+2a)t}H_a(h,s,t)$, [respectively, $G(h,h',s,t)=e^{-(h-1+2a)t}H_a(h',s,t)$]. Introducing $h_a(s,t) = \int_0^1 dh' H_a(h',s,t)$, one can express, after some algebra, the s-cluster density $\rho_s(t)$ as

$$\rho_s(t)=\frac{e^{-4at}}{t^2}H_a(s,t)+2\frac{e^{-2at}}{t}h_a(s,t) + 2\int_0^t dt' e^{-2at'}\left[\frac{1-e^{-t'}}{t'}\right]\left[\left[\frac{1}{t'}+a\right]h_a(s,t') + ah_a(s-1,t')\right], \quad s \geq 1 , \quad (27)$$

where the function $h_a(s,t)$ can be determined from the equation

$$\frac{\partial h_a(s,t)}{\partial t} = e^{-t}\left[a+\frac{1}{t}\right]h_a(s,t) + e^{-2at}\left[\frac{1-e^{-t}}{t}\right]\left[\frac{1}{t}+a\right]H_a(s,t) + a\left[e^{-2at}\left[\frac{1-e^{-t}}{t}\right]H_a(s-1,t) + e^{-t}h_a(s-1,t)\right], \quad s \geq 1 , \quad (28)$$

with the initial condition

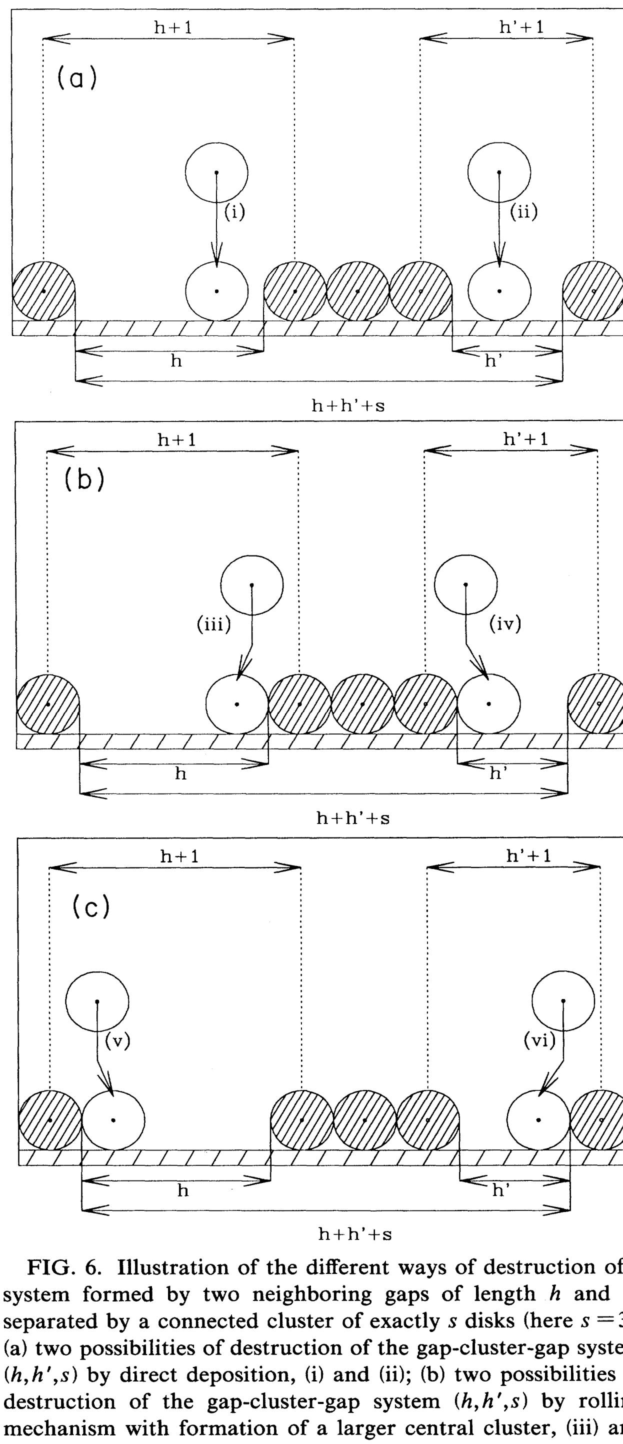

FIG. 6. Illustration of the different ways of destruction of a system formed by two neighboring gaps of length h and h' separated by a connected cluster of exactly s disks (here $s=3$): (a) two possibilities of destruction of the gap-cluster-gap system (h,h',s) by direct deposition, (i) and (ii); (b) two possibilities of destruction of the gap-cluster-gap system (h,h',s) by rolling mechanism with formation of a larger central cluster, (iii) and (iv); (c) two possibilities of destruction of the gap-cluster-gap system (h,h',s) by rolling mechanism with an unchanged central s cluster, (v) and (vi).

$$h_a(0,t)=\frac{H_a(t)}{2a}\left[\frac{1-e^{-t}}{t}\right]. \tag{29}$$

Integrating Eq. (27) by parts, the s-cluster density can be written as

$$\rho_s(t)=\tilde{\rho}_{s-1}(t)-\tilde{\rho}_s(t) \tag{30}$$

with

$$\tilde{\rho}_s(t)=2a\int_0^t dt'\frac{e^{-2at'}}{t'}\left[h_a(s,t')+\frac{e^{-2at'}}{t'}H_a(s,t')\right],\quad s\geq 0\,. \tag{31}$$

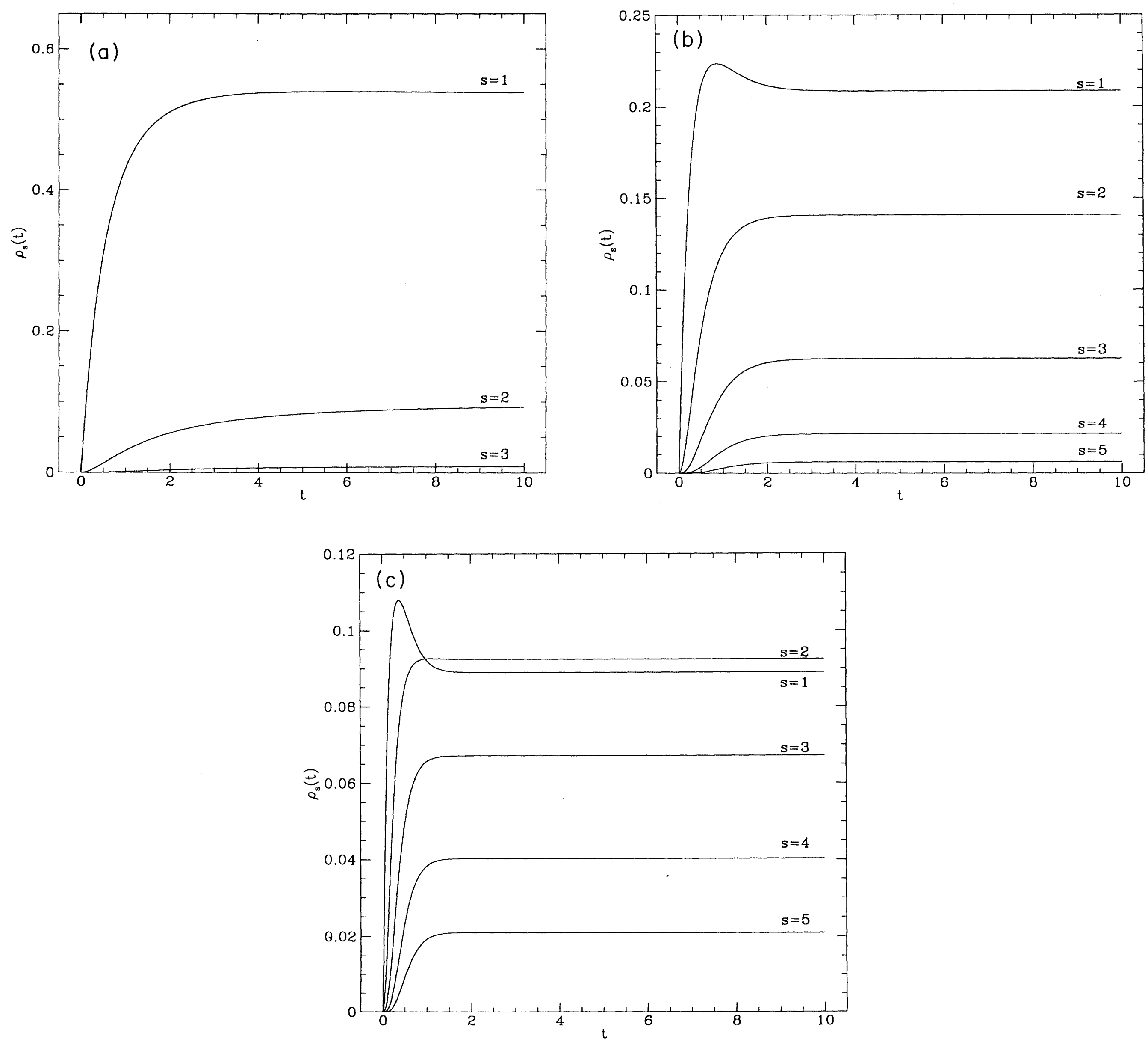

FIG. 7. Time dependence of the cluster densities $\rho_s(t)$ for different values of $a>0$ and s. (a) $a=0.1$, $s=1,2,3$, (b) $a=1$, $s=1,2,3,4,5$, (c) $a=3$, $s=1,2,3,4,5$. Note that the density of monomers $\rho_1(t)$ has a maximum at a finite value for $a=1$ and 3 but not for $a=0.1$. For $a=3$, the efficiency of the rolling mechanism leads to a density of dimers larger than the density of monomers at the jamming limit.

In the above equation, $\tilde{\rho}_s(t)$ denotes the density of cluster of size strictly larger than s. One checks directly from Eqs. (28)–(31) that the two sum rules given by Eqs. (21) and (22) are satisfied: cf. Appendix A. Also as expected, the s-cluster densities $\rho_s(t)$ vanish for $s>1$ when $a=0$ (RSA).

After some algebra (the details are given in Appendix B), an explicit solution for $\tilde{\rho}_s(t)$ can be obtained:

$$\tilde{\rho}_s(t)=\frac{2a^s}{(s-1)!}\int_0^t dt_1 e^{-2at_1}\frac{\sqrt{H_a(t_1)}}{t_1}\int_0^{t_1} dt_2\frac{\sqrt{H_a(t_2)}}{t_2}\Bigg\{e^{-t_2}(e^{-t_1}-e^{-t_2})^{s-1}+\sum_{j=0}^{s-1}C_{s-1}^{j}2^j(-1)^{s-1-j}(e^{-t_2}+e^{-t_1})^{s-1-j}\times\left[\frac{j}{2}\left[\frac{e^{-2at_2}-e^{-(j+1)t_2}}{j+1-2a}\right]-a\left[\frac{e^{-2at_2}-e^{-(j+2)t_2}}{j+2-2a}\right]\right]\Bigg\}. \tag{32}$$

Figure 7 illustrates cluster formation versus time for different values of a. At short times, the s-cluster density $\rho_s(t)$ increases as

$$\rho_s(t)\sim a^{s-1}t^s . \tag{33}$$

We may interpret this result by noting that the first appearance of a cluster of size s is only possible after s depositions of disks and by a sequence of $(s-1)$ rolling processes [21]. Equation (32) can be also used to derive the behavior of the s-cluster density at the jamming limit for asymptotically large clusters ($s\rightarrow+\infty$):

$$\rho_s(\infty)\sim\frac{(2a)^{s-1}}{(s)!} . \tag{34}$$

This large-s behavior is very different from that expected at a percolation threshold and an infinite cluster can only be obtained in the limit $a\rightarrow\infty$ corresponding to a close-packed configuration. Interestingly, faster-than-exponential decays similar to Eq. (34) occur in models of irreversible filling of a one-dimensional lattice. This appears to be a general property of 1D irreversible deposition processes which do not permit cluster-cluster coalescence [22].

IV. CONCLUSION

In this article, we have presented a solvable model of monolayer formation, which generalizes both random sequential adsorption and ballistic deposition: gap-distribution functions and the density of adsorbed disks are obtained. This generalized ballistic-deposition process involves formation of connected clusters of all sizes: by means of higher-order distribution functions, the time evolution of the cluster distribution is also derived. In one dimension, an infinite cluster cannot be formed for a finite value of a and the percolation threshold is not reached.

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APPENDIX A: SUM RULES

The expression for the gap density is easily recovered thanks to successive cancellation in the summation over s [compare Eq. (22) with Eqs. (30) and (31)]. One finds

$$n_G(t)=2a\int_0^t dt'\frac{e^{-2at'}}{t'}\left[h_a(0,t')+\frac{e^{-2at'}}{t'}H_a(0,t')\right] . \tag{A1}$$

Combining Eq. (A1) with the initial conditions given in Eqs. (26) and (29), we indeed obtain Eq. (15).

The number density $\rho(t)$ is the sum of the contributions of clusters of all sizes; using Eqs. (21) and (31) leads to

$$\rho(t)=2a\int_0^t dt\sum_{i=0}^{+\infty}\frac{e^{-2at}}{t}\left[h_a(i,t)+\frac{e^{-2at}}{t}H_a(i,t)\right] . \tag{A2}$$

Let $U(t)$ be defined as

$$U(t)=\frac{1}{H_a(t)}\sum_{i=1}^{+\infty}\left[h_a(i,t)+\frac{2e^{-2at}}{t}H_a(i,t)\right] . \tag{A3}$$

This function obeys the following equation:

$$\frac{dU(t)}{dt}=\frac{e^{-t}}{t}[1-U(t)] . \tag{A4}$$

With the initial condition $U(0)=1$, Eq. (A4) has the unique solution $U(t)=1$. Since the number density $\rho(t)$ can be reexpressed as

$$\rho(t)=\int_0^t dt'\frac{H(t')}{t'}\left[\frac{1}{t'}+2aU(t')\right]e^{2a(1-t'-e^{-t'})} , \tag{A5}$$

one then recovers Eq. (14).

APPENDIX B: CLUSTER DENSITIES

To obtain a solution for the s-cluster density $\rho_s(t)$, one introduces more adapted functions

$$A_a(s,t)=\frac{2}{\sqrt{H_a(t)}}\left[h_a(s,t)+\frac{e^{-2at}}{t}H_a(s,t)\right] \tag{B1}$$

and

$$B_a(s,t)=\frac{2H_a(s,t)}{H_a(t)} . \tag{B2}$$

By using Eqs. (25) and (28), one derives

$$\frac{dA_a(s,t)}{dt}=ae^{-t}A_a(s-1,t)+a\frac{\sqrt{H_a(t)}e^{-2at}}{t}[B_a(s-1,t)-B_a(s,t)] , \tag{B3}$$

$$\frac{dB_a(s,t)}{dt}=2ae^{-t}B_a(s-1,t) , \tag{B4}$$

with the initial conditions $A_a(0,t)=\sqrt{H_a(t)}/(at)$ and $B_a(0,t)=e^{(2a-1)t}/a$. Thus, the density of clusters of size s is given by

$$\rho_s(t)=\int_0^t dt'ae^{-2at'}\frac{\sqrt{H_a(t')}}{t'}[A_a(s-1,t')-A_a(s,t')] , \tag{B5}$$

where $A_a(s,t)$ is

$$A_a(s,t)=a^s\int_0^t dt_1e^{-t_1}\int_0^{t_1}dt_2e^{-t_2}\cdots\int_0^{t_{s-1}}dt_se^{-t_s}\frac{\sqrt{H_a(t_s)}}{at_s}+\sum_{i=1}^{s}a^i\int_0^t dt_1e^{-t_1}\int_0^{t_1}dt_2e^{-t_2}\cdots\int_0^{t_{i-1}}dt_ie^{-2at_i}\frac{\sqrt{H_a(t_i)}}{t_i}[B_a(s-i,t_i)-B_a(s+1-i,t_i)] , \tag{B6}$$

with

$$B_a(s,t)=(2a)^s\int_0^t dt_1e^{-t_1}\int_0^{t_1}dt_2e^{-t_2}\cdots\int_0^{t_{s-1}}dt_se^{-t_s}\frac{e^{(2a-1)t_s}}{a} . \tag{B7}$$

Using the change of variable $y=a(1-e^{-t})$ in Eq. (B7), one finds

$$B_a(s,y)=2^s\int_0^y dy'\frac{(y-y')^{s-1}}{(s-1)!}B_a(0,y') , \tag{B8}$$

and performing the same procedure in Eq. (B6), one obtains after inserting Eq. (B8)

$$A_a(s,y)=\frac{2}{(s-1)!}\left[\int_0^y dy'(y-y')^{s-1}A_a(0,y')+\int_0^y dy'\frac{A_a(0,y')}{B_a(0,y')}\int_0^{y'}dy''B_a(0,y'')[(s-1)(y-2y''+y')^{s-2}-(y-2y''+y')^{s-1}]\right] . \tag{B9}$$

Changing back the variable y to t and combining Eqs. (B5) and (B9) leads finally to Eq. (32).

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