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Random sequential adsorption of straight rigid rods on a simple cubic lattice

G.D. García [a], F.O. Sanchez-Varretti [a], P.M. Centres [b], A.J. Ramirez-Pastor [b,*]

[a] *Universidad Tecnológica Nacional, Regional San Rafael, Gral. Urquiza 314, M5602GCH, San Rafael, Mendoza, Argentina*

[b] *Departamento de Física, Instituto de Física Aplicada, Universidad Nacional de San Luis-CONICET, Ejército de Los Andes 950, D5700HHW, San Luis, Argentina*

HIGHLIGHTS

- The RSA problem of straight rigid rods on a simple cubic lattice is studied.
- A new procedure to calculate jamming coverage θ_j is presented.
- The behavior of the jamming coverage as a function of the rod size is obtained.
- The value of θ_j obtained for dimers corrects previous calculations in the literature.

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ABSTRACT

Random sequential adsorption of straight rigid rods of length k (k-mers) on a simple cubic lattice has been studied by numerical simulations and finite-size scaling analysis. The k-mers were irreversibly and isotropically deposited into the lattice. The calculations were performed by using a new theoretical scheme, whose accuracy was verified by comparison with rigorous analytical data. The results, obtained for k ranging from 2 to 64, revealed that (i) the jamming coverage for dimers ($k = 2$) is $\theta_j = 0.918388(16)$. Our result corrects the previously reported value of $\theta_j = 0.799(2)$ (Tarasevich and Cherkasova, 2007); (ii) θ_j exhibits a decreasing function when it is plotted in terms of the k-mer size, being $\theta_j(\infty) = 0.4045(19)$ the value of the limit coverage for large k's; and (iii) the ratio between percolation threshold and jamming coverage shows a non-universal behavior, monotonically decreasing to zero with increasing k.

1. Introduction

The study of systems of hard non-spherical particles is one of the central problems in statistical mechanics and has been attracting a great deal of interest since long ago. In a seminal contribution to this subject, Onsager [1] predicted that rod-like particles interacting with only excluded-volume interaction can lead to long-range orientational (nematic) order. This nematic phase, characterized by a big domain of parallel molecules, is separated from an isotropic state by a phase transition occurring at a finite critical density. Numerous model systems of anisotropic objects, ranging from inorganic to organic and also biological particles, were analyzed by using the Onsager theory. An extensive overview of this field can be found in the excellent reviews of Refs. [2,3].

* Corresponding author.
E-mail address: antorami@unsl.edu.ar (A.J. Ramirez-Pastor).

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The long-range orientational order disappears in the case of irreversible adsorption (no desorption), where the distribution of adsorbed objects is different from that obtained at equilibrium. Thus, for the equilibrium problem, a nematic phase is observed at intermediate density, and a second phase transition (from a nematic order to a non-nematic state) occurs at high density [4–8]. On the other hand, the final state generated by irreversible adsorption is a disordered state (known as jamming state), in which no more objects can be deposited due to the absence of free space of appropriate size and shape. The jamming state has infinite memory of the process and the orientational order is purely local [9–11].

The phenomenon of jamming plays an important role in numerous systems where the deposition of objects is irreversible over time scales of physical interest [12]. The random sequential adsorption (RSA), introduced by Feder [13], has served as a paradigm for modeling irreversible deposition processes. The main features of the RSA model are: (1) the objects are put on randomly chosen sites, (2) the adsorption is irreversible and (3) at any time only one object is being adsorbed, so that the process takes place sequentially. In this theoretical frame, anisotropic particles of different shapes and sizes have been studied: linear k-mers (particles occupying k adjacent lattice sites) [14–18], flexible polymers [19,20], T-shaped objects and crosses [21], squares [22–25], disks [26], regular and star polygons [27], etc. In all cases, the limiting or jamming coverage, θ_j, strongly depends on the shape and size of the depositing particles.

In the case of lattice models of straight rigid rods (or linear k-mers), which is the topic of this paper, the RSA problem has been exactly solved for one-dimensional (1D) substrates. In this limit, $\theta(t)$ can be written as [14],

$$\theta(t) = k \int_0^t \exp\left[-u - 2\sum_{j=1}^{k-1}\left(\frac{1-e^{-ju}}{j}\right)\right] du. \tag{1}$$

From Eq. (1), the dependence on k of the jamming coverage can be obtained. Note that $\theta(t)$ represents the fraction of lattice sites covered at time t by the deposited objects and, consequently, $\theta(t=\infty)=\theta_j$. For $k \to \infty$, the jamming threshold tends to Rényi's Parking Constant $\theta_j \to c_R \approx 0.7475979202$ [28].

In two and three dimensions, the inherent complexity of the system still represents a major difficulty to the development of accurate analytical solutions, and computer simulations appear as a very important tool for studying this subject. In this direction, Bonnier et al. investigated the deposition of linear k-mers (with k ranging between 2 and 512 on a two-dimensional (2D) square lattice [15]). The authors found that the jamming concentration monotonically decreases and tends to 0.660(2) as the length of the rods increases.

Vandewalle et al. studied the percolation and jamming phenomena for straight rigid rods of size k on 2D square lattices [16] and found that, for values of k between 2 and 10, the ratio θ_p/θ_j (being θ_p the percolation threshold) remains constant $\theta_p/\theta_j \approx 0.62$, regardless of the length of the particle. Based on this finding, the authors suggested that both critical phenomena (percolation and jamming) are intimately related.

Kondrat and Pękalski [17] extended the study of Ref. [16] to larger lattices (lattice size $L = 30$, 100, 300, 1000 and 2500) and longer objects ($1 \le k \le 2000$). The results obtained revealed that: (1) as reported in Ref. [15], the jamming coverage decreases monotonically approaching the asymptotic value of $\theta_j = 0.66(1)$ for large values of k; (2) the percolation threshold is a nonmonotonic function of the size k: it decreases for small rod sizes, goes through a minimum around $k = 13$, and finally increases for large segments; and (3) the ratio of the two thresholds θ_p/θ_j has a more complex behavior: after initial growth, it stabilizes between $k = 3$ and $k = 7$, and then it grows again.

Recently, Lebovka et al. studied an anisotropic RSA of straight rigid rods on 2D square lattices [18]. In this model, the vertical and horizontal orientations occur with different probabilities, and the degree of anisotropy of the system can be characterized by an order parameter measuring the difference (normalized) between the number of line segments oriented in the vertical direction and the number of line segments oriented in the horizontal direction. The authors investigated the effect of k-mer alignment on the jamming properties and found important differences with respect to the isotropic case. In the limit of isotropic systems (order parameter equal to zero), the results obtained by Lebovka et al. are in excellent agreement with previous simulation data in Ref. [17].

The RSA problem becomes more difficult to solve when the objects are deposited on three-dimensional (3D) lattices, and only very moderate progress has been reported so far. In the line of present work, Tarasevich and Cherkasova examined the percolation and jamming properties of dimers (straight rigid rods with $k = 2$) on 3D simple cubic lattices and found that $\theta_p = 0.2555(1)$, $\theta_j = 0.799(2)$ and $\theta_p/\theta_j \approx 0.32$ [29]. This value of θ_p/θ_j differs from the corresponding value obtained for square lattices and small values of k ($2 \le k \le 10$), $\theta_p/\theta_j \approx 0.62$ [16].

In the present paper, the study of Tarasevich and Cherkasova is extended to larger k-mers ($2 \le k \le 64$). Using computational simulations and a new method introduced here, the jamming properties of straight rigid rods on 3D simple cubic lattices are studied. The method is based on the calculation of the probability $W_L(\theta)$ that a lattice composed of $L \times L \times L$ elements reaches a coverage θ. As it will be discussed in detail in the next section, the intersection point of the curves $W_L(\theta)$ for different values of L gives an accurate estimation of the jamming threshold in the infinite system.

It is quite obvious that the model considered here is highly idealized and is not meant to reproduce a particular experimentally studied system. However the objectives of this work are (1) to introduce a new procedure to calculate jamming properties, which can be widely applied in RSA problems; (2) to evaluate the accuracy and applicability of the new technique; (3) to determine, for the first time, the dependence of the jamming coverage on the size of the deposited k-mers for 3D cubic lattices; (4) to improve previous calculations of θ_j for dimers ($k = 2$) [29]; and (5) to stimulate the development of more sophisticated models which can be able to reproduce concrete experimental systems. With respect to the last point, the 3D adsorption of k-mers layer by layer could be an interesting topic for further research.

The outline of the paper is as follows. In Section 2 we describe the deposition model, along with the simulation scheme and the procedure used to calculate the jamming coverage. In Section 3 we present the results and discussion of the simulations. Finally, the general conclusions are given in Section 4.

2. Model, basic definitions and simulation scheme

The following scheme is usually called standard model of deposition or RSA. Let us consider the substrate is represented by a three-dimensional simple cubic lattice of $M = L \times L \times L$ sites and periodic boundary conditions. In the filling process, straight rigid rods of length k (with $k \geq 2$) are deposited randomly, sequentially, and irreversibly on an initially empty lattice. The procedure of deposition is as follows: (i) one of the three possible lattice directions (x, y, z) and a starting site are randomly chosen (random and isotropic deposition); (ii) if, beginning at the chosen site, there are k consecutive empty sites, then a k-mer is deposited on those sites. Otherwise, the attempt is rejected. When N rods are deposited, the concentration is $\theta = kN/M$.

Due to the blocking of the lattice by the already randomly deposited dimers, the limiting or jamming coverage, $\theta_j \equiv \theta(t = \infty)$ is less than that corresponding to the close packing ($\theta_j < 1$). Note that $\theta(t)$ represents the fraction of lattice sites covered at time t by the deposited objects. Consequently, θ ranges from 0 to θ_j for objects occupying more than one site, and the jamming coverage depends on the size of the deposited object [9,14–18].

It is well known that it is a quite difficult matter to analytically determine the value of the jamming coverage for a given lattice. For some special types of lattices, geometrical considerations enable to derive their jamming thresholds exactly [14]. For systems which do not present such a topological advantage, jamming properties have to be estimated numerically by means of computer simulations.

In order to calculate the jamming thresholds, we introduce the probability $W_L(\theta)$ that a lattice composed of $L \times L \times L$ elements reaches a coverage θ. In the simulations, the procedure to determine $W_L(\theta)$ consists of the following steps: (a) the construction of the $M = L \times L \times L$ lattice (initially empty) and (b) the deposition of particles on the lattice up to the jamming limit θ_j. In the latter step, the quantity $m_i(\theta)$ is calculated as

$$m_i(\theta) = \begin{cases} 1 & \text{for } \theta \leq \theta_j \\ 0 & \text{for } \theta > \theta_j. \end{cases} \tag{2}$$

n runs of such two steps (a)–(b) are carried out for obtaining the number $m(\theta)$ of them for which a lattice reaches a coverage θ,

$$m(\theta) = \sum_{i=1}^{n} m_i(\theta). \tag{3}$$

Then, $W_L(\theta) = m(\theta)/n$ is defined and the procedure is repeated for different values of lattice sizes. For infinite systems ($L \to \infty$), $W_L(\theta)$ is a step function, being 1 for $\theta \leq \theta_j$ and 0 for $\theta > \theta_j$. For finite values of L, $W_L(\theta)$ varies continuously between 1 and 0, with a sharp fall around θ_j. In conclusion, $W_L(\theta)$ appears as a good parameter evidencing the jamming threshold and its definition is computationally convenient.

Using a system of fully aligned rods deposited on 3D cubic lattices as an example of application,[1] we show in the following how to determine the corresponding jamming coverage from the probabilities $W_L(\theta)$. This model of fully aligned rods can be exactly solved [14], which allows us not only to illustrate the use of the method introduced here, but also to test its validity and accuracy by comparing our simulation results with those of Ref. [14]. Five k-mer sizes were chosen for the comparison $k(=2, 3, 4, 5, 6)$. Then, a set of $n = 10^5$ runs were carried out for each pair k and $L/k(=18, 24, 30, 36, 42)$.

In Fig. 1, the probabilities $W_L(\theta)$ are presented for a system of aligned dimers ($k = 2$) and several values of L as indicated. As it can be observed from the figure, curves for different lattice sizes cross each other in a unique point, which is located at a very well defined value in the θ-axes determining the jamming coverage for each k. The procedure in Fig. 1 was repeated for $k(=3, 4, 5, 6)$ (data do not shown here for reasons of space) and the results are collected in the second column of Table 1.

The high accuracy of the simulation data can be asserted by comparison with exact results for θ_j presented in Ref. [14]. In fact, for a fully aligned system, as studied here, the k-mers are deposited only along an axis of the lattice, and the jamming problem reduces to the 1D case. In this limit, θ_j can be calculated from Eq. (1) [14]. The results are shown in the first column of Table 1. A remarkable agreement is obtained between theoretical and simulation data, validating the applicability the method introduced here to calculate jamming thresholds.

In the next section, we will analyze the dependence of the jamming coverage on the size of the deposited objects for a system of straight rigid rods on a simple cubic lattice.

[1] In this case, the k-mers are deposited along one of the directions of the lattice, forming a nematic phase.

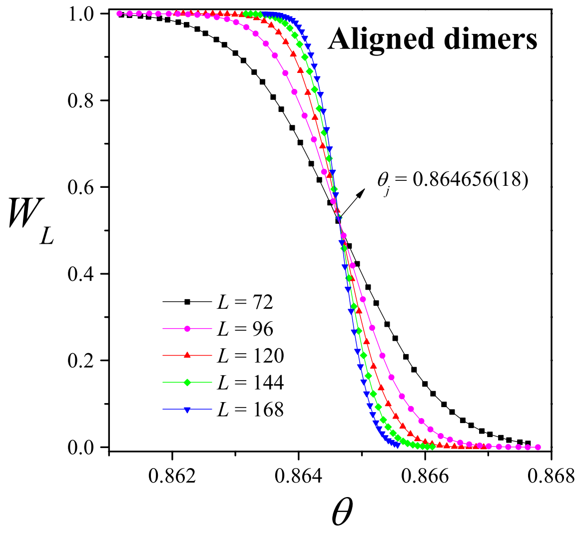

Fig. 1. Curves of W_L vs. θ for fully aligned dimers on simple cubic lattices of different sizes as indicated. From their intersections one obtained θ_j. In this case, $\theta_j = 0.864656(18)$.

Table 1
Simulation and theoretical values of θ_j (as indicated in the text) for fully aligned rods on a simple cubic lattice and k ranging from 2 to 6.

k	Aligned rods	
	θ_j (Eq. (1))	θ_j (W_L's)
2	0.864665	0.864656(18)
3	0.823653	0.823646(16)
4	0.803893	0.803892(12)
5	0.792276	0.792284(15)
6	0.784630	0.784637(16)

3. Simulation results: random sequential adsorption of straight rigid rods on a simple cubic lattice

The procedure described in Section 2 was applied to calculate the jamming coverage corresponding to a system of straight rigid rods on a simple cubic lattice. k-mers of length $k = 2 \ldots 64$ were considered, and the finite-size study was performed for lattices ranging between $(36 \times 36 \times 36)$ and $(1152 \times 1152 \times 1152)$ sites. In addition, $n = 10^5$ runs were carried out for each pair k and L. These values of the parameters allowed us to obtain very accurate measurements of the jamming thresholds, with errors less than 0.008% in all cases.

In Fig. 2, the probabilities $W_L(\theta)$ are shown for two different values of k ($k = 2$ and $k = 5$ as indicated) and L/k(=18, 24, 30, 36, 42). From a simple inspection of the figure (and from data do not shown here for a sake of clarity) it is observed that: (a) for each k, the curves cross each other in a unique point W_L^*; (b) those points do not modify their numerical value for the different k used (ranged between $k = 2$ to $k = 64$). In the cases of Fig. 2, $W_L^* = 0.509(9)$ for $k = 2$, and $W_L^* = 0.505(9)$ for $k = 5$; (c) those points are located at very well defined values in the θ-axes determining the jamming threshold for each k, being $\theta_j = 0.918388(16)$ for $k = 2$, and $\theta_j = 0.736061(12)$ for $k = 5$; and (d) θ_j decreases for increasing k-mer sizes.

In the case of $k = 2$, the value of θ_j obtained in Fig. 2 corrects the previously reported value of $\theta_j = 0.799(2)$ [29]. Due to the methodology used in this contribution, our estimate of θ_j is expected to be more accurate than that reported previously.[2]

The case of dimers was also studied by using an independent algorithm developed to measure the coverage as a function of time $\theta(t)$. Fig. 3 shows $\theta(t)$ for dimers adsorbed on a simple cubic lattice with $L = 320$. The curve in the figure was averaged over 100 000 independent runs. As is standard in the literature [10], the dimensionless time variable t was defined in terms of the number of deposition attempts per lattice site. In this case, for the M-site lattice, the time step $\Delta t = 1$ corresponds to $0.1M$ deposition attempts. The jamming limit is reached when it is not possible to adsorb any more dimers on the surface, this point is reached for $t \approx 10^2$. A typical jamming configuration in a $16 \times 16 \times 16$ system is shown in the inset of Fig. 3, different colors represent different directions. The value of θ_j obtained in Fig. 3, $\theta_j = 0.9184(1)$, coincides, within numerical errors, with the value calculated from the crossing of the probabilities $W_L(\theta)$. This leads to independent control and consistency check of the numerical value of the threshold θ_j.

2 After publication of Ref. [29], the authors found that the method of filling used in Ref. [29] produces a slightly anisotropic configurations. This effect would be the reason for the observed differences [30].

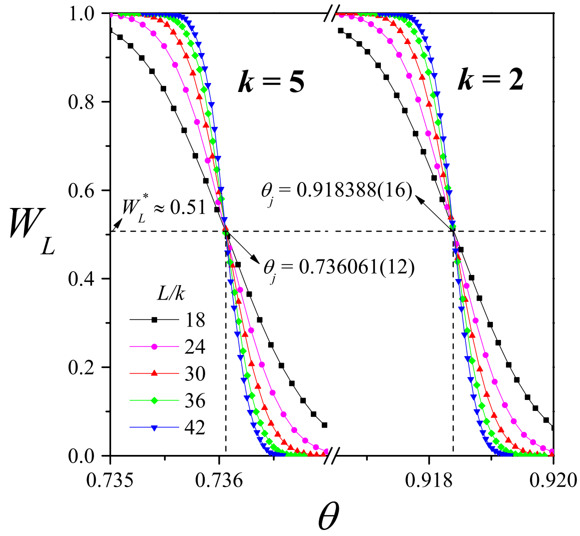

Fig. 2. Curves of W_L as a function of the density θ for two values of k-mer size ($k = 2$, curves on the right, and $k = 5$, curves on the left) and lattice sizes ranging between $L/k = 18$ and $L/k = 42$. Horizontal dashed line shows the W_L^* point. Vertical dashed lines denote the jamming thresholds in the thermodynamic limit.

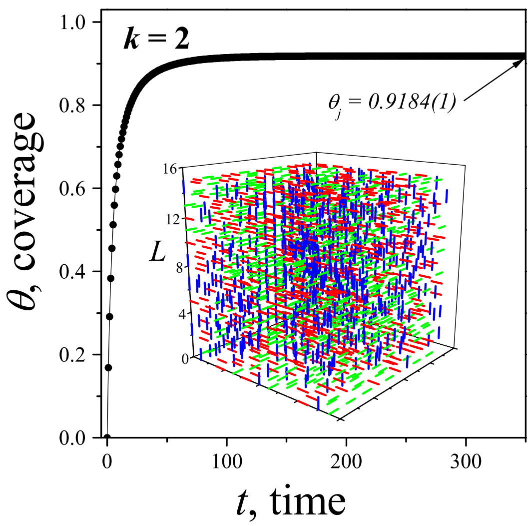

Fig. 3. Numerical values of coverage θ as a function of time t for dimers adsorbed on a simple cubic lattice with $L = 320$. Inset: Typical jamming configuration corresponding to dimers adsorbed on a $(16 \times 16 \times 16)$ lattice. Different colors represent different directions. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

The procedure of Fig. 2 was repeated for k ranging between 2 and 64, and the results are shown in Fig. 4 (solid circles) and second column of Table 2. The points corresponding to k =32, 48 and 64 were calculated for three relatively small values of L/k (12, 18, 24). The jamming coverage decreases upon increasing k. At the beginning, for small values of k, the curve rapidly decreases. However, it flatten out for larger values of k and finally asymptotically converges toward a definite value as $k \to \infty$.

Following a similar scheme to that proposed in Ref. [15] for straight rigid rods on 2D square lattices, the curve of θ_j vs. k was fitted to a function

$$\theta_j(k) = A_1 + \frac{A_2}{k} + \frac{A_3}{k^2} \quad (k \geq 6), \tag{4}$$

being $A_1 = \theta_j(\infty) = 0.4045(19)$ the result for the limit coverage of a simple cubic lattice by infinitely long k-mers. In addition, $A_2 = 2.63(5)$ and $A_3 = -5.2(2)$. As shown in the upper inset of Fig. 4, the plot of θ_j as a function of $1/k$ allows a better visualization of the limit value $\theta_j(\infty)$.

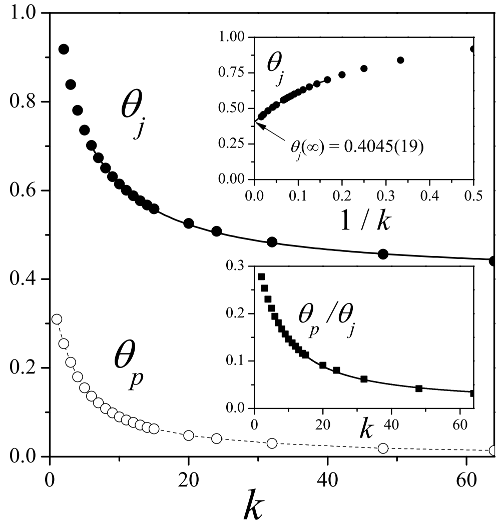

Fig. 4. The thresholds θ_j (solid circles) and θ_p (open circles) as a function of k. The values of θ_p correspond to data reported in Ref. [31]. Lower inset: Ratio θ_p/θ_j as a function of k. Upper inset: θ_j as a function of $1/k$. The solid lines represent the best fits of the numerical values as indicated in the text.

Table 2
Values of θ_j, θ_p and θ_p/θ_j (as indicated in the text) for isotropic rods on a simple cubic lattice and k ranging from 2 to 64.

k	Isotropic rods		
	θ_j (W_L's)	θ_p Ref. [31]	θ_p/θ_j
2	0.918388(16)	0.2555(1) [29]	0.2777
3	0.838860(14)	0.2129(1)	0.2538
4	0.780344(16)	0.1800(1)	0.2307
5	0.736061(12)	0.1555(1)	0.2113
6	0.701346(13)	0.1364(1)	0.1945
7	0.673355(14)	0.1218(1)	0.1809
8	0.650282(11)	0.1089(1)	0.1675
9	0.630901(12)	0.0990(1)	0.1569
10	0.614384(15)	0.0901(1)	0.1467
11	0.600130(10)	0.0831(1)	0.1385
12	0.587696(11)	0.0772(1)	0.1314
13	0.576780(14)	0.0714(1)	0.1238
14	0.567044(11)	0.0661(1)	0.1164
15	0.558360(13)	0.0632(1)	0.1128
20	0.525676(18)	0.0478(1)	0.0909
24	0.507750(18)	0.0411(1)	0.0807
32	0.483360(34)	0.0299(1)	0.0620
48	0.456071(32)	0.0191(1)	0.0417
64	0.440655(34)	0.0143(1)	0.0318

Fig. 4 includes also the behavior of the percolation threshold θ_p as a function of k for the system studied here (open circles). The corresponding numerical values, obtained from Ref. [31], are tabulated in the third column of Table 2. The percolation threshold is a decreasing function of k. Combining the results of θ_j and θ_p, the ratio θ_p/θ_j was calculated (see lower inset in Fig. 4 and fourth column of Table 2). As for 2D square lattices, the ratio between percolation and jamming thresholds exhibits a non-universal behavior. In this case, θ_p/θ_j decreases in all range of k and can be fitted to a function

$$\theta_p/\theta_j = B_1 + \frac{B_2}{k} + \frac{B_3}{k^2} \quad (k \geq 6), \tag{5}$$

where $B_1 \approx 0$, $B_2 = 1.87(5)$ and $B_3 = -4.5(3)$ and, consequently, $\theta_p/\theta_j \to 0$ as $k \to \infty$. This result is consistent with the behavior observed for θ_p and large values of k [$\theta_p(k \to \infty) \approx 0$] [31].

4. Conclusions

Random sequential adsorption of straight rigid k-mers deposited on a simple cubic lattice has been studied by numerical simulations and finite-size analysis. Several conclusions can be drawn from the present work.

- A new theoretical scheme to determine jamming thresholds was introduced here. The method relies on the definition of the probability $W_L(\theta)$ that a lattice composed of $L \times L \times L$ elements reaches a coverage θ. The value of θ_j can be obtained from the crossing point of the curves of W_L for different lattice sizes. Comparisons with rigorous analytical results demonstrated the method's accuracy. Further applications to more complex RSA problems, such as continuum adsorption, deposition objects of different sizes and shapes, and multilayer adsorption, would in principle be feasible. In this sense, the 3D adsorption of k-mers layer by layer could be an interesting topic for future research.
- For the first time, the jamming coverage dependence on the k-mer size has been reported. On the basis of the behavior of θ_j vs. k in the range $2 \leq k \leq 64$, and the best fit to this curve, the expression $\theta_j(k) = A_1 + A_2/k + A_3/k^2$ was found, being $A_1 = \theta_j(\infty) = 0.4045(19)$ the result for the limit coverage of a simple cubic lattice by infinitely long k-mers. In addition, $A_2 = 2.63(5)$ and $A_3 = -5.2(2)$.
- In the case of dimers ($k = 2$), the value of $\theta_j = 0.918388(16)$ obtained in the present paper corrects the value $\theta_j = 0.799(2)$ calculated by Tarasevich and Cherkasova [29]. This important finding was corroborated by using an independent algorithm developed to measure the coverage as a function of time $\theta(t)$.
- Finally, and based on previous investigation from our group [31], the possible relationship between percolation threshold and jamming coverage was examined. The results showed a non-universal behavior for the ratio θ_p/θ_j, which decreases upon increasing k and tends to 0 for large values of k.

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