On the random filling of $\mathbb{R}^d$ by non-overlapping d-dimensional cubes

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Abstract

We compute the time-dependent coverage in the random sequential adsorption of aligned d-dimensional cubes in $\mathbb{R}^d$ using time-series expansions. The seventh-order series in 2, 3 and 4 dimensions is resummed in order to predict the coverage at jamming. The result is in agreement with Monte-Carlo simulations. A simple argument, based on a property of the perturbative expansion valid at arbitrary orders, allows us to analytically derive some generalizations of the Palásti approximation.

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1 Introduction

Random sequential adsorption (RSA) is a model for the random adsorption of non-overlapping objects on a surface. RSA is a conceptually simple model for irreversible behaviour. It has a rich variety of forms which are explained, for example, in the review paper of J.W. Evans [1]. Here, we consider the random filling of $\mathbb{R}^d$ by non-overlapping aligned d-dimensional cubes. This is a simple model, whose one-dimensional version has been solved since a long time [2]. In comparison, the level of understanding of this model in higher dimensions is low. It has been studied through 2, 3 and 4 dimensional Monte-Carlo (MC) simulations and through a 2-d perturbative analysis [3]. A density series expansion [4] can also be used to analyze this model. It is equivalent to a direct perturbative expansion of the coverage in the time variable.

A striking particularity of the model concerns the behavior of the fraction of space, $\theta_d$, filled at jamming. $\theta_d$ is numerically found to be close to the jamming coverage in one dimension raised to the d-th power. The relation $\theta_d = \theta_1^d$, proposed long ago by Palásti [5], is, in fact, one of the motivations of some MC experiments with an increasing precision. Nowadays, they allow us to rule out this exact relation. Nevertheless, as an approximation it appears to have a puzzling accuracy, e.g. of the order of 0.5% in two dimensions [6, 7].

The Palásti relation is one of our main interests. More generally, we undertake the reconstruction of the coverage $\theta_d(T)$ in 2, 3 and 4-d using as basic input its perturbative expansion (PE) in the time variable $T$.

In the next section, this expansion is derived up to seventh order by using diagramatic methods already given in Ref.[3, 8]. The complexity of this expansion, even at moderate orders (6th or 7th), necessitates a computer algorithm. We will only give the results in 2, 3 and 4- (because the full analytical result in d is too long).

Using standard approximation methods, which combine PE and asymptotic ($T \to \infty$) expansions, we extend the analysis of $\theta_d$ of Ref.[3] to 3 and 4 dimensions. This analysis is done in section 2, where it is found that $\theta_d \approx \theta_1^d$ with some decreasing precision as $d$ increases. In section 3, we provide a functional understanding of the Palásti relation as a consequence of a particular property of the PE, valid at arbitrary orders. Finally, by taking into account the asymptotic regime, we give some improved generalizations of the Palásti approximation.

2 The d-dimensional perturbative expansion

We will study the RSA of hypercubes in the continuum limit on $\mathbb{R}^d$. This is defined as the scaling limit of RSA of cubes on a d-dimensional hypercubical...
lattice. The discrete model, in which the size of the adsorbed cubes enters as a parameter, has its own interest and it has been also investigated [2, 3, 7, 9] in 1 and 2-dimensions. At small sizes, the Palásti approximation works perfectly, and more generally, the model is easier to handle [3, 10]. We consider here the discretisation as a convenient regularisation of the PE from which we will keep, order by order, the scaling limit.

We follow the definition of the discrete model given in Ref.[7]. The adsorbed cubes have their edges parallel to the axes of the lattice, and they are made of \(k^d\) unit cells, \(k\) being an integer. Their volume \(l^d = (kb)^d\) enters in the definition of the dimensionless time, \(T, T = \rho t^d\), in which \(b\) is the lattice spacing. \(\rho\) is the attempt rate to adsorb cubes per volume unit. On the lattice, the number of attempts per site after time \(t\) is thus \(\rho tb^d\), i.e. \(T/k^d\). Therefore the coverage can be written as

\[
\theta_d(k, T) = k^d \rho_d(k; T/k^d) \tag{1}
\]

The coverage \(\theta_d(T)\) in the continuum limit is then given by the limit of \(\theta_d(k, T)\) as \(k\) goes to infinity with \(T\) fixed.

The PE of \(\rho_d(k, t)\) is:

\[
\rho_d(k, t) = t - \sum_{n \geq 2} C_n(k, d) \frac{(-t)^n}{n!} \tag{2}
\]

where the first order in time is \(t\), because the first adsorption attempt on an empty lattice is always accepted. We refer to Ref.[3] for a comprehensive and complete derivation of the PE. It starts with an operator formalism of the RSA mechanism and ends up with diagrammatic rules, that are sketched below, for computing the coefficients \(C_n(k, d)\).

To generate the n-th order diagrams, one expands the expression:

\[
\prod_{i=2}^{n} (1 - i - 1 \prod_{j=1}^{i-1} (1 - K_{i, j}))
\]

The set of variables defining a deposition is denoted by \(i\), and \(K_{i, j}\) is a hard Mayer function (\(K_{i, j} = 1\) only if \(i\) and \(j\) are overlapping objects, 0 otherwise). All monomials of this polynomial are connected labeled graphs which are regrouped in classes of the same topology. Thus, they appear with combinatorial weights \(w_{n,i}\) in such a way that

\[
C_n(k, d) = \sum_i w_{n,i} I(\Gamma_{n,i}) \tag{3}
\]

\(I(\Gamma_{n,i})\) is the graph contribution which embodies the details of the model.

In order to compute \(I(\Gamma_{n,i})\), one has to do the summation over \(\{x_l\}\), the n vertices of \(\Gamma_{n,i}\). At this stage, the vertices, \(\{x_l\}\), are to be understood as d-dimensional lattice points, i.e. \(x_l \equiv (x^1_l, x^2_l, \ldots, x^d_l)\), because a d-dimensional
aligned hypercube can be characterized by the position of a distinguished point, for example a corner. Finally, the graph contribution is

\[ I(\Gamma_{n,i}) = \sum_{\{x\}} \prod_{p} K(x_{lp}, x_{mp}) \equiv I_{n,i}(k, d) . \]  

(4)

The product \( \prod_{i} \) is done on the links of \( \Gamma_{n,i} \) and the sum is over all the lattice degrees of freedom for every variable \( x_{l} \), except one which can be frozen at the origin by translational invariance.

Any graph contribution, \( I_{n,i}(k, d) \), is thus the \( d \)-th power of the contribution of the same graph in one dimension.

\[ I_{n,i}(k, d) = I_{n,i}(k, 1)^{d} . \]  

(5)

This simple property, that we shall use in the next section, is a consequence of the construction rule (4) and of the factorization property of hard Mayer functions for hypercubes: \( K_{d}(x, y) = \prod_{i=1}^{d} K_{1}(x^{i}, y^{i}) \).

Finally, in the continuum limit, the PE of \( \theta_{d}(T) \) is, from (1) and (2):

\[ \theta_{d}(T) = T - \sum_{n \geq 2} C_{n}(d) \frac{(-1)^{n}}{n!} T^{n} \]  

(6)

where, from (3-5)

\[ C_{n}(d) = \sum_{i} w_{n,i} \lim_{k \to \infty} \{ k^{1-n} I_{n,i}(k, 1) \}^{d} = \sum_{i} w_{n,i} I_{n,i}^{d} \]  

(7)

The coefficients \( C_{n}(d) \) are well defined, because \( I_{n,i}(k, 1) \) are polynomials of degree \( (k-1) \) in the variable \( k \) that we have analytically computed. Their leading coefficients are denoted by \( I_{n,i} \) in Eq.(7).

We have thus, stored in our computer algorithm, all the \( I_{n,i} \) and \( w_{n,i} \) necessary to know \( C_{n}(d) \) analytically in \( d \), up to the seventh order in \( T \). Nevertheless we do not explicitly write them here due to their number (for \( n = 7 \) there are about \( 10^{3} \) graphs). At fixed \( d \), \( C_{n}(d) \) are just the rational numbers that we give in Table I for \( d = 2, 3 \) and 4. We have performed various tests of our expansion. The simplest is for \( d = 0 \) where \( \theta_{0}(T) = 1 - e^{-T} \). For \( d = 1 \) the exact solution is known for any \( k \) and in the scaling limit \( \theta_{1}(T) \) reads:

\[ \theta_{1}(T) = \int_{0}^{T} dt e^{-2g(t)} \quad \text{with} \quad g(t) = \int_{0}^{t} dx \frac{1 - e^{-x}}{x} . \]  

(8)

The agreement was checked analytically in \( k \), including the scaling limit. Finally, in 2-d, we recover the expansions given for \( k = 2 \) and \( k = \infty \) in Ref.[3].

These coefficients \( C_{n}(d) \) can be used in various ways to approximate \( \theta_{d}(T) \). We give, in the following, our analysis. In addition to the PE Eq.(3), one knows how the jamming limit, \( \theta_{d} = \theta_{d}(T = \infty) \), is approached [1, 7].

\[ \theta_{d}(T) - \theta_{d} = O[\ln T^{d-1}/T] \]  

(9)
Standard methods which embody these two kinds of information have shown, in other contexts, their ability to reasonably approximate the coverage even in its asymptotic regime [3, 8].

We give, as a first attempt, the predictions for $\theta_d$ from a straightforward extension of the method used in Ref. [3]. One uses the mapping $\omega(T)$,

$$
\omega(T) = 1 - \frac{1 + \ln (1 + \frac{b-1}{d-1}T)^{d-1}}{1 + bT}, \quad b \geq 1
$$

which behaves as $T$ for small $T$ and as $1 - \ln T^{d-1}/bT$ for $T \to \infty$, to convert the PE, Eq.(6), into a power series of $\omega$. The Padé approximants formed from this series, when evaluated at $\omega = 1$, give a $\theta_d$ consistent with the behaviour (9), for any choice of the $b$ which is used as a variational parameter.

Let us first recall the 2-d results from Ref.[3]. Among the sixth and seventh order approximants, $[5,2]$, $[3,4]$, $[4,2]$ and $[3,3]$ cross near $b = 1.30$ yielding $\theta_2 = 0.52625(5)$, in agreement with the data of Ref.[3], $\theta_2 = 0.562009(4)$. As $d$ increases, we expect some continuity, and we indeed observe a crossing at $b = 1.40$ for the $[4,2]$ and $[3,4]$ approximants, and for the remaining ones $[5,2]$ and $[3,3]$ there is a crossing at $b = 1.45$. Nevertheless, the prediction, now, loses its accuracy, because these intersections give $\theta_3 = 0.40$ and 0.46, respectively. Moreover, in 4-d this intersections disappear and the best determination of $b$ is $b \simeq 2.3 - 2.5$.

At this value, the $[4,3]$, $[3,4]$, $[3,3]$ and $[3,2]$ Padé-approximants give $\theta_4 = 0.30(1)$. These values are consistent with the MC data since $\theta_3 = 0.4227(6)$, 0.4262 , 0.430(8) and 0.422(8) according to the Refs. [12], [13], [14], [15] respectively and $\theta_4 = 0.3129$ or 0.3341 from Ref. [12] and Ref. [13] respectively. Obviously, the Palásí values, $\theta_1^d = 0.7476, 0.5589, 0.4178$, and 0.3124, $d = 2, 3, 4$ cannot be discussed (except in 2-d) within the poor precision of the previous results. Nevertheless, we show in the following section that perturbation theory is able to provide a functional insight into the Palásí approximation. It goes beyond a numerical coincidence.

3 Variational approach to the Palásí approximation

The RSA mechanism of deposition of aligned cubes is characterized by power law behaviors in $d$ for both the coverage at jamming, in the Palásí approximation, and the graph-integrals, in the framework of the perturbative expansion. We shall give in this section a variational derivation of this puzzling approximation together with an extension to finite values of $T$.

Starting with the representation of $\theta_d(T)$ given by Eqs.(6,7),

$$
\theta_d(T) = \sum_{i, n \geq 1} \Lambda_{i,n} I_{i,n}^d T^n
$$

(11)
we write, in order to emphasize the role of the power law behavior (3), that

\[ I_{i,n}^{d} = (I_{i,n} - ab^{n})^{d} + \sum_{r=0}^{d-1} (-1)^{d-r+1} \frac{d!}{r!(d-r)!} a^{d-r} b^{n(d-r)} I_{i,n}^{r} \quad (12) \]

This is nothing but a binomial identity, valid for any \( a \) and \( b \). We can insert (12) into (11) to obtain a two-component representation of \( \theta_{d}(T) \):

\[ \theta_{d}(T) = \tilde{\theta}_{d}(T) + \epsilon_{d}(T) \quad (13) \]

where

\[ \tilde{\theta}_{d}(T) = \sum_{r=0}^{d-1} (-1)^{d-r+1} \frac{d!}{r!(d-r)!} a^{d-r} \theta_{r}(b^{d-r}T) \quad (14) \]

\[ \epsilon_{d}(T) = \sum_{i, n \geq 1} \Lambda_{i,n} (I_{i,n} - ab^{n})^{d} T^{n} \quad (15) \]

In the representation (13-14), \( \tilde{\theta}_{d}(T) \) involves all the lower dimensionality coverages, and the a priori \( d \)- and \( T \)- dependent parameters \( a, b \). A natural way to choose of \( a \) and \( b \) is thus to look for maxima of \( \tilde{\theta}_{d}(T) \), i.e. minima of the perturbative component \( \epsilon_{d}(T) \).

Beginning with the 2-d case, one finds that \( \tilde{\theta}_{2}(T) = 2a\theta_{1}(bT) - a^{2}\theta_{0}(b^{2}T) \), has a maximum with respect to \( a \):

\[ \theta_{2}^{\ast}(T) = \frac{\theta_{2}^{1}(bT)}{\theta_{0}(b^{2}T)} \]

for \( a = \theta_{1}(bT)/\theta_{0}(b^{2}T) \), i.e. a value of \( a \) where \( \epsilon_{1} \) vanishes. If \( \epsilon_{2} = 0 \), this is nothing but the Palásti approximation at \( T = \infty \).

This result can be extended to any \( d \): Under the assumption \( \epsilon_{r} = 0 \) for \( 1 \leq r \leq d-1 \), \( \tilde{\theta}_{d}(T) \) has an extremum \( \theta_{d}^{\ast}(T) \):

\[ \theta_{d}^{\ast}(T) = \frac{\theta_{d}^{1}(b^{d-1}T)}{\theta_{d-1}^{1}(b^{d}T)} = \frac{\theta_{d}^{2}(bT)}{\theta_{d-2}^{2}(b^{2}T)} \]

for \( a = \theta_{1}(b^{d-1}T)/\theta_{0}(b^{d}T) \).

This is simply because the minimizing condition can be reexpressed in terms of \( \epsilon_{d-1} \) if the \( a \) parameter is assumed to be \( d \)-independent. The relation is:

\[ \frac{\partial \epsilon_{d}(T)}{\partial a} = -d \epsilon_{d-1}(bT) \ . \]

We are thus led to write the PE (13) of \( \theta_{d}(T) \) under the form:

\[ \theta_{d}(T) = \theta_{d}^{\ast}(T) + \epsilon_{d}(T) \quad (16) \]

where we choose

\[ \theta_{d}^{\ast}(T) = \frac{\theta_{d-1}^{2}(bT)}{\theta_{d-2}^{2}(b^{2}T)} \quad (17) \]
as a convenient generalization of the Palásti approximation on the whole time-range. To go further, one can eliminate $T$, order by order, in terms of $\theta_\ast^d(T)$ ($\theta_\ast^d(T) \sim T$ at $T = 0$ and $\theta_\ast^d(T)$ increases if $b \geq 1$), and then, substitute this expression in $\epsilon_d(T)$, ($\epsilon_d(T) \sim T^2$). One finally obtains $\theta_d(T)$ as a power series in $\theta_\ast^d(T)$, i.e. as a perturbative expansion where the first order is the Palásti approximation.

In this approach, it remains to take into account the asymptotic behaviour. One observes that $\frac{d}{dT}(T \theta_d(T))$ and $\theta_\ast^d(T)$ reach their asymptotic limit in the same way, $O[\ln T^{d-2}/T]$. On the other hand, $\frac{d}{dT}(T \theta_d(T))$ is perturbatively of the form $O(T^0)$, so that the previous arguments apply. Thus we propose:

$$\theta_d(T) = \frac{1}{T} \int_0^T dT \{\theta_\ast^d(t) + \epsilon_d(t)\}$$

as an approximation of $\theta_d(T)$ with a minimal component $\epsilon_d(T)$. The jamming coverage is then $\theta_d = \theta_\ast^d(\infty) + \epsilon_d(\infty)$, where

$$\theta_\ast^3(\infty) = \frac{\theta_2^2}{\theta_1} = 0.4225$$
$$\theta_\ast^4(\infty) = \frac{\theta_3^2}{\theta_2} = 0.3176.$$ 

We have to verify, in PE, that $\epsilon_d(T)$ is small and that a precise determination of the $b$ parameter can be done. We express $\epsilon_d(T)$ as a power series in $\theta_\ast^d(T)$ and form its Padé table. We find $\epsilon_d(T = \infty)$ from these approximants when $\theta_\ast^d = \theta_\ast^d(\infty)$. Here, $b$ is used as a variational parameter.
In 2-d, 3-d and 4-d, we find a minimal dispersion of the [3,4], [4,3], [5,2], [4,2], 
[3,3], [2,4], [2,3] and [3,2] Padé at the values:

\[
d = 2 \quad b \simeq 1.32 \quad \epsilon_2(\infty) = 0.004(2) \\
 d = 3 \quad b = 1.35(3) \quad \epsilon_3(\infty) = 0.00(1) \\
 d = 4 \quad b = 1.40(5) \quad \epsilon_4(\infty) = 0.01(1) .
\]

\(\epsilon_d(T)\) is found to be positive on the whole physical region (except near \(T = \infty\) for some marginal solutions in 3-d) and is of the order of \(\epsilon_d(\infty)\). We finally obtain that

\[
\theta_3 = 0.42(1) \quad \text{and} \quad \theta_4 \simeq 0.32(1) .
\]

In most of the cases, the positivity of \(\epsilon_d(T)\) indicates the violation of the Palásti conjecture.

\section{Conclusion}

This study has been devoted to the RSA coverage of \(R^d\) by \(d\)-dimensional aligned hypercubes, especially in 3 and 4 dimensions. For these dimensions, we have explicitly presented the perturbation result to seventh-order. The series in other dimensions and any discrete case are available upon request. The continuum model at long times is expected to be more difficult to describe than its discrete analogue. This is supported by the fact that we cannot predict \(\theta_d\) beyond a moderate accuracy with our perturbative information. Nevertheless, we indicate, in this framework, how one can approach the Palásti conjecture and its corrections.

To conclude, we want to mention that the simple rule \(\theta_d \sim \theta_{d-1} + \theta_{d-2}/\theta_{d-3}\) applies to various discrete models, in particular RSA on \(d\)-dimensional lattice with nearest-neighbour exclusion \cite{16}, where \(\theta_0 = 1/2\) and \(\theta_1 = (1 - e^{-2})/2\). As \(d\) runs from 2 to 6, we obtain the sequence of approximations (0.374, 0.307, 0.254, 0.229, 0.206) which are quite close to the corresponding data (0.364, 0.304, 0.264, 0.233, 0.209).
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| n | d=2 | d=3 | d=4 |
|---|-----|-----|-----|
| 2 | 4   | 8   | 16  |
| 3 | 23  | 101 | 431 |
| 4 | 168 | 45920| 432032|
| 5 | 105895| 30622397| 7785588283|
|   | 72   | 864 | 10368|
| 6 | 6709687| 11820085427| 17179096575091|
|   | 450  | 13500| 405000|
| 7 | 1385692277| 5815177641671| 183314989218969359|
|   | 8100 | 233280| 65610000|

Table I: Coefficients of the time-series expansion of the coverage \( \theta_d(T) \) defined by

\[
\theta_d(T) = T - \sum_{n \geq 2} \frac{(-1)^n}{n!} C_n(d) T^n .
\]