Random sequential adsorption and diffusion of dimers and $k$-mers on a square lattice

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We have performed extensive simulations of random sequential adsorption and diffusion of $k$-mers, up to $k = 5$ in two dimensions with particular attention to the case $k = 2$. We focus on the behaviour of the coverage and of vacancy dynamics as a function of time. We observe that for $k = 2, 3$ a complete coverage of the lattice is never reached, because of the existence of frozen configurations that prevent isolated vacancies in the lattice to join. From this result we argue that complete coverage is never attained for any value of $k$. The long time behaviour of the coverage is not mean field and non analytic, with $t^{-1/2}$ as leading term. Long time coverage regimes are independent of the initial conditions while strongly depend on the diffusion probability and deposition rate and, in particular, different values of these parameters lead to different final values of the coverage. The geometrical complexity of these systems is also highlighted through an investigation of the vacancy population dynamics.

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I. INTRODUCTION

Random Sequential Adsorption (RSA) of hard particles on lattice is employed to describe a wide class of irreversible phenomena in physics, chemistry and biology. The prototype of this process is the dimerization of monomers on a chain. Each monomer occupies initially a site of the chain and can react reversibly with one of its neighbors to form a dimer: irreversible dimerization takes place sequentially until no more pairs of neighbor monomers are available. According to Flory, this process can be viewed as the random sequential adsorption of non-overlapping dimers onto a one dimensional lattice: a dimer can be irreversibly deposited if two neighbor free sites are available. At a given stage only single isolated sites will be available and further deposition will be no more allowed. At that stage, the density of dimers is finite and takes a non random value in the limit of infinite system, the so called jamming coverage $\theta_J$. This study was followed by the introduction and investigation of a variety of RSA processes involving different objects, lattices and rules. The quantities of interest are in general the time behavior of the coverage $\theta(t)$ and its jamming value. Exact solutions are available in few cases, typically for dimers in one dimension, whereas only numerical or approximate results exist for more complex objects and in higher dimension.

In recent years, the class of systems for which RSA yields effective description has widened by the observation that in many experimental processes particles also diffuse on the lattice after deposition. Diffusional Relaxation (DR) affects the spatial distribution of free sites, allowing for further deposition of particles whenever neighbor empty sites are created. This produces a different time behavior of $\theta(t)$ and different values of the asymptotic coverage $\theta(\infty)$. The effect of DR on RSA models has been investigated by many authors (see Refs. therein), often displaying the emergency of non trivial behaviors both for $\theta(t)$ and $\theta(\infty)$. In the case of dimers on a line, for instance, the introduction of DR produces a very slow increase of coverage with time, terminating eventually with the complete filling $\theta(\infty) = 1$. Exact results 4 and numerical simulations 5 show that, after a transient depending on the initial conditions, $1 - \theta(t)$ decreases asymptotically as $t^{-1/2}$, and thus an infinite time is needed to reach the completely filled state. The exponent 1/2 is due to the relevance of statistical fluctuations in the formation of neighbors free sites, so that any computation based on mean field treatments is not effective. It is also for this reason that solutions of RSA problems with Diffusion (RSAD) are very rare and usually limited to one dimensional systems (see Refs. therein) and some particular cases. Some two dimensional models have been recently investigated 6 and features sometimes different from the one dimensional case have been observed. Eisenberg and Baram have found a stable asymptotic coverage $\theta(\infty) < 1$ for the

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RSAD of crosses on a square lattice, whereas such a behavior is not observed in the case of squares in which the complete coverage is eventually reached for an infinite system, although through coexistence of different phases. The pure RSA of dimers in two dimensions was first investigated by Nord and Evans and was later extended also considering anisotropic deposition rates. Grigera et al. computed the asymptotic coverage \( \theta(\infty) \) in presence of diffusion and rotation of dimers and found that it is less than unity. They also observed indications for an exponential approach to the limit coverage \( \theta(\infty) \).

In this paper we present extensive Monte Carlo (MC) simulations of the RSAD of linear particles on a square lattice. In the systems under consideration hard particles, the \( k \)-mers, are dropped on an initially empty square lattice and are allowed to diffuse. Each \( k \)-mer occupies \( k \) consecutive sites and cannot overlap to the others. We consider different initial conditions as well as a set of different deposition and diffusion rates and probabilities. Whereas the former only have influence on initial and intermediate stages of the coverage, the latter influence the late stage of dynamics and different values may lead to different final states. In fact, we show that the value of the final coverage depends on these parameter and, as expected, decreases with increasing \( k \). In this paper the focus will be mainly on the case \( k = 2 \), since simulations show that its main features are also shared by systems with higher values of \( k \), while the time required for simulations rapidly increases with \( k \). For this reason Secs. II-V are dedicated mainly to the RSAD of dimers.

In Sec. II we define the model and the procedure for the Monte Carlo simulation. We also test possible algorithms to save computational time when simulating such slow relaxation processes, and describe the one we adopted. Section III is devoted to investigate the dynamical features of the coverage process as well as of the final state and of their dependence on the deposition and diffusion rates. In Sec. IV the results are put in relation with the behavior of the vacancies in the lattice, whose statistical behavior is also studied. The \( k \)-mers RSAD model with \( 2 < k \leq 5 \) is addressed in Sec. V, where similarities and differences with the case \( k = 2 \) are pointed out. Finally sec. VI is devoted to a summary of our main findings and to some general conclusions.

II. MODEL AND COMPUTATIONAL DETAILS

A. RSAD dynamics

The process that we have more extensively investigated in this paper consists in the deposition and diffusion of dimers on a square lattice containing \( N = L \times L \) sites with periodic boundary conditions. Each dimer occupies two nearest neighbor sites, or equivalently a bond, and cannot overlap with other dimers. For \( t > 0 \) it is free to diffuse along the direction of its axis towards empty sites. We have assumed deposition to occur with equal probability on horizontal and vertical bonds and we have considered two possible different initial conditions: a) jamming coverage, i.e. \( \theta(0) = \theta_J \) and b) empty lattice, i.e. \( \theta(0) = 0 \). In the case a) the initial stage consists of pure RSA. Dimers are deposited on the lattice until no more free pair of sites is available and the jamming state is reached. The jamming coverage for this kind of model is known from previous work to be \( \theta_J = 0.900654 \). It is reached exponentially and is higher than the corresponding one in one dimension. At this stage deposited dimers are allowed to diffuse and more dimers can be possibly added when suitable space is made available. In case b) the deposited dimers can move from the beginning, so that diffusion in the initial stages largely dominates the deposition process, giving rise to a large amount of vacancies.

TABLE I. Coefficients of fit Eq. (4) for \( \theta(0) = \theta_J \). The curve for \( p = 0.5 \) can be adequately fitted only retaining the terms up to the third order.

| \( p \) | \( r_0 \) | \( r_1 \) | \( r_2 \) | \( r_3 \) | \( r_4 \) |
|---|---|---|---|---|---|
| 0.2 | 0.00150 | 0.040 | 2.167 | 7.447 | 7.456 |
| 0.5 | 0.00110 | 0.104 | 0.607 | -1.105 | -1.105 |
| 0.9 | 0.00120 | 0.252 | 0.702 | -3.983 | 4.799 |

TABLE II. Coefficients of fit Eq. (4) for \( \theta(0) = 0 \).

| \( p \) | \( r_0 \) | \( r_1 \) | \( r_2 \) | \( r_3 \) | \( r_4 \) |
|---|---|---|---|---|---|
| 0.2 | 0.0016 | 0.0628 | 1.358 | 2.152 | -4.836 |
| 0.5 | 0.00084 | 0.1239 | 0.4223 | 0.2943 | -0.3776 |
| 0.9 | -0.0014 | 0.3797 | -0.6506 | 1.021 | -0.4146 |
FIG. 2. Temporal behavior of density of empty sites (solid lines) for $p = 0.2$ (a), $p = 0.5$ (b) and $p = 0.9$ (c). The long dashed lines are fits to the data according to Eq. (1). The initial condition is $\theta(0) = \theta_J$ and the size of the system is $L = 100$.

We have computed the time behavior of the coverage and of the abundance of different kinds of vacancies in the above processes by means of MC simulations. At each time step deposition of dimers is attempted with probability $p$ and diffusion with probability $1 - p$. Both directions of diffusion are equiprobable.Deposition happens at rate $R$, i.e. $pRN$ dimers are tried to be deposited on the average in a MC step and a unitary jumping rate $d = 1$ follows for diffusion by assuming unitary lattice spacing and simulation (micro) time discretization $\delta t = 1/N$.

B. Saving time algorithms

The above dynamics is extremely time consuming to simulate, mainly in the late stage of the process, since due to the exclusion rules most of diffusion and deposition attempts fail. A large amount of computational time can however be saved and dynamics sped up by suitable algorithms that maximize the number of successful events. We have adopted the strategy of storing in memory the dimers that can successfully diffuse along a given direction, $n_{\text{dif}H}$ and $n_{\text{dif}V}$ and the bonds available for deposition $n_{\text{dep}H}$ and $n_{\text{dep}V}$ respectively for the horizontal and vertical directions.

By accounting for the different probabilities and rates at which these actions may take place, it turns out that on the average a successful event occurs with probability

$$p_{\text{succ}} = p \left( \frac{n_{\text{dep}H} + n_{\text{dep}V}}{2N} \right) + \frac{1 - p}{2} \left( \frac{n_{\text{dif}H} + n_{\text{dif}V}}{2N} \right).$$

(1)

Since the law of large numbers is expected to hold (at least for system size large enough), at each step of the simulation the MC time is increased by the amount

$$\Delta t = \frac{1}{p_{\text{succ}}}. \tag{2}$$

We adopted the above method that produces a time saving of about one order of magnitude with respect to the usual MC simulation with equivalent results.

As a further check we have also tested the method adopted by Gan and Wang which is based on the probability distribution of time lags $\Delta t$ between two consecutive successful events

$$P(\Delta t = m) = p_{\text{succ}}(1 - p_{\text{succ}})^{m-1} \tag{3}$$

and also yields results equivalent to those of usual MC. The identity of results confirms the reliability of both of
these methods; however time saving is much more efficient via Eq. (1), since in fact this latter method corresponds to an average of the former over a large number of successful events. Both of them require additional memory occupation for the storage of dimers and bonds that may change their state, but this does not seriously hinder simulations of large systems if compared with required computational time.

![Graph](image)

**FIG. 4.** Comparison of the temporal behavior of density of empty sites for an initial jammed lattice (solid line) and an initial empty lattice (long dashed line), for \( p = 0.5 \) The size of the system is \( L = 100 \).

### III. TIME COVERAGE

#### A. Asymptotic coverage

Mean field approximations are known to work very badly for RSAD of small particles in one dimension. Diffusional dynamics of dimers along the lattice can be described in terms of mobile vacancies, that annihilate through the deposition of a dimer when they meet on neighbor sites. As already mentioned, both exact solutions and numerical simulations show that the process of vacancies annihilation is dominated by random fluctuations of density rather than by average values, thus leading asymptotically to a coverage described by a random walk process, \( \theta(\infty) - \theta(t) \simeq t^{-1/2} \), opposed to the \( t^{-1} \) coverage estimated by mean field. Random walk process with regular exponent 0.5 concerns the dynamics of vacancies, since particles suffer an anomalous random walk with exponent \( \leq 0.5 \), as discussed in Ref. [3]

Mean field is generally speaking expected to work better in higher dimensions. However studies on RSAD of different objects in \( d = 2 \) found that fluctuations of isolated vacancies still dominate relaxational dynamics [4]. Investigations of Eisenberg and Baram [5] on RSAD of monomers particles with nearest neighbors and next nearest neighbors exclusion have shown that \( \theta(\infty) - \theta(t) \simeq t^{-1/2} \) to the leading order in the asymptotic regime. Systems made by superposition of several one dimensional lattices to form multilayer structures, that allow for both intra-layer and inter-layer diffusion, have been found to behave in a similar fashion [6].

Our simulations show that also the RSAD of dimers in \( d = 2 \) displays a regime in which \( t^{-1/2} \) dominates, thus implying the recombination of one-site vacancies to be the main process, in analogy with the \( d = 1 \) case (we will turn to this point in Sec. [7]). This result has shown to be independent from the parameter values and initial conditions chosen for the simulation. Nevertheless in \( d = 2 \) there is a stage of the covering process at which the dynamics definitely changes. At that stage we observe that the annihilation rate slows down and correspondingly the filling slowly stops. The system is prevented to reach the complete coverage by the formation of isolated vacancies that are trapped at one site or along closed loops. As firstly pointed out by Grigera et al. [14] this seems to be a genuine property of the system, since independent of the lattice size. The results displayed in Fig. [1] where systems of different sizes approach the same asymptotic coverage \( \theta(\infty) \simeq 0.998 \), support the idea of a dynamically jammed state related to local configurations of the system. We find, in agreement with the authors in [14], that finite-size effects are reasonably negligible for \( L \geq 40 \).

In Fig. [2] we show the coverage behavior for different choices of \( p \). Different values of \( \theta(\infty) \) are reached, implying that different choices of \( p \) and \( R \) can lead to different final coverages and can therefore be regarded as different quench rates of an initially annealed system.

![Graph](image)

**FIG. 5.** Temporal behavior of density of empty sites for infinite deposition rate (solid line). The long dashed line is a fit to the data according to Eq. (1). For comparison we also show the curve for \( p = 0.5 \) (dot-dashed line). The initial condition is \( \theta(0) = 0 \) and the size of the system is \( L = 100 \).
FIG. 6. Temporal behavior of different kinds of vacancies for $p = 0.2$ (a), $p = 0.5$ (b) and $p = 0.9$ (c), for $\theta(0) = \theta_J$: the solid line is the total density of empty sites, the dotted line is the density of one-site vacancies, the dashed line is the density of sites formed by two-site vacancies and the dot-dashed line is the density of sites formed by non one-site vacancies. The size of the system is $L = 100$.

B. System evolution

Fig. 3 displays the time coverage when $\theta(0) = 0$. After an initial stage ($1 < t < 10^2$) in which deposition attempts dominate and the density progressively increases, the system approaches the same regime as for $\theta(0) = \theta_J$.

A fit of this curve to the results of numerical simulations shows that the terms beyond the fourth in the series can be neglected (cfr. Table III) and is displayed in Fig. 3. As expected, the effect of an increased diffusion is to decorrelate the system, and thus to produce a faster coverage, as shown by higher weights for the $t^{-1}$ term. The coverage depends on the initial condition only at early times.

![Graphs showing temporal behavior of vacancies](image)

FIG. 7. Temporal behavior of different kinds of vacancies for $p = 0.2$ (a), $p = 0.5$ (b) and $p = 0.9$ (c), for $\theta(0) = 0$: the solid line is the total density of empty sites, the dotted line is the density of one-site vacancies, the dashed line is the density of sites formed by two-site vacancies and the dot-dashed line is the density of sites formed by non one-site vacancies. The size of the system is $L = 100$.

TABLE III. Coefficients of fit Eq. (6) for $\theta(0) = \theta_J$.

| $p$ | $x_0$ | $x_1$ | $x_2$ | $x_3$ | $x_4$ |
|-----|-------|-------|-------|-------|-------|
| 0.2 | 0.00131 | 0.093 | 0.834 | 2.932 | 2.833 |
| 0.5 | -0.00158 | 0.174 | 0.602 | -2.916 | 3.016 |
| 0.9 | 0.00117 | 0.249 | 0.575 | -4.801 | 6.437 |

TABLE IV. Coefficients of fit Eq. (7) for $\theta(0) = \theta_J$.

| $p$ | $y_0$ | $y_1$ | $y_2$ | $y_3$ | $y_4$ |
|-----|-------|-------|-------|-------|-------|
| 0.2 | 0.00222 | 0.0509 | 0.371 | -0.446 |
| 0.5 | 0.00306 | -0.0946 | 0.925 | -1.484 |
| 0.9 | -0.00010 | -0.0033 | 0.0896 | -0.1268 |
IV. VACANCY DYNAMICS

As mentioned above, RSAD can be described in terms of reaction and diffusion of vacancies on the lattice. In the general case of $k$–mers with diffusion coefficient $D$ and deposition rate $R$, one deposition corresponds to the annihilation of $k$ vacancies on contiguous sites.

If one adopts this point of view the system behavior appears to be ruled by the dynamics of vacancies. In order to give a full account of this dynamics, knowledge of vacancies correlation functions is necessary. Nevertheless a qualitative understanding of some of the main dynamical features can be obtained also through the investigation of the sole vacancy population.

A. Initial jammed state

Fig. 3 compares the coverage behavior with the change in time of vacancy population for different values of $p$. Solid and dotted lines represent respectively the total fraction of empty sites, $1 - \theta$, and of one-site vacancies, $\rho_1$. At early and intermediate times these two quantities display some differences, more visible at lower values of $p$ where formation of many-site vacancies is enhanced by diffusion. For a jammed initial state, an increase in two-site vacancies takes place in the early stage of the process and, for low values of $p$, can last for several decades both in density ($\rho_2$) and time. In fact, two dimensional geometry allows for the formation of many different kinds of voids; besides vertical and horizontal two-site vacancies, there can be $L$ many-site vacancies with different orientations, square vacancies and so on.

| TABLE V. Jamming densities of $k$–mers. |
|----|---|
| $k$ | $\theta_j$ |
| 4 | 0.8470 |
| 5 | 0.7877 |

TABLE VI. Coefficients of fit Eq. (5) for $k$–mers when $\theta(0) = \theta_j$.

| $k$ | $r_0$ | $r_1$ | $r_2$ | $r_3$ |
|----|----|----|----|----|
| 2 ($p = 0.5$) | 0.00110 | 0.104 | 0.607 | -1.105 |
| 3 ($p = 0.5$) | 0.00075 | 0.329 | 1.562 | -8.275 |
| 4 ($p = 0.5$) | 0.00036 | 0.682 | 0.399 | -6.709 |
| 5 ($p = 0.5$) | 0.00228 | 1.013 | -1.774 | 0.8537 |
| 2 ($p = 0.1$) | 0.00126 | 0.027 | 4.174 | -19.30 |
| 3 ($p = 0.1$) | 0.00036 | 0.384 | 6.483 | -47.35 |
| 4 ($p = 0.1$) | -0.00031 | 1.033 | 0.474 | -17.13 |
| 5 ($p = 0.1$) | 0.00075 | 1.663 | -6.330 | 17.38 |
An initially empty lattice produces early and intermediate stages in vacancy dynamics different from the jammed case. Whereas the late stage is still dominated by recombination of one-site vacancies, an initial stage exists in which the abundance of this kind of defects increases monotonically up to reach about the same density of many-site vacancies, as displayed in Fig. 6. On the other hand, right at that stage the latter’s population starts to be less than that of one-site vacancies. Interestingly single empty sites rule the dynamics for $t > 10^2$, that is, as already explained, the same time range in which density no longer depends on initial conditions (cfr. Fig. 4). It can be noted that two-site vacancies grow at pace with one-site ones up to a maximum, where they start to decay at the same rate of larger ones. The location of this maximum as well as the intersection between $2\rho_2$ and $1 - \rho_1 - \theta$ tend to move to early times when $p$ increases, suggesting that enhanced deposition rate destroys many-site vacancies more efficiently. The functional forms that fit well $\rho_2$ and $1 - \rho_1 - \theta$ are the same as those for the initial jammed lattice case (cfr. Eqs. (6) and (7)).

V. RSAD OF $K$–MERS

We now address the problem of RSAD of $k$–mers on a square lattice, which is a generalization of the model described in Sec. II that corresponds to $k = 2$. Since for increasing $k$ the simulations of the dynamics becomes more and more time consuming and finite-size effects more important, we concentrate on low values of $k$, namely $k = 3, 4, 5$.

We have simulated the RSAD dynamics always starting with an initial jammed state and then switching on the diffusive part of the process. We checked that the jamming densities obtained as initial conditions after the pure RSA process were in agreement with the values found earlier by Bonnier et al. It turns out that there were no discrepancies with their results within the numerical precision. The jamming densities obtained are summarized in the Table V.

In Fig. 8 we compare the temporal evolution of the $k$–mers density for two values of $p$.

**TABLE VII.** Crossover time for one site vacancy domination.

| $k$ | $t_c$ |
|-----|-------|
| 3   | 80    |
| 4   | 400   |
| 5   | 1680  |
The major aspect to underline is the very sluggish dynamical behavior for increasing $k$, which is a common feature with the one dimensional case: the probability for $k$ vacancies to meet and annihilate decreases when $k$ gets larger. It can be noted that, as for dimers, a dynamical saturation at long times exists, although our data are less clear for $k = 5$, since the time at which this plateau sets in is an increasing function of $k$. Such a saturation is absent in $d = 1$, where the density follows a single power law for long times also for larger values of $k$. It appears therefore to be a feature typical of the two dimensional geometry and independent of the particle size, related to the complexity of the configurational space accessible to the system. A proof of that is that the curves in Fig. 8 can be fitted by the same functional form adopted for dimers (Eq. (4)) with the parameters of Table VI. It is seen that the $t^{-1/2}$ term is more important at larger $k$, in agreement with a slow down in the coverage process.

Arguments based on diffusive reaction (5) have shown in the past that a critical dimension $d_c = 2/(k-1)$ should exist above which fluctuations would be negligible and mean field behavior ($1 - \theta \sim t^{-1/k-1}$) would dominate. This would imply that for $d = 2$ the mean field law should work for $k \geq 3$; nevertheless we have found no evidence of that, at least for $k = 3$. In order to draw such a conclusion for larger $k$, longer runs would be required, since a change in the dynamical behavior could occur at very large times.

Finally we have compared the data for the density of one-site vacancies and of many-site vacancies, for $k = 3, 4, 5$ and $p = 0.1$ (Fig. 8). At variance with the case $k = 2$, many-site vacancies tend now to prevail for a long time and disappear more slowly for larger $k$; the time behavior of their density can be well approximated by a single power law:

$$1 - \rho_1(t) - \theta(t) \sim t^{-\beta_k},$$

with $\beta_3 = 0.94$, $\beta_4 = 0.75$ and $\beta_5 = 0.61$. Moreover, the crossover time $t_c$ after which one-site vacancies dominate increases with $k$, as shown in Table VII of $t_c$ values, where we note that for higher $k$ values diffusive regime sets in at later time.

VI. SUMMARY AND CONCLUSIONS

We studied irreversible deposition and diffusion of dimers and $k$-mers on a square lattice, using a driven algorithm which allowed to decrease the CPU time needed for Monte Carlo simulations. Both time coverage behavior and vacancy dynamics have been analyzed in detail as a function of initial conditions and deposition rate and probability. A wider variety of dynamical regimes with respect to the one dimensional case has been observed, reflecting the complexity of the configurational space accessible to dimers in $d = 2$ with respect to $d = 1$. In particular, the existence of dynamically frozen configurations, that only depend on the local environment, slows down the asymptotic dynamics and does not allow the system to reach the complete coverage. Different choices of deposition and diffusion rates and probabilities can lead to different final values of the asymptotic coverage. Simulation shows that the behavior of coverage $\theta$ strongly deviates from the mean field prediction. Deviations from mean field are less pronounced when the diffusion probability is higher. Furthermore $\theta$ is observed to depend on the initial conditions only in the early stage of the dynamics. Starting from the jammed state correlations are higher and the dynamics evolves more slowly. Vacancy dynamics is dominated not only by single-site vacancies, as in $d = 1$, but also by larger vacancies.

It was also shown that the dynamical behavior is severely slowed down with the increase of the length $k$ of the particles. We do not have evidences of a mean field behavior, in spite of the fact that it would be expected to hold for $k \geq 3$. We have also pointed out that the dimensionality plays an important role in reaction-diffusion models with the result of a rich dynamical behavior in the investigated quantities.

As an open possibility for the future, we think that in order to better understand the dynamics of these models it would be useful to vary the kind of relaxation mechanisms, investigating the possibility of occurrence of collective effects in $d = 2$ and $d = 3$.

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