Extended series expansions for random sequential adsorption

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We express the coverage (occupation fraction) $\theta$, in powers of time $t$ for four models of two-dimensional lattice random sequential adsorption (RSA) to very high orders by improving an algorithm developed by the present authors [J. Phys. A 29, L177 (1996)]. Each of these series is, to the best of our knowledge, the longest at the present. We analyze the series and deduce accurate estimates for the jamming coverage of the models.

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I. INTRODUCTION

A large variety of physical, chemical, biological, and ecological processes can be modeled by random sequential adsorption (RSA) on a lattice. In this irreversible process, particles are deposited randomly on a surface one after another. In the simplest form, the depositing particles are represented by non-overlapping hard core extended objects. These particles, once adsorbed on a surface, are assumed to be fixed in their spatial positions and exclude certain regions from further occupation. It is therefore interesting to study the coverage (fraction of occupied sites) of the system as time evolves, in particular the jamming coverage, a standstill beyond which no deposition is possible.

Exact solutions for RSA models are usually restricted to special cases only [1]. For those models which resist an exact theoretical treatment, different approximate methods have been developed [2]. Series expansion is one of the powerful analytical methods in RSA studies [4–10]. In this work we present an improved algorithm for series expansions based on hierarchical rate equations [11,12]. It is then applied to four lattice RSA models where we are able to obtain, to the best of our knowledge, the longest series known for each of model that we have studied.

II. MODELS

Four models are studied in this work. For each of these models, we start with an initially empty, infinite lattice. Particles are dropped randomly and sequentially at a rate of $k$ per lattice site per unit time, onto the lattice. Hereafter we set $k$ equal to unity without loss of generality. In the first model, monomers are deposited on a square lattice with nearest-neighbor (NN) exclusion, where an adsorbed monomer excludes its occupied sites and its four nearest-neighbors from further occupation [3]. In the second model, dimers of random orientations are deposited on a square lattice. In this model, a dimer can occupy a pair of nearest-neighbor sites only if both of the chosen sites are vacant [13]. The third and the fourth models are defined similarly as the first and the second models, respectively, except that depositions are performed on a hexagonal lattice [1].

III. MODIFIED ALGORITHM

We shall first summarize the original algorithm presented in Ref [3]. We are interested in expressing the coverage $\theta$ as a function of time $t$, which is related to the probability of finding a single vacant site (denoted by $^\circ$). More precisely, we write $\theta(t) = 1 - P(N, t)$. In general we are interested in expressing the probability of finding any particular configuration of interest, $C_0$, which we write it as $P_{C_0} = P(C_0)$, as a power series expansion of time $t$, with the expansion point $t = 0$. We write $P_{C_0}(t) = \sum_{n=0}^{\infty} P_{C_0}^{(n)} t^n / n!$, with $n$th derivative of $P_{C_0}$ given by $P_{C_0}^{(n)} = \frac{d^n P_{C_0}(t)}{dt^n} \bigg|_{t=0}$.

Let $G_i$ denote the set of new configurations generated in the calculation of the $i$th derivative of $P_{C_0}$, and $G_i^{(j)}$ the corresponding $j$th derivatives of the set of configurations. We observe that $G_0^{n-1}, G_0^{n-2}, \ldots, G_0^{n-1}$ [determined at the $(n-1)$th derivative], $G_0^{n-2}, G_0^{n-3}, \ldots, G_0^{n-2}$ [determined at the $(n-2)$th derivative], $\ldots, G_0^{0}$ are predetermined before calculating the $n$th derivative of $P_{C_0}$. In the calculation of $n$th derivative of $P_{C_0}$, we determine systematically $G_0^{0}, G_1^{n-1}, \ldots, G_{n-1}^{0}, G_n^{0}$, by recursive use of rate equations. This algorithm consumes the memory quickly due to storage of intermediate results. In this algorithm, each configuration is transformed into a unique canonical representation and stored as such in memory. If a configuration is already generated before, only a pointer reference to
the previous configuration is made. The checking of the existence of a particular configuration is done efficiently through the hashing technique \[13\].

An instructive way of looking at this algorithm is through the concept of tree traversal \[14\]. We make the first configuration \(C_0\), of which its highest order of derivative to be obtained is \(h\), as the root node of the tree. The depth at which the root node resides is set arbitrarily to 0, this corresponds to the index \(i = 0\) in the symbol \(G_i\) introduced above. We see that a complete breadth-first tree traversal will introduce at new set of leaves nodes of depth which is one more than the depth of leaves nodes in the previous breadth-first traversed tree. Therefore at the end of \(D\)th times of tree traversal we will introduce a set of new leaves nodes of depth \(D\). Since we have to store all new configurations or nodes that appear in the process, the breadth-first tree traversal is thus a very memory demanding algorithm. To curb the exponential memory growth, we stop the breadth-first traversal of tree when the leaves nodes have a depth \(D'\) where a new and different strategy is followed. Notice now that the problem of finding the \(h\)th order derivative of \(C_0\) amounts to pre-determining the first, second, \ldots, through \((h−D')\)th order derivatives for all the leaves nodes of depth \(D'\). If we can do that, then by another \((h−D')\) times of breadth-first traversal of the tree from the root node \(C_0\) again, we can calculate all derivatives of \(C_0\) up to order \(h\). Now the problem is to calculate the first through \((h−D')\)th order derivatives for each of the leaves nodes of depth \(D'\). This can be achieved by treating each of the leaves nodes of depth \(D'\) as a new root node in another breadth-first tree traversal subproblem. Each of these subproblems can be dealt with one at a time, which means that we can use the memory allocated for a subproblem for the next subproblem. It is through this way that the use of memory is kept at roughly a fixed amount. A drawback of this approach is that the derivatives for some configurations have to be calculated more than once, however there is an important observation that the calculations of all these subproblems can be parallelized.

As another effort to cut the growth of the memory requirement, we observe that the storing of the nodes is not really necessary for nodes which have an absolute depth very close to \(h\). This can be achieved by using a depth-first tree traversal. When we are considering the rate equation of a particular configuration \(C\), we recursively traverse down the tree whenever a configuration belongs to the right hand side of the rate equation for node \(C\), which we shall call it a child node of \(C\), appears. The merit of this depth-first approach is that we do not need to consider all symmetry operations that have to be applied to a configuration. These operations are required before any configuration can be kept in the memory. Although the depth-first strategy effectively saves the troubles of finding a canonical representation, the time to traverse a depth-first tree can be quite long if the height of the tree is large.

A detailed description of the implementation of the original algorithm can be found in Ref \[10\]. It can be modified readily to suit this new improved algorithm. Here we should mention the details of the task of performing the rate equation expansion. When the canonical representation for a new configuration is required, we make a list of 2-column vectors of entries which are the \(x\) and \(y\) coordinates of vacant sites specifying the configurations stored in a “working matrix”. Symmetry operations are applied to the new configuration through pre-multiplying the vectors in the list with appropriate transformation matrices. Criteria are set so that we can determine a canonical representation out of the list of new vectors.

Series expansions are parallelized using PVM version 3.3.11 \[14\] to utilize the hardware resources to the fullest. Calculations were performed on a 16-node cluster of Pentium Pro 200 with a bonded dual channel 100 BaseT Ethernet connection. All runs that we have done used less than 60 Megabytes of main memory, with the longest run took an elapsed time of 120 hours per machine in a dedicated computing environment. To compare the performance of this new algorithm and the original algorithm \[8\], we notice that we are able to obtain 4 more coefficients for the dimer RSA model on a square lattice using the former algorithm. The later algorithm requires 706 Megabytes of memory even at the 14th order derivative calculation. The results for all the models are displayed in Table \[I\] and Table \[II\] for square lattice RSA and hexagonal lattice RSA, respectively. We note that our 18th order coefficient in the expansion of powers of \(u = e^{−t}−1\), is 3560240252651011168 for monomer RSA with NN exclusion on a square lattice, which is slightly different from the value reported in Ref \[8\].

**IV. ANALYSIS OF SERIES**

The approach to the jamming state for lattice RSA is often exponential. This behavior enables us to use transformations of variables which reflect the actual approach. The transformations that we have employed are similar to that used in Refs \[9\], \[10\]. First we transform the coverage \(\theta(t)\) in term of \(y = 1−e^{−t}\). Another transformation \(z = (1−e^{-by})/b\) is performed. This transformation is suggested by the fact that the exact jamming approach of dimer RSA on a linear lattice corresponds to \(\theta(t) = z\) with \(b = 2\) \[8\]. For other RSA models, we set \(b\) to be a free variable so that when different orders of Padé approximants are applied to \(\theta(z)\), a crossing region between different orders of Padé approximants is to be located so as to give a good estimate of jamming coverage \(\theta_{\infty}\). As an example, Fig. \[\text{a}\] shows the crossing region of Padé approximants of orders \([11,10]\), \([10,11]\), \([12,9]\), \([9,12]\), \([13,8]\), \([8,13]\), \([14,7]\), \([7,14]\), \([10,10]\), \([11,9]\), \([9,11]\), \([12,8]\), \([8,12]\), \([10,9]\), \([9,10]\), \([11,8]\), and \([8,11]\) for the series of monomer RSA with NN exclusion on a square lattice, giving an estimate of
\( \theta_\infty = 0.3641323(1) \), where the last digit denotes the uncertainty. This estimate is in good agreement with the estimate of \( \theta_\infty = 0.3641330(5) \) by Baram and Fixman [8]. Analyzing the series using the square cactus as a reference model [3], we obtain \( \theta_\infty = 0.364132(1) \). All these estimates from the series analysis agree well with the simulation result of \( \theta_\infty = 0.36413(1) \) [15].

For the dimer RSA on a square lattice, we obtain \( \theta_\infty = 0.906823(2) \). This is to be compared with the simulation results of Oliveira et al. [6] of \( \theta_\infty = 0.90677(6) \) and of Wang and Pandey [16] of \( \theta_\infty = 0.906820(2) \). A somewhat biased estimate of \( \theta_\infty = 0.8788088(4) \) was obtained in Ref [9] as too small a range of \( b \) was used.

For monomer RSA on a hexagonal lattice, \( \theta_\infty = 0.37913944(1) \). This result is much more accurate than the simulation results of \( \theta_\infty = 0.38(1) \) and \( \theta_\infty = 0.379 \) reported in Refs [17] and [18], respectively. For the dimer RSA on hexagonal lattice, we obtain \( \theta_\infty = 0.8789329(1) \). This is in good agreement with the estimate \( \theta_\infty = 0.878809 \) in Ref [14].

V. CONCLUSIONS

In this work we have substantially extended the series for \( \theta(t) \) by at least 3 terms for RSA models defined on a square lattice. The series for the hexagonal lattice RSA are also of very high orders. We noted that with the improvement of the original algorithm, very long series and accurate estimates for \( \theta_\infty \) can be obtained. The generality of this computational method allows us to handle a variety of problems based on rate equations. We are currently applying this algorithm to provide more insights into the dynamics of the two-dimensional kinetic Ising model [14].

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| n | monomer RSA with NN exclusion | dimer RSA |
|---|-----------------------------|-----------|
| 1 | 1                          | -3        |
| 2 | 5                          | 15        |
| 3 | -37                        | -99       |
| 4 | 349                        | 807       |
| 5 | -3925                      | -7803     |
| 6 | 50845                      | 87039     |
| 7 | -742165                    | -1097139  |
| 8 | 12017245                   | 1538607   |
| 9 | -213321717                 | -25701699 |
| 10| 4113044061                 | 397893519 |
| 11| -85493084853               | -3989473277203116 |
| 12| 19038867855277             | 126863203272268 |
| 13| -45187885535477            | -3889473277203116 |
| 14| 1137973688508989           | 126863203272268 |
| 15| -3028952020394205          | -436543174155008620 |
| 16| 849248887429012733         | 15862169269805453164 |
| 17| -2500725970924817749       | -6058617368871081964076 |
| 18| 771322713104711008093      | 24259302255088935132428 |
| 19| -24860884250598911506045   | -24259302255088935132428 |
| 20| 1986803685767519515997     | 835568036857675195155997 |
| 21| -29226711255970839546587445 | -29226711255970839546587445 |

| n | monomer RSA with NN exclusion | dimer RSA |
|---|-----------------------------|-----------|
| 1 | -1                          | -3        |
| 2 | 4                          | 15        |
| 3 | -22                        | -99       |
| 4 | 154                        | 807       |
| 5 | -1306                      | -7803     |
| 6 | 12946                      | 87039     |
| 7 | -146026                    | -1097139  |
| 8 | 1837666                    | 1538607   |
| 9 | -25429018                  | -23701699 |
| 10| 382667218                  | 397893519 |
| 11| -6208467946                | -71934898755 |
| 12| 107847467914               | 1396766151303 |
| 13| -1994552336218             | -2893259695051 |
| 14| 39089479606162             | 63635537367831 |
| 15| -806280414455066           | -14801609728262739 |
| 16| 1759710793505354           | 362819105840203479 |
| 17| -4016844356616661234       | -934388660601611115 |
| 18| 959327364964882706         | 252096661713036722415 |
| 19| -239168310954693706954     | -7109230409530804525155 |
| 20| 6211745800773276251122    | 20908565591788638504551 |
| 21| -16776509906814548846842  | -6400562190944268227677947 |
| 22| 4703750163644209363257538  | 203577552403910264228987775 |
| 23| -136703798845635451539146 | -136703798845635451539146 |
| 24| 411246853135639607791658706 | 411246853135639607791658706 |
FIG. 1. Padé approximant estimates for the jamming coverage $\theta_\infty$ as a function of the transformation parameter $b$, for the monomer RSA with NN exclusion on a square lattice.