Use of Model Solutions in Random Sequential Adsorption on a Lattice

Y. Fan and J. K. Percus[(a)]

Courant Institute of Mathematical Sciences, New York University, 251 Mercer Street, New York, New York 10012

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We consider random sequential adsorption on a lattice. We use analytical results on the Bethe lattice and cactus as references to develop systematic perturbationlike expansions which are very rapidly convergent. The latter produces the jamming density of a square lattice with an accuracy within 10^{-5}. This expansion is based on both physical and mathematical considerations and is not restricted to random sequential adsorption.

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(1) Introduction.— A number of processes in the physical, chemical, and biological sciences can be modeled by random sequential adsorption (RSA) on a lattice. In the prototypical version of this model, hard objects arriving randomly at sites of a lattice are irreversibly adsorbed unless they are in the exclusion zone of previously adsorbed objects, in which case they leave and try again. In spite of considerable effort, analytical results are sparse and largely confined to one-dimensional systems [1-3] or lattices with tree properties [4,5]. Various hierarchies have been developed for both continuous [6] and discrete [4,7] systems, but they seem more useful for uncontrolled approximations than as systematic evaluation procedures.

Two rather different approaches to systematic evaluation have been studied. The first, which has been carried out extensively, involves the selection of a suitable expansion parameter, typically the time, and the corresponding development of a power-series expansion. Primitive expansions tend to converge very poorly, and so are used, after modest convergence-accelerating transformations, to construct Padé approximants. Judged by comparison with numerical simulations, the results achieved this way in Refs. [8] and [9] attest to the reliability of the methods. The second method has been only briefly mentioned, but is at the heart of this paper. It takes advantage of increasingly sophisticated exactly solved models to incorporate series information, and thus relies also upon the physics of the situation being studied, rather than solely upon general mathematical convergence procedures. In summary, we first review very briefly the expansions derived by Evans [10] and Dickman, Wang, and Jensen [9], and then quote our previous results on Bethe lattices. We use these results in two different ways to develop the particle coverage on these lattices and conclude by extending our reference systems for triangular and square lattices to the corresponding cacti, producing very rapidly convergent estimates.

(2) Brief review.— According to Evans [10] and Dickman, Wang, and Jensen [9], if one considers a RSA process on an initially empty translation-invariant lattice with nearest-neighbor exclusion, the average density (coverage) in suitably scaled time has the expansion

$$\rho(t)=\sum_{k=1}^{\infty}\frac{(-1)^{k-1}}{k!}S(k-1)t^k\,, \tag{2.1}$$

where $S(k)$ is the number of paths, starting at 0, with k further sites, each a repeat or a nearest neighbor of a previous site. By rearranging terms, (2.1) can be transformed into [5]

$$\rho(t)=N(1-e^{-t})\,,\qquad N(x)=\sum_{k=1}^{\infty}\frac{(-1)^{k-1}}{k!}N_{k-1}x^k\,, \tag{2.2}$$

where N_k counts the subset of walks of $S(k)$ in which no repeats of previous sites are permitted.

It was shown that (2.2) is convergent for a one-dimensional lattice, for two-row square ladders, and also for a Bethe lattice of coordination number q providing that $|x|<1/(q-2)$ [5]. In the latter case, the analytic result is [5]

$$N(x)=\tfrac{1}{2}\{1-[1+(q-2)x]^{-2/(q-2)}\}\,. \tag{2.3}$$

For a general D-dimensional cubic lattice, one can argue that (2.2) does indeed converge for $x=1$, but very slowly, even for $D=2$. However, the Bethe-lattice results should be reasonable estimates if the local structure of the lattice is "close" enough to that of the Bethe lattice. The justification of close is not entirely clear. Roughly, we can consider how many steps L one needs to form a loop, with $L=\infty$ for a Bethe lattice. For triangular, square, and honeycomb lattices, one has $L=3$, 4, and 6, respectively. Comparisons of asymptotic coverage (by computer simulation) between these three and the corresponding Bethe lattices are provided in Table I. Although the direction of the deviation from Bethe to regular lattices is not uniform, the size of the deviation shows that honeycomb, with $q=3$, $L=6$, is smallest, in agreement with the above argument.

(3) Bethe-lattice reference.— There are many options

TABLE I. Asymptotic coverage, $\rho(\infty)$.

Lattice type	Coordination number q	Loop size L	Regular lattice	Bethe lattice
Triangular	6	3	0.231 [a]	0.276
Square	4	4	0.364 [a]	0.333
Honeycomb	3	6	0.380 [b]	0.375

[a]Reference [11].
[b]Reference [12].

TABLE II. Series expansion for $\rho(\infty)$ on a square lattice.

k	$N_k/(k+1)!$	a_k	b_k	c_k	ρ_k^a	ρ_k^b	ρ_k^c
0	1	2		1	0.3333		0.569334
1	2	0	2	−1	0.3333	0.3333	0.245193
2	4	0	0	6.66667(−1)	0.3333	0.3333	0.368222
3	7.333	1.3333	−1.3333	1.66667(−1)	0.3846	0.4010	0.385734
4	12.53	−1.6	0.6667	−0.4	0.3171	0.3677	0.361806
5	20.22	1.1556	0.6667	−4.44444(−2)	0.3714	0.3508	0.360293
6	31.10	0.6984	−0.8667	2.09524(−1)	0.3910	0.3696	0.364355
7	45.95	−3.0524	−0.0857	2.46032(−2)	0.1743	0.3718	0.364627
8	65.54	4.5346	0.6877	−1.09436(−1)	0.4176	0.3590	0.363939
9	90.68	−3.3194	−0.1947	−7.23986(−3)	0.3182	0.3617	0.363913
10	122.1	−1.2978	−0.5273	9.25261(−2)	0.1557	0.3700	0.364102
11	160.5	7.8651	0.3966	5.62378(−2)	0.4463	0.3640	0.364213
12	206.5	−12.4173	0.3294	−1.32377(−2)	0.6613	0.3604	0.364205
13	260.3	10.0714	−0.5410	−2.62243(−2)	0.4279	0.3662	0.364195

for accelerating convergence of the series (2.2). Perhaps the most directly motivated from a reference-model point of view is as follows. We have observed that although the direct power expansion of (2.3) does not converge at $x=1$ ($t=\infty$) it can be rewritten as

$$[1-2N(x)]^{-(q-2)/2}=1+(q-2)x\,, \quad (3.1)$$

a series which truncates at the second term. This suggests that if we expand

$$[1-2N(x)]^{-(q-2)/2}=1+\sum_{k=1}^{\infty} a_{k-1}x^k \quad (3.2)$$

for a regular lattice of coordination number q, the higher coefficients in the power series should decrease more rapidly, so that fewer are required for a given accuracy. We have carried out (3.2) for the square lattice, obtaining the a_k for $k\le 13$ from the N_k ($k\le 13$) [13]. Table II lists the $N_k/(k+1)!$, the corresponding a_k, and the cumulatives of the resulting expansion at $x=1$. It is seen that the size of the coefficients has been greatly compressed; the fourteenth term in (3.2) is only a few percent of that in (2.2). $\rho(\infty)$ in the former is, at this stage, no place near the correct result, while the latter remains within 0.2 of it. The huge reduction of the coefficients seems to indicate that some sort of diagram reduction is taking place, but the apparently random sign of a_k shows that expressing this incisively will not be an easy task.

There is a nominally minor change one can make in the above procedure which further improves convergence. We notice that for the Bethe lattice,

$$N'(x)^{-(q-2)/q}=1+(q-2)x\,, \quad (3.3)$$

and correspondingly write

$$N'(x)^{-(q-2)/q}=1+\sum_{k=1}^{\infty} b_k x^k\,. \quad (3.4)$$

If the b_k decrease in magnitude about as rapidly as the a_k, then the integration required to produce $N(x)$ from $N'(x)$ will certainly smooth out the oscillations and accelerate convergence. According to Table II, this is certainly the case. It is clear from these two examples that application of Occam's razor is effective: The solution of the reference system leaves us very little choice as to how to organize the series calculation. With the confidence thus engendered, we can now proceed to even better reference systems.

(4) Cactus reference.—The more closely the reference lattices mimic the local structure of the lattice under consideration, the more accurate its prediction should be, and the more effective the expansions directed by its structure. Exact analysis of RSA on lattices of increasing connectivity is not trivial. But we may start with an instance in which it is surprisingly feasible, that of a triangular cactus, a cactus in which three triangles meet at each vertex. For a cactus, the meaning of "close" is even more nebulous than for a Bethe lattice. We simply look at the deviation of the string of N_k's from those of the real lattice measured by computing the ratio of the two. These deviations increase with k in both cases, but considerably more slowly for the cactus [see Table III, where γ_β (γ_c) is the ratio of the N_k's between Bethe (cactus) and regular lattices].

It is an easy exercise to show that for a triangular cactus, $N_k/N_{k-1}=3(k+1)$, so that [5]

$$N(x)=x/(1+3x)\,. \quad (4.1)$$

On the one hand, this yields $\rho(\infty)=0.25$, which according to Table I is a substantial improvement over the Bethe-lattice result. On the other hand, adopting the improved strategy of Sec. 3, we observe that $N'(x)=(1+3x)^{-2}$, and correspondingly choose

$$[N'(x)]^{-1/2}=1+\sum_{k=1}^{\infty} d_k x^k \quad (4.2)$$

TABLE III. The comparison of N_k for a triangular lattice.

k	Regular	Bethe	Cactus	γ_β	γ_c
0	1	1	1	1	1
1	6	6	6	1	1
2	48	60	54	1.25	1.125
3	468	840	648	1.7949	1.3846
4	5328	15120	9720	2.8378	1.8243

TABLE IV. Series expansion for $\rho(\infty)$ on a triangular lattice.

k	$N_k/(k+1)!$	d_k	ρ_k
1	3	3	0.25
2	8	1.5	0.2191
3	19.5	−1.5	0.2349
4	44.4	0.375	0.2316

as our model expansion. Doing so, we find (Table IV) a greatly accelerated convergence, leading rapidly to the computer-simulation result.

In a square cactus, two squares meet at each vertex (Fig. 1). The ratio of the N_k's is calculated and shown in Table V with, again, a more slowly increasing deviation for the cactus, and we might anticipate that use of such a model as reference would be extremely effective at accelerating convergence. The square cactus can also be solved exactly; the solution, which requires a certain amount of effort, is carried out in the Appendix and yields

$$N(x)=\tfrac{1}{2}-(\tfrac{1}{3}y+\tfrac{1}{6}y^4)\,,\qquad x=3\int_y^1 dy'/(2y'^3+1)\,. \tag{4.3}$$

A first observation is that here $\rho(\infty)=N(1)=0.3507$, a substantial improvement over the Bethe-lattice result. A second observation is that since N is already a truncated power series in y, Occam's razor dictates that the regular square lattice be expanded in y as well, instead of x. There is just one snag: The power series for $x(y)$ does not converge in the region $(2^{-1/3},1)$ so that it makes more sense to expand in $z=1-y$. In other words, we write

$$N[x(z)]=\sum_{k=1}^{\infty}c_{k-1}z^k\,,\qquad x=3\int_0^z dz'/[1+2(1-z')^3]\,, \tag{4.4}$$

and fit the c_k from the known coefficients N_k. Convergence to $\rho(\infty)$ (see last column, Table II) is extremely rapid, and suggests that $\rho(\infty)=0.364\,20(\pm1)$, a small correction to the simulation result $[0.364\,13(\pm1)]$ [11].

Encouraged by the square cactus as a reference system for the square lattice, we also carried out in a very similar fashion the cubic cactus model of a cubic lattice. The resulting $\rho(\infty)=0.305\,69$ is indeed already very close to the cubic-lattice simulation result, $\rho(\infty)=0.304(\pm1)$ [9]. It seems to us that the simulation results in all three cases may be a little bit lower than they should be.

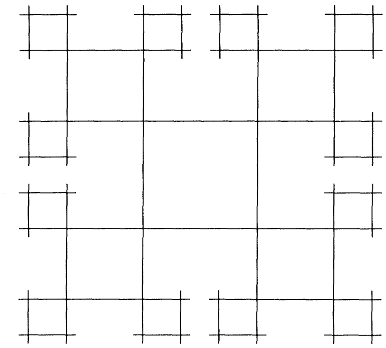

FIG. 1. Part of a square cactus. The different size of squares serves plotting convenience.

Remarks.— We have introduced a technique for the solution of RSA models, which we believe is of much more general utility. It organizes perturbationlike calculations according to both physical and mathematical considerations. The general idea is to first seek a variable in terms of which some function of the solution of a solvable reference is represented by a low-order polynomial. A systematic expansion of the same function for the "perturbed" system in terms of the same variable would then be expected to converge very rapidly, depending upon how close the solvable system is to the real one. Of course, the prescription is not unique and becomes even less so if the physics depends upon too many relevant variables. This method is not restricted to RSA. We have recently extended it to the equilibrium Ising model as well and will report on this in a future publication.

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Appendix: RSA on a square cactus.— We let n_i be the number of squares with i sites occupied, k the total number of occupied sites (with nearest-neighbor occupancy

TABLE V. The comparison of N_k for a square lattice.

k	Regular	Bethe	Cactus	γ_B	γ_c
0	1	1	1	1	1
1	4	4	4	1	1
2	24	24	24	1	1
3	176	192	184	1.0909	1.0455
4	1504	1920	1712	1.2766	1.1383
5	14560	23040	18688	1.5824	1.2835
6	156768	322560	233888	2.0576	1.4919

dynamics). One sees by induction that

$$\sum_{i=1}^{4} n_i = k+1, \quad \sum_{i=1}^{4} i n_i = 2k \tag{A1}$$

(so that $n_1 - 2 = n_3 + 2n_4$ is maintained). Now define

$$N_k(n_1,n_2,n_3) = N(n_1,n_2,n_3,n_4) \tag{A2}$$

as the number of "walks" specified by $\{n_i\}$. It is easily seen by induction that [14]

$$N(n_1,n_2,n_3,n_4) = 2n_1 N(n_1,n_2-1,n_3,n_4) + 2(n_2+1)N(n_1-1,n_2+1,n_3-1,n_4) + (n_3+1)N(n_1-1,n_2,n_3+1,n_4-1) + \delta_{n_1,2}\delta_{n_2,0}\delta_{n_3,0}\delta_{n_4,0}\,. \tag{A3}$$

Introducing the generating function

$$N(x_1,x_2,x_3,x) = \sum_{n_1=2}^{\infty} \sum_{n_{2,3,4}=0}^{\infty} \left\{ \frac{(-1)^{k+1}}{k!} N_k(n_1,n_2,n_3) x_1^{n_1} x_2^{n_2} x_3^{n_3} x^k \right\}, \tag{A4}$$

(A3) implies that

$$N(x_1,x_2,x_3,x) = x x_1^2 - 2\sum \frac{n_1 x_2 x}{k+1}\{\cdots\} - 2\sum \frac{1}{k+1}\frac{n_2 x_1 x_3 x}{x_2}\{\cdots\} - \sum \frac{1}{k+1}\frac{n_3 x_1 x}{x_3}\{\cdots\}, \tag{A5}$$

where the $\{\cdots\}$ are the same as that in (A4). It follows that

$$\left[\frac{\partial}{\partial x} + 2x_1 x_2 \frac{\partial}{\partial x_1} + 2x_1 x_3 \frac{\partial}{\partial x_2} + x_1 \frac{\partial}{\partial x_3} \right] N \equiv DN = x_1^2, \quad N(x_1,x_2,x_3,0) = 0\,. \tag{A6}$$

Now $Dx = 1$, and $Dc_j = 0$ for $j = 1,2,3$, where

$$c_1 = x_2 - x_3^2, \quad c_2 = x_1 - 2c_1 x_3 - \tfrac{2}{3}x_3^3, \quad c_3 = x + \int_{x_3}^{1} dy/(c_2 + 2c_1 y + \tfrac{2}{3}y^3), \tag{A7}$$

so that if

$$N(x_1,x_2,x_3,x) = \bar{N}(c_1,c_2,c_3,x) \text{ with } x_3 = x_3(c_1,c_2,c_3,x), \tag{A8}$$

then

$$d\bar{N}/dx = x_1^2 = (c_2 + 2c_1 x_3 + \tfrac{2}{3}x_3^3)^2 = (c_2 + 2c_1 x_3 + \tfrac{2}{3}x_3^3)\,dx_3/dx\,.$$

Hence

$$\bar{N} = (c_2 x_3 + c_1 x_3^2 + \tfrac{1}{6}x_3^4) - (c_2 \bar{x}_3 + c_1 \bar{x}_3^2 + \tfrac{1}{6}\bar{x}_3^4), \text{ where } \int_{\bar{x}_3}^{1} dy/(c_2 + 2c_1 y + \tfrac{2}{3}y^3) = c_3\,. \tag{A9}$$

At $x_1 = x_2 = x_3 = 1$, we have $c_1 = 0$, $c_2 = \frac{1}{3}$, $c_3 = x$, and so $N(x) = N(1,1,1,x) = \bar{N}(0,\frac{1}{3},x,x) = \frac{1}{3}(1-\bar{x}_3) + \frac{1}{6}(1-\bar{x}_3^4)$, i.e.,

$$N(x) = \tfrac{1}{2} - \tfrac{1}{3}\bar{x}_3 - \tfrac{1}{6}\bar{x}_3^4, \text{ where } \int_{\bar{x}_3}^{1} dy/(\tfrac{1}{3} + \tfrac{2}{3}y^3) = x\,. \tag{A10}$$

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[a]Also at Department of Physics, New York University, New York, NY 10003.

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