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1992 J. Phys. A: Math. Gen. 25 L493

(http://iopscience.iop.org/0305-4470/25/8/019)

J. Phys. A: Math. Gen. 25 (1992) L493–L498. Printed in the UK

LETTER TO THE EDITOR

Random sequential adsorption on a quasi-one-dimensional lattice: an exact solution

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Received 15 October 1991

Abstract. The dynamics of a random sequential absorption process on a quasi-one-dimensional lattice is solved exactly. It is shown that the filling process exhibits some features typical to two-dimensional systems. In particular the jamming density is $\rho_r = \frac{1}{2}(1-(1/2e)) = 0.408\,030\,1397\ldots$, considerably lower than the one-dimensional jamming density

Random sequential adsorption (RSA) is an irreversible random packing process. Particles are absorbed randomly, one at a time, into a d-dimensional space, obeying the following conditions: (a) particles do not overlap, (b) absorbed particles are permanently fixed in their spatial positions. Thus at each step a new particle is either rejected from the volume, or it is added at random to an accessible point in the diminished volume formed by all previously adsorbed particles. The filling process ends at a jammed state, when no more particles can be absorbed. Clearly the average jamming density ρ_r is smaller than the corresponding density of closest packing ρ_0.

A variety of physical, chemical, biological and ecological irreversible processes are realized by RSA models [1–6]. Furthermore, since the RSA phase is a non-equilibrium disordered phase for all values of ρ for which it exists, it has been suggested as a phenomenological model for glasses and supercooled liquids [7].

Exact RSA results are available only in one-dimensional systems [1, 2, 8]. For higher-dimensional systems most of the available information stems from numerical machine calculations [9–12]. Recently formal series expansions for the dynamics of RSA models have been derived [13–16]. The graphical expansion of [13], used here, represents the time evolution of multi-particle correlation functions in terms of diagrams of lattice animals. The diagramatic expansion is valid for all dimensions d. For the two limiting dimensionalities $d=1$ and $d=\infty$ the expansions simplify considerably. The analytical $d=1$ results are easily recovered, while at large d the expansion reduces to a Cayley tree type approximation [17], predicting for the one particle correlation function, i.e. the coverage density:

$$\rho(u) = \ln[1+a_2(1-u)]/a_2 \tag{1}$$

where $u=\exp(-t)$ is the natural dynamical variable of [13] and a_2 is the second graphical coefficient (see below), related to the corresponding equilibrium second virial coefficient B_2 by: $a_2 = 2B_2 - 1$. In particular, one obtains for the jamming density:

$$\rho_r = \ln(1+a_2)/a_2. \tag{1a}$$

In this letter we utilize the graphical method [13] to obtain an exact solution for RSA on a quasi-one-dimensional lattice, consisting of a strip of two infinite rows. Some of the properties of RSA on a $d=2$ cubic lattice are already manifested in the time evolution of the process on the strip. Therefore it is worthwhile to study this case.

Consider particles absorbing at random on a $2*\infty$ strip of a square lattice, with mutual nearest neighbours (NN) exclusion and periodic boundary conditions. The time evolution of the filling process is formulated in terms of a time-dependent distribution function $P(s, t)$, $s=\{s_i\}$, s_i are two state occupation variables which are defined as 1 for empty sites and 0 for occupied ones. P satisfies a master equation

$$\mathrm{d}P(s,t)/\mathrm{d}t=\sum_{i=1}^{N}[W(1-s_i)P(s_1,\ldots,1-s_i,\ldots,s_N,t)-W(s_i)P(s,t)] \quad (2)$$

whose initial conditions, for an initially empty lattice, are $s_i(t=0)=1$ for all i. More general initial conditions may be implemented as well. The transition rate W_i defined by:

$$W_i=s_i\prod_j s_j \quad (3)$$

represents the mutual repulsion of the adsorbed particles. It vanishes if the site i or one of its NN are occupied (j runs over the NN of i). Macroscopic correlation functions are obtained by appropriate tracing of P; in particular the coverage density $\rho(t)$ is related to the one particle probability distribution by:

$$\rho(t)=1-\langle s_i\rangle \quad (4)$$

where the brackets $\langle\,\rangle$ denote an ensamble average. Using (2) and (3) one obtains

$$\mathrm{d}\langle s_i\rangle/\mathrm{d}t=-\langle W_i\rangle. \quad (5)$$

Similarly the nth time derivative of $\langle s_i\rangle$ is given by a sum of all possible combinations of n-point connected lattice animals. Since at $t=0$ $(u=1)$ $s_i=1$, all the resulting correlators are equal to unity at this limit, which enables one to construct expansions for macroscopic observables in powers of $(1-u)$ [13].

The density is given by:

$$\rho(t)=\sum_{n=1}^{\infty}(-1)^{(n+1)}b_n t^n/n!=\sum_{n=1}^{\infty}(-1)^{(n+1)}a_n(1-u)^n/n! \quad (6a)$$

where the relation between the two sets of coefficients is given by:

$$b_n=\sum_{m=1}^{n}a_m S_{n,m}. \quad (6b)$$

$S_{n,m}$ is a Stirling number of the second kind [18]. The coefficients b_n are identical to those of Dickman *et al* [15]. Since the coefficients a_n are equal to the number of connected lattice animals containing n points, they are equal to positive integers, and their calculation reduces to an enumeration problem.

The simplicity of the one-dimensional models is reflected in a set of a few generating configurations for the connected animals, since the number of boundary points at each step is independent of n. In particular for the NN case there is a single generating configuration, resulting in the simple recursion relation: $a_n=2a_{n-1}$. Similarly to the

$d=1$ case, the time evolution on the strip is characterized by a finite set of generating configurations:

$$A_n=\prod_{i=1}^{n} s_{1,i}s_{2,i} \tag{7a}$$

$$B_n=A_n s_{2,0}s_{2,n+1} \tag{7b}$$

$$C_n=A_n s_{2,n+1} \tag{7c}$$

$$D_n=A_n s_{1,0}s_{2,n+1} \tag{7d}$$

whose time evolution is given by the closed set of coupled differential equations:

$$\mathrm{d}A_n/\mathrm{d}u=4C_n \tag{8a}$$

$$\mathrm{d}B_n/\mathrm{d}u=2C_{n+1}+2uB_{n+1} \tag{8b}$$

$$\mathrm{d}C_n/\mathrm{d}u=A_{n+1}+B_n+D_n+uC_{n+1} \tag{8c}$$

$$\mathrm{d}D_n/\mathrm{d}u=2C_{n+1}+2uD_{n+1}. \tag{8d}$$

It is easy to see that, as in the $d=1$ case, the evolution of the configurations A–D is n independent. Furthermore the evolution of B and D is identical, hence (8) reduces to:

$$\mathrm{d}A/\mathrm{d}u=4C \tag{9a}$$

$$\mathrm{d}B/\mathrm{d}u=2C+2uB \tag{9b}$$

$$\mathrm{d}C/\mathrm{d}u=A+2B+uC \tag{9c}$$

with the initial conditions $A(u=1)=B(1)=C(1)=1$. It can be easily checked that the solution of (9) has the form:

$$A(u)=u^2\,\mathrm{e}^{(u^2-1)} \tag{10a}$$

$$B(u)=\tfrac{1}{4}(u^4+2u^2+1)\,\mathrm{e}^{(u^2-1)} \tag{10b}$$

$$C(u)=\tfrac{1}{2}(u^3+u)\,\mathrm{e}^{(u^2-1)}. \tag{10c}$$

The coverage density ρ is given by the solution of:

$$-\frac{\mathrm{d}^2\rho(u)}{\mathrm{d}u^2}=A(u)+2B(u) \tag{11}$$

With the initial conditions: $\rho(1)=0$ and $-(\mathrm{d}\rho/\mathrm{d}u)(1)=1$. Integrating (11) one obtains:

$$\rho(u)=1-u-\int_u^1 \mathrm{d}u'\int_{u'}^1 \mathrm{d}u''(A(u'')+2B(u'')). \tag{12}$$

The jamming density $\rho_r=\rho(u=0)=\rho(t=\infty)$ can be easily evaluated:

$$\rho_r=\frac{1}{2}\left(1-\frac{1}{2e}\right)=0.408\,030\,139\ldots. \tag{13}$$

This result has been recently obtained by Fan and Percus [19] using a different method. The value of ρ_r is substantially lower than the corresponding one dimensional jamming density, which is $\rho_r(d=1)=(1-\mathrm{e}^{-2})/2=0.432\,332\ldots$. It is close to the average of the densities of the $d=1$ and the square lattice models. The jamming density of the latter is estimated to be $\rho_r(d=2)=0.364\,13\ (1)$ [13, 15].

The following four-term recursion relation results from (10*b*)

$$B_{n+1}=3B_n+(3n+5)B_{n-1}-4(n+1)B_{n-2}-2(n+3)(n-2)B_{n-3}. \tag{14}$$

The coefficients a_n of (6*a*) are related to B_n by:

$$a_n=4[B_{n-2}-B_{n-3}-(n-3)B_{n-4}]. \tag{15}$$

It is easily seen that both B_n and a_n increase asymptotically like $\sqrt{n!}$. This behaviour is in sharp contrast with the characteristic exponential dependence of the $d=1$ case. It is identical to the asymptotic growth of lattice animals on the $d=2$ lattice [20]. Thus the $2*\infty$ solution may provide useful information regarding the process on the $d=2$ lattice.

It is worthwhile to note that although the lattice is a low-dimensional one, the large-d approximation for the coverage density on the strip $\rho_{\text{ap}}=\ln[1+3(1-u)]/3$ is quite a good approximation for $\rho(u)$ especially in the short time regime.

In contrast to continuum RSA models, where the deviation of the density from its limiting value ρ_r decays asymptotically like a power law [21, 22], in lattice systems the decay is exponential. Indeed, the long time behaviour of the coverage density is:

$$\rho(u)=\rho_r-c_1u+O[u^2] \tag{16}$$

where

$$c_1=\frac{1}{8}\left(1+\frac{1}{e}\int_0^1 e^{u^2}\,du\right)=0.192\,259\,938\,36\ldots.$$

In figure 1 the exact time evolution is compared to the long time approximation (16), and to the large-d approximation (1). Both approximations bound the exact values from above for all t. For $u<0.4$, i.e. $t>0.916$, the errors in the long time approximate values are less than 5%, while the errors in the large-d approximation do not exceed the same value for $u>0.65$, i.e. $t<0.431$.

Multi-particle correlation functions are directly related to ρ. It is easy to see that the two particle occupation function for NN sites is given by:

$$\langle s_i(t)s_j(t)\rangle=1-2\rho(t) \tag{16}$$

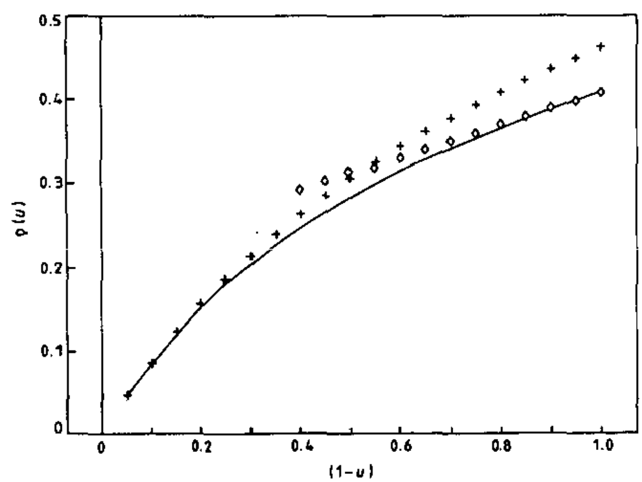

Figure 1. The exact time evolution of the coverage density $\rho(u)$ (continuous line) is shown as a function of $(1-u)$. The long time approximation (equation (14) (□)), and the large-d approximation (equation (1)(+)) bound the exact values from above.

At the jamming limit the local pair distribution has one of the two possible configurations

$$\begin{matrix} 1 & 0 \\ 0 & 1 \end{matrix} \quad \text{or} \quad \begin{matrix} 1 & 1 & 0 \\ 0 & 1 & 1 \end{matrix} \qquad \text{(recall that 0 designates an occupied site).}$$

The weight of the second configuration is given by (16), and the corresponding pair distribution function is:

$$g(2,1)=(1-2\rho_r)/2\rho_r=0.225\,39. \tag{17a}$$

The complementary function is:

$$g(1,1)=0.774\,61. \tag{17b}$$

In general $g(k,k')$ is given as a sum of products of $g(1,1)$ and $g(2,1)$

$$g(k,k')=\sum_{l,m}\binom{l+m}{l}g(1,1)^l g(2,1)^m \tag{18}$$

where $2m+l=k$, and $k'=l+m$ modulo 2. The binomial coefficient is the number of different ways to perform a (k,k') shift in l steps of type $(1,1)$ and m steps of type $(2,1)$. Although $g(1,1)$ is substantially higher than $g(2,1)$, the correlation loss is very fast, and $g(k,k')$ converges to its asymptotic value ρ_r for relatively small k values. As an example, for $k=4$ the occupation difference of the two sublattices relative to their average value is

$$2[g(4,0)-g(4,1)]/[g(4,0)+g(4,1)]=0.012$$

while the relative deviation from ρ_r is of the order of 0.5%.

Finally it is interesting to compare the RSA model with a similar equilibrium problem. Consider the Hamiltonian

$$H=\beta\left[\sum_{i=1}^{N}W_i+\sum_{i,j}(1-s_i)(1-s_j)\right]. \tag{19}$$

In the infinite strong coupling limit the configurations of H are identical to those of the jammed RSA process. Since the second term in H excludes NN occupation, the first term that plays the role of a short ranged pseudo attraction suppresses configurations in which a site and all its NN are empty. Let c_N and n_N denote the number of allowed configurations and their average occupation number respectively for a $2*N$ strip. Then using the following recursion relations

$$c_N=c_{N-2}+2c_{N-3}+c_{N-4}$$

and

$$n_N=[n_{N-2}c_{N-2}+2n_{N-3}c_{N-3}+n_{N-4}c_{N-4}]/c_N+2$$

one obtains for the average density of the infinite H system $\rho_H=0.361\,803,\ldots,$ which is smaller than ρ_r. A similar relation has been found for the one-dimensional case [13]. We conjecture that this result is general, and the jamming density is always higher than the average density of a corresponding Hamiltonian system. The discrepancy results from the fact that all the configurations have the same statistical weight in the equilibrium case, while in the dynamical RSA process the statistical weight of a configuration is proportional to its occupation number.

We thank the referee and Professor J W Evans for bringing [19] to our attention.

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