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Inhomogeneous Random Sequential Adsorption on a Lattice

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We study idealized random sequential adsorption on a lattice, with adsorption probabilities inhomogeneous both in space and in time, and including the possibility of cooperativity. Attention is directed to the mean occupancy of a given site as a function of time, which is represented by a weighted random walk on the lattice. In the special case of nearest neighbor exclusion, the walk is transformed to one in which only neighbors of occupied sites can be occupied, but with a renormalized probability. Reduction theorems are presented, with which the general case of a tree lattice is completely solved in inverse form.

KEY WORDS: Random sequential adsorption; irreversible kinetics; nonuniform lattice; simply connected network; inverse response.

1. BASIC FORMULATION

In this communication, we consider the process of random sequential adsorption or addition (RSA) on a lattice in a version that emphasizes inhomogeneity in space and time, and show how exact solutions for networks of low connectivity may be obtained. RSA models a large number of physical and chemical processes, which have been discussed extensively in the literature (see, e.g., ref. 1).

The idealized form that we study is characterized by a particle flux $f_x(t)$ into site x at time t, with a sticking probability per incoming particle of

$$w_x = \exp\left(-\sum_z \phi_{xz} n_z\right) \tag{1.1}$$

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Here n_z is the occupation, restricted to 0 or 1, of site z, and we will choose $\phi_{xx} = \infty$. The possibility of cooperative dependence on the environment of site x dates back to Hoffman.[2] Now, in time interval dt, the configuration of site x remains the same if $n_x = 1$, remains the same with probability $1 - w_x f_x(t)\,dt$ if $n_x = 0$, and transforms to $n_x = 1$ with probability $w_x f_x(t)\,dt$ if $n_x = 0$. We adopt a representation in which the site occupations are given by

$$|0\rangle = \begin{pmatrix} 1 \\ 0 \end{pmatrix}, \qquad |1\rangle = \begin{pmatrix} 0 \\ 1 \end{pmatrix} \tag{1.2}$$

In terms of the occupation operator, "hole" occupation operator, and particle creation operator at x:

$$n_x = \begin{pmatrix} 0 & 0 \\ 0 & 1 \end{pmatrix}, \qquad \bar{n}_x = \begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix}, \qquad a_x^+ = \begin{pmatrix} 0 & 0 \\ 1 & 0 \end{pmatrix} \tag{1.3}$$

The site x transformation operator over interval dt is then given by

$$\begin{aligned} \mathscr{T}_x(t, dt) &= \begin{pmatrix} 0 & 0 \\ 0 & 1 \end{pmatrix} + f_x(t) \begin{pmatrix} 0 & 0 \\ 1 & 0 \end{pmatrix} w_x\,dt + \begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix} [1 - f_x(t) w_x\,dt] \\ &= 1 + \mathscr{T}_x(t)\,dt \end{aligned} \tag{1.4}$$

where[3,4]

$$\mathscr{T}_x(t) = f_x(t)(a_x^+ - \bar{n}_x) w_x \tag{1.5}$$

Denoting the probability state vector of the full lattice by $|P(t)\rangle$, we conclude that $|P(t+dt)\rangle = |P(t)\rangle + \sum_x \mathscr{T}_x\,|P(t)\rangle\,dt$, and hence that

$$\frac{\partial\,|P(t)\rangle}{\partial t} = \sum_x \mathscr{T}_x(t)\,|P(t)\rangle \tag{1.6}$$

2. RANDOM WALK REPRESENTATION

Suppose that initially, say at $t = -\infty$, the lattice is empty: $|P(-\infty)\rangle = |0\rangle$, where $|0\rangle$ here is the unoccupied state of the full lattice. Then (1.6) can be solved in standard time-ordered exponential form

$$\begin{aligned} |P(t)\rangle &= T\left[\exp \sum_x \int_{-\infty}^{t} \mathscr{T}_x(t')\,dt'\right] |0\rangle \\ &= \sum_{N=0}^{\infty} \int_{t \geqslant t_1 \geqslant \cdots \geqslant t_N} \cdots \int \prod_{j=1}^{N} \sum_x \mathscr{T}_x(t_j)\,|0\rangle\,dt_N \cdots dt_1 \end{aligned} \tag{2.1}$$

Written as

$$|P(t)\rangle=\sum_{N=0}^{\infty}\int_{t\geqslant t_1\geqslant\cdots\geqslant t_N}\cdots\int\sum_{x_1,\dots,x_N}\prod_{j=1}^{N}\mathscr{T}_{x_j}(t_j)\,|0\rangle\,dt_N\cdots dt_1 \tag{2.2}$$

this represents a weighted sum over walks $x_1,\dots,x_N$ on the lattice, taken for convenience as moving backward in time from t to $-\infty$. The sum can be substantially simplified. To do so, consider the typical term

$$|I\rangle=\prod_{1}^{N}\left[(a_{x_j}^{+}-\bar{n}_{x_j})\exp\left(-\sum_{z}\phi_{x_jz}n_z\right)\right]|0\rangle \tag{2.3}$$

and insert pairs of reciprocal exponentials to rewrite it as

$$\begin{aligned}|I\rangle=\prod_{1}^{N}&\left\{\left[\exp\left(-\sum_{i=1}^{j-1}\sum_{z}\phi_{x_iz}n_z\right)\right](a_{x_j}^{+}-\bar{n}_{x_j})\exp\left(\sum_{i=1}^{j-1}\sum_{z}\phi_{x_iz}n_z\right)\right\}\\&\times\exp\left(-\sum_{i=1}^{N}\sum_{z}\phi_{x_iz}n_z\right)|0\rangle\end{aligned} \tag{2.4}$$

Since $e^{-\alpha n}(a^{+}-\bar{n})e^{\alpha n}=e^{-\alpha}a^{+}-\bar{n}$, and $n\,|0\rangle=0$, it follows that

$$|I\rangle=\prod_{1}^{N}\left(\left[\exp\left(-\sum_{i=1}^{j-1}\phi_{x_ix_j}\right)\right]a_{x_j}^{+}-\bar{n}_{x_j}\right)|0\rangle \tag{2.5}$$

But the $x_1,\dots,x_N$ need not be distinct. Suppose we denote the location of the first occurrence (in the order $1,\dots,N$) of the jth distinct site by $\sigma(j)$, $j=1,\dots,d\leqslant N$. Then, since

$$(e^{-\alpha}a^{+}-\bar{n})(e^{-\beta}a^{+}-\bar{n})=-(e^{-\alpha}a^{+}-\bar{n}) \tag{2.6}$$

(2.5) is further reduced to

$$|I\rangle=(-1)^{N-d}\prod_{1}^{d}\left\{\left[\exp\left(-\sum_{i=1}^{j-1}\phi_{x_ix_{\sigma(j)}}\right)\right]a_{x_{\sigma(j)}}^{+}-\bar{n}_{x_{\sigma(j)}}\right\}|0\rangle \tag{2.7}$$

whose appropriate sum and integral then yields $|P(t)\rangle$. Of course, we are not concerned with the full probability distribution, but rather with significant expectations. In particular, the mean occupation of site x at time t is given by

$$n_x(t)=\sum_{\{n_z=0,1\}}\langle\{n_z\}|\,n_x\,|P(t)\rangle=\langle U|\,n_x\,|P(t)\rangle \tag{2.8}$$

where U represents the direct product of the row vectors

$$|u_z\rangle=(1,1) \tag{2.9}$$

Since $\langle u|\, e^{-\alpha}a^{+} - \bar{n}\,|0\rangle = e^{-\alpha} - 1$, the $j=1$ term in (2.7), inserted into (2.8), will be nonvanishing only if $x_1 = x$, in which case $\langle u|\, n(a^{+} - \bar{n})\,|0\rangle = 1$. Hence, (2.7) applied to (2.2) via (1.5) yields the desired representation

$$n_x(t) = \sum_{N=1}^{\infty} \int_{t \geqslant t_1 \geqslant \cdots \geqslant t_N} \cdots \int \sum_{\{x_1,\ldots,x_N\}} \delta_{x_1,x}(-1)^{N-d} \prod_1^N f_{x_i}(t_i) \times \prod_{j=2}^{d} \left[\exp\left(-\sum_{i=1}^{j-1} \phi_{x_i x_{\sigma(j)}}\right) - 1\right] dt_1 \cdots dt_N \tag{2.10}$$

Note that in the case of a continuum of sites, where the probability of two sites on a walk coinciding goes to zero, (2.10) reduces simply to

$$n(x,t) = \sum_{N=1}^{\infty} \int_{t \geqslant t_1 \geqslant \cdots \geqslant t_N} \cdots \int \int \cdots \int \delta(x_1 - x) \prod_{j=2}^{N} \left(\exp\left[-\sum_{i=1}^{j-1} \phi(x_i, x_j)\right] - 1\right) \times \prod_1^N f(x_i, t_i)\, dx_1 \cdots dx_N\, dt_1 \cdots dt_N \tag{2.11}$$

Equation (2.11) can be cast in a form similar to the equilibrium Mayer expansion by using

$$\prod_{\alpha} A_\alpha - 1 = \sum_{\phi \neq \Lambda \subset \{\alpha\}} \prod_{\alpha \in \Lambda} (A_\alpha - 1)) \tag{2.12}$$

to rewrite (2.11) as[1,5]

$$n(x,t) = \sum_{N=1}^{\infty} \int_{t \geqslant t_1 \geqslant \cdots \geqslant t_N} \cdots \int \int \cdots \int \delta(x_1 - x) \prod_1^N f(x_i, t_i) \times \sum_{\Lambda} \prod_{(i,j) \in \Lambda} (e^{-\phi(x_i, x_j)} - 1)\, dx_1 \cdots dx_N\, dt_1 \cdots dt_N \tag{2.13}$$

where Λ denotes a subset of ordered pairs of indices (i,j), $i < j \leqslant N$, for which each value of j occurs at least once.

3. EXCLUSION INTERACTION

Let us return to RSA on a lattice and confine our attention to strictly hard interactions, i.e., those for which $\phi_{xy} = \infty$ for $x \in \mathscr{E}(y)$, the exclusion region of y, and otherwise $\phi_{xy} = 0$; we will also assume that $x \in \mathscr{E}(y) \Rightarrow y \in \mathscr{E}(x)$. Now Eq. (2.10) simplifies considerably: each factor of $\prod_{j=2}^{d}$ is either 0 or -1, and takes the latter value when $x_{\sigma(j)}$ is in the exclusion region of some x_i for $i < j$. Since every repeat of $x_{\sigma(j)}$ will automatically be in the exclusion region of a previous site, (2.10) can then be rewritten, slightly redundantly, as

$$n_x(t) = \sum_{N=1}^{\infty} (-1)^{N-1} \times \int \cdots \int_{t \geq t_1 \geq \cdots \geq t_N} \sum_{\{x_1,\dots,x_N\}}^{x_1 = x,\ x_j \in \bigcup_{i<j} \mathscr{E}(x_i)} \prod_1^N f_{x_i}(t_i)\, dt_1 \cdots dt_N \tag{3.1}$$

Roughly speaking, the walk forward in time with exclusion is replaced by one which is backward in time but in which a landing must be made in some exclusion zone. This, together with the factor $(-1)^{N-1}$, suggests some sort of inclusion–exclusion relation, and indeed may be derived on purely combinatorial grounds.[6]

The fact that (3.1) does contain repeats suggests that something like (2.10), in which distinct sites are singled out, may be more appropriate. To make this more concrete, we observe that in the full sum (3.1), a first occurrence of y at t' will be followed by, say, s occurrences of y at $t'_1,\dots, t'_s$ which have no effect on the restrictions on succeeding members of the walk. Since $t'_1,\dots, t'_s$ can appear arbitrarily, subject only to $t' \geq t'_1 \cdots \geq t'_s$, $f_y(t')$ then appears only in the combination

$$\begin{aligned} F_y(t') &= f_y(t') \sum_{s=0}^{\infty} (-1)^s \int \cdots \int_{t' \geq t'_1 \geq \cdots \geq t'_s} \prod_1^s f_y(t'_i)\, dt'_1 \cdots dt'_s \\ &= f_y(t') \sum_{s=0}^{\infty} (-1)^s/s! \left[\int_{-\infty}^{t'} f_y(\tau)\, d\tau \right]^s \\ &= f_y(t') \exp - \int_{-\infty}^{t'} f_y(\tau)\, d\tau \end{aligned} \tag{3.2}$$

With this amalgamation of terms, repeated sites are no longer present, and so (3.1) is transformed to[4]

$$n_x(t) = \sum_{N=1}^{\infty} (-1)^{N-1} \times \int \cdots \int_{t \geq t_1 \geq \cdots \geq t_N} \sum_{\{x_1,\dots,x_N\}}^{x_1 = x,\ x_j \in \bigcup_{i<j} \mathscr{E}(x_i) - \bigcup_{i<j} x_i} \prod_1^N F_{x_i}(t_i)\, dt_1 \cdots dt_N \tag{3.3}$$

It will also be useful to record the time derivative of $n_x(t)$,

$$\dot{n}_x(t) = F_x(t) \sum_{N=1}^{\infty} (-1)^{N-1} \times \int \cdots \int_{t \geq t_2 \geq \cdots \geq t_N} \sum_{\{x_1 = x,\, x_2,\dots,x_N\}}^{x_j \in \bigcup_{i<j} \mathscr{E}(x_i) - \bigcup_{i<j} x_i} \prod_2^N F_{x_i}(t_i)\, dt_2 \cdots dt_N \tag{3.4}$$

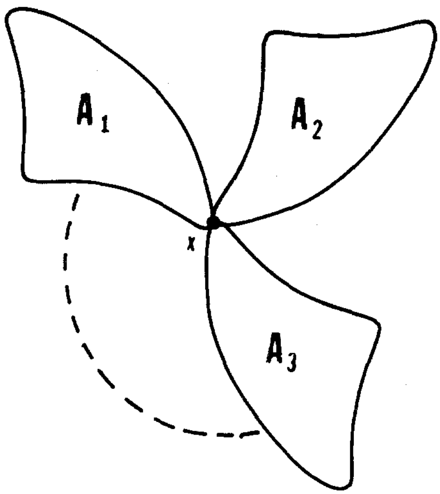

Fig. 1. Lattice configuration with articulation point.

Explicit evaluation of (3.3) depends very much upon the complexity of the lattice and the exclusion regions. Let us now suppose that only nearest neighbor sites are excluded (as well as the site itself). The simplest local form of a lattice is an articulation point, a site x whose excision cuts the lattice L into disconnected pieces $A_1, A_2,..., A_s$: $L = \bigcup_i A_i$, $A_i \cap A_j = x$ for $i \neq j$ (Fig. 1). Consider $\dot{n}_x^L(t)$ (superscript denoting the lattice being referred to) under these circumstances. In any sequence $x, x_2,..., x_N$, the sites belonging to A_1 can occur at arbitrary—but ordered—times, and similarly with $A_2, A_3,...$. Thus the s subsequences occur independently in $x, x_2,..., x_N$ aside from the common root x at t. We conclude at once that[7]

$$\text{if} \quad L = \bigcup_i A_i, \qquad \text{where} \quad A_i \cap A_j = x \quad \text{for} \quad i \neq j$$

then for nearest neighbor exclusion

$$\dot{n}_x^L(t)/F_x(t) = \prod_i \left[\dot{n}_x^{A_i}(t)/F_x(t)\right] \tag{3.5}$$

Equation (3.5) is particularly relevant to a lattice that is a tree, i.e., for which all sites are articulation points, but is of course not sufficient to solve for $n_x(t)$. For this purpose, we need another reduction formula. Suppose that L is constructed by hanging another site x onto a site $y \in A$ (Fig. 2). Then any term in $\dot{n}_x(t)$ starts with $F_x(t)$, is necessarily followed by $F_y(t_2)$, and has no further restriction enforced by x. It follows at once that[7] if $L = A \cup (x, y)$, where $A \cap (x, y) = y$

then for nearest neighbor exclusion

$$\frac{d}{dt}\left(\frac{\dot{n}_x^L(t)}{F_x(t)}\right) = -\dot{n}_y^A(t) \tag{3.6}$$

We can now proceed to the solution of RSA on a tree, which will appear in the increasingly common inverse form: $f_x(t)$ as a functional of $\{n_y(t)\}$. As a preliminary, we apply the preceding to the case of two neighboring articulation points x and y, roots of sublattices A and B (Fig. 3). Denote the complementary sublattice of A, together with the root x, as $\bar{A}$, and similarly with $\bar{B}$. Now, according to (3.5), we have

$$F_x(t)\,\dot{n}_x(t) = \dot{n}_x^A(t)\,\dot{n}_x^{\bar{A}}(t) \tag{3.7a}$$

$$F_y(t)\,\dot{n}_y(t) = \dot{n}_y^B(t)\,\dot{n}_y^{\bar{B}}(t) \tag{3.7b}$$

and, according to (3.6),

$$\frac{d}{dt}\frac{\dot{n}_x^{\bar{A}}(t)}{F_x(t)} = -\dot{n}_y^B(t) \tag{3.8a}$$

$$\frac{d}{dt}\frac{\dot{n}_y^{\bar{B}}(t)}{F_y(t)} = -\dot{n}_x^A(t) \tag{3.8b}$$

Combining (3.7b) and (3.8a) tells us (no superscript implies the full lattice) that

$$\frac{\dot{n}_x^{\bar{A}}(t)}{F_x(t)}\frac{d}{dt}\frac{\dot{n}_y^{\bar{B}}(t)}{F_y(t)} = -\dot{n}_x(t) \tag{3.9}$$

and similarly from (3.7a) and (3.8b),

$$\frac{\dot{n}_y^{\bar{B}}(t)}{F_y(t)}\frac{d}{dt}\frac{\dot{n}_x^{\bar{A}}(t)}{F_x(t)} = -\dot{n}_y(t) \tag{3.10}$$

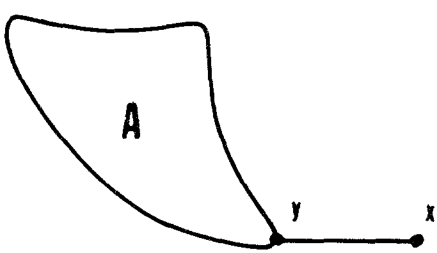

Fig. 2. Lattice configuration with hanging bond.

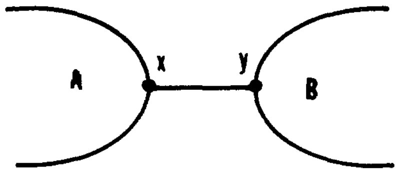

Fig. 3. Lattice configuration with neighboring articulation points.

From (3.9) and (3.10),

$$\frac{d}{dt}\left(\frac{\dot{n}_x^{\bar{A}}(t)}{F_x(t)}\frac{\dot{n}_y^{\bar{B}}(t)}{F_y(t)}\right) = -\dot{n}_x(t) - \dot{n}_y(t) \tag{3.11}$$

but $\dot{n}_x^C(t)/F_x(t) \to 1$ as $t \to -\infty$ for any $C \ni x$, so that

$$\frac{\dot{n}_x^{\bar{A}}(t)}{F_x(t)}\frac{\dot{n}_y^{\bar{B}}(t)}{F_y(t)} = 1 - n_x(t) - n_y(t) \tag{3.12}$$

Finally, dividing (3.10) by (3.12), we obtain

$$\frac{\dot{n}_y(t)}{1 - n_x(t) - n_y(t)} = -\frac{F_x(t)}{\dot{n}_x^{\bar{A}}(t)}\frac{d}{dt}\frac{\dot{n}_x^{\bar{A}}(t)}{F_x(t)} \tag{3.13}$$

which integrates at once to the desired

$$\frac{\dot{n}_x^{\bar{A}}(t)}{F_x(t)} = \exp - \int_{-\infty}^{t} \frac{\dot{n}_y(\tau)}{1 - n_x(\tau) - n_y(\tau)}\, d\tau \tag{3.14}$$

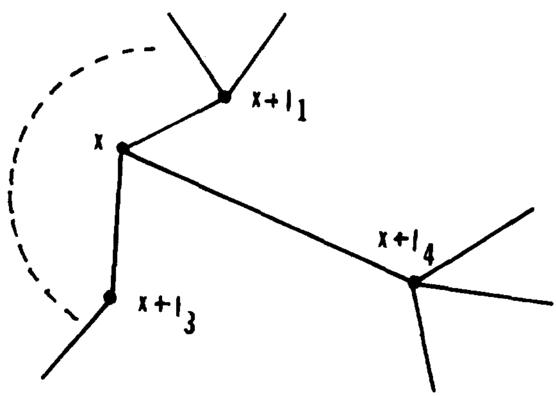

Fig. 4. Tree configuration with specified vertex.

Now consider the point x on a tree, together with its neighbors $x+\sigma$, $\sigma = l_1, l_2,\ldots$ (Fig. 4). In obvious notation patterned after the above, we have

$$\frac{\dot{n}_x(t)}{F_x(t)} = \prod_\sigma \frac{n_x^{\bar{A}_\sigma}(t)}{F_x(t)} \tag{3.15}$$

and so, inserting the result (3.14) for each branch, conclude that

$$F_x(t) = \dot{n}_x(t) \exp \sum_\sigma \int_{-\infty}^t \frac{\dot{n}_{x+\sigma}(\tau)}{1 - n_x(\tau) - n_{x+\sigma}(\tau)}\, d\tau \tag{3.16}$$

in the inverse form that was promised.

4. DISCUSSION

Analysis of inhomogeneous RSA on simply connected lattices with nearest neighbor exclusion is, as we have seen, not difficult and not complicated. This suggests that more complex lattices which locally resemble such ideal cases can use them as effective references, and such preliminary investigations have been made. However, a deeper feeling for the structure of RSA on non-simply-connected lattices can best be achieved by exact solution of as many models as feasible, starting, e.g., with Husimi trees or cacti,[8] or perhaps with a few connected circuits. These studies will be reported in the near future.

Returning briefly to the tree lattices that have been treated here, we may ask under what circumstances (3.16) can be cast in the more direct operational form: given the adsorption rates $\{f_x(t)\}$, determine the densities, or coverages, $\{n_x(t)\}$? The simplest case of course is that in which the process is uniform on an infinite tree or Bethe lattice, of coordination number q. Then the subscript x can be dropped, and (3.16) reduces to

$$\begin{aligned} F(t) &= \dot{n}(t) \exp q \int_{-\infty}^t \dot{n}(\tau)/[1 - 2n(\tau)]\, d\tau \\ &= \dot{n}(t)/[1 - 2n(t)]^{q/2} \end{aligned} \tag{4.1}$$

From the definition (3.2), we can integrate (4.1) to obtain

$$1 - \exp - \int_{-\infty}^t f(\tau)\, d\tau = \{[1 - 2n(t)]^{(2-q)/2} - 1\}/(q-2) \tag{4.2}$$

and hence solve as

$$n(t) = \tfrac{1}{2}\left(1 - \left\{(q-1) - (q-2)\exp\left[-\int_{-\infty}^t f(\tau)\, d\tau\right]\right\}^{2/(2-q)}\right) \tag{4.3}$$

which is a trivial generalization (with an obvious renormalization of time) of a previously derived result.[4, 9]

Matters become potentially more interesting when some degree of nonuniformity is present. Simplest perhaps is one which is locally bipartite: we divide the lattice into A and B sites so that A has only B as a neighbor and vice versa, and allow $f_B(t) \neq f_A(t)$. Then (3.16) reduces to the pair

$$\begin{aligned} F_A(t) &= \dot{n}_A(t) \exp q \int_{-\infty}^{t} \frac{\dot{n}_B(\tau)\, d\tau}{1 - n_A(\tau) - n_B(\tau)} \\ F_B(t) &= \dot{n}_B(t) \exp q \int_{-\infty}^{t} \frac{\dot{n}_A(\tau)\, d\tau}{1 - n_A(\tau) - n_B(\tau)} \end{aligned} \tag{4.4}$$

Let us adopt the notation

$$S_x(t) = \dot{n}_x(t)/F_x(t) \tag{4.5}$$

for the normalized response, so that $S_x(-\infty) = 1$. Now multiplying the pair (4.4) gives us

$$S_A(t)\, S_B(t) = \exp - q \int_{-\infty}^{t} [\dot{n}_A(\tau) + \dot{n}_B(\tau)]/[1 - n_A(\tau) - n_B(\tau)]\, d\tau$$

or

$$S_A(t)\, S_B(t) = [1 - n_A(t) - n_B(t)]^q \tag{4.6}$$

or substituting back into the derivatives of (4.4),

$$\begin{aligned} \dot{S}_A(t) &= -qF_B(t)[S_A(t)\, S_B(t)]^{1-1/q} \\ \dot{S}_B(t) &= -qF_A(t)[S_A(t)\, S_B(t)]^{1-1/q} \end{aligned} \tag{4.7}$$

In terms of

$$R_x(t) = S_x(t)^{1/q} \tag{4.8}$$

these yield the relatively simple pair

$$\begin{aligned} \dot{R}_A(t) &= -F_B(t)\, R_B(t)^{q-1} \\ \dot{R}_B(t) &= -F_A(t)\, R_A(t)^{q-1} \end{aligned} \tag{4.9}$$

Although the solution of (4.9) is not transparent, the line lattice case, $q = 2$, is relatively clear. In terms of $(1 \geqslant X \geqslant 0)$

$$X = \int_t^{\infty} F_A(\tau)\, d\tau = \exp\left[-\int_{-\infty}^{t} f_A(\tau)\, d\tau \right] \tag{4.10}$$

we have $R'_A = F_A F_B R_B$, $R'_B = R_A$, or

$$R''_B(X) = F_A(X)\, F_B(X)\, R_B(X) \tag{4.11}$$

which can be examined at leisure. But it seems clear from the foregoing that the inverse form is the approach of choice for inhomogeneous RSA. There remains the task of setting up thermodynamic generating functions to make this approach more compact, as in the customary equilibrium theory. Possible ways of doing so are now under study.

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REFERENCES

1. G. Tarjus, P. Schaaf, and J. Talbot, *J. Stat. Phys.* **63**:167 (1991).
2. D. K. Hoffman, *J. Chem. Phys.* **65**:95 (1976).
3. R. Dickman, J. S. Wang, and I. Jensen, *J. Chem. Phys.* **94**:8252 (1991).
4. Y. Fan and J. K. Percus, *Phys. Rev. A* **44**:5099 (1991).
5. J. A. Given, *Phys. Rev. A* **45**:816 (1992).
6. Y. Fan and J. K. Percus, *Comb. Prob. Comp.* **1**:219 (1992).
7. J. K. Percus, CECAM Workshop, Paris (1992).
8. Y. Fan and J. K. Percus, *Phys. Rev. Lett.* **67**:1677 (1991).
9. J. W. Evans, *Phys. Rev. Lett.* **62**:2642 (1989).

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