Inhomogeneous random sequential adsorption with equilibrium initial conditions

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We study the exact solution for the irreversible addition of nearest-neighbor hard-core particles to a lattice structure occupied at an initial time by particles of the same kind that are in a thermal equilibrium state. The adsorption probabilities are inhomogeneous in both time and space. This problem is attacked via the Bethe lattice, whose topology provides local forms of inverse relations with site coverages as controlling variables, separately for equilibrium as well as for nonequilibrium regimes. It is shown that the interference of the two inverse formats does not break their locality, due to a factorization property of equilibrium multisite correlations. The complete inverse solution is used to point out the absence of nonequilibrium phase transitions within irreversible stochastic dynamics.

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I. INTRODUCTION

Random sequential adsorption (RSA) [1–4] is the extreme of an irreversible process where the desorption of particles adsorbed onto a surface, and their diffusion, are neglected on a short time scale. The consequent lack of detailed balance leads to an asymptotic probability distribution which is very different from that in equilibrium.

Idealized RSA is perhaps the best candidate for studying the role of cooperativity in nonequilibrium by means of standard methods of equilibrium thermodynamics. Such attempts have already been made within the framework of Kirkwood-Salsburg equations [5] and the liquid-state replica method [6,7]. The existence of a close relationship between the formal structure of RSA and the canonical formalism as known in equilibrium statistics is supported by a recent study [8] of the RSA version that emphasizes the inhomogeneity of adsorption probabilities in both space and time. Within the inverse format, with particle densities as controlling variables, the (inverse) particle flux-site coverage relation turns out to be local for the addition of gas particles with a nearest-neighbor (NN) exclusion zone onto an initially empty tree lattice. Exactly the same behavior has been observed in the inverse treatment of the equilibrium counterpart of the gas system, formulated on lattice structures possessing articulation points [9–11]: the inhomogeneous external potential depends locally on the evoked density profile. This topological property provides an explicit local form for the associated density functional, which is the starting point for accounting for nonlocal global effects due to chain closing [12], or to network multiconnectivity of superbond type [13], via collective modes.

A next natural step towards the understanding of the role of simple connectivity in the inverse description is the fusion of equilibrium and nonequilibrium local formats. In this paper, we report the study of this fusion via the most general version of the RSA process which is, in terms of adsorption probabilities, inhomogeneous in space time and starts from an arbitrary equilibrated initial state. This corresponds to an experimental situation of monolayer deposition from solution onto a surface which is not free of particles before deposition (for simplicity, NN hard-core range is considered for adsorbing as well as "surface" particles). From a theoretical point of view, consideration of a nonempty initial state is the natural condition for potential RSA generation of glassy configurations in a two-dimensional (2D) hard disc system [5,14], with saturation density higher than the freezing density. The study of RSA on simply connected structures, e.g., a Bethe lattice that mimics locally a regular structure of the same coordination number, could provide a significant picture of the interplay between equilibrium and nonequilibrium regimes, as well as time evolution of realistic systems. Exact analysis shows that, owing to a factorization form of equilibrium multisite correlations, interference of the equilibrium and nonequilibrium inverse formats does not break their locality. The problem of translationally invariant RSA process with nontrivial initial conditions has been studied in Ref. [15], using the "shielding property" of empty sites, and in Ref. [5], within Kirkwood-Salsburg theory.

We first outline a derivation of the exact inverse format for the inhomogeneous 1D case (Secs. II and III) in order to motivate an exact treatment of the Bethe lattice (Sec. IV). As a by-product of this treatment, we present in Sec. V the solution of the continuous homogeneous version of 1D RSA, known as the random parking problem [16–18], starting from an equilibrium initial state. In Sec. VI, a proof of the absence of nonequilibrium transition from a fluid to a crystal phase under homogeneous adsorption rates is presented.

II. RANDOM WALK FORMULATION

A number of different techniques have been used to obtain exact solutions for RSA on selected lattices. Let us

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start our investigation by recalling one of these [8], which proceeds by constructing the transition operator for a dynamical event and then iterating it. We will adopt the notation that a site x is either empty, symbolized by $\sigma_x=0$, or occupied, $\sigma_x=1$; we will also define the "hole" occupation $\bar{\sigma}_x \equiv 1-\sigma_x$. Attention will be restricted to events that obey the nearest-neighbor exclusion rule: if $\sigma_x=1$, then $\sigma_y=0$ for all $\langle y,x\rangle$ (read y is a nearest neighbor of x; the expression "nearest neighbor" can be replaced by that of belonging to the exclusion zone E_x of site x). Then for a particle flux $f_x(t)$ per unit time into x at time t, the transition rate operator at site x will clearly be given by

$$T_x(t)=f_x(t)(a_x^\dagger-\bar{\sigma}_x)\prod_{\langle y,x\rangle}\bar{\sigma}_y \,, \qquad \text{where } a_x^\dagger g(\sigma_x)=\sigma_x g(0) \,, \tag{2.1}$$

$a_x^\dagger$ is the particle creation operator at x. Correspondingly, the dynamics of the full lattice is determined by

$$\frac{\partial}{\partial t}p(\sigma,t)=T(t)p(\sigma,t) \,, \qquad T(t)=\sum_x T_x(t) \,, \tag{2.2}$$

where $p(\sigma,t)$ is the occupation distribution function for the full lattice. Equation (2.2) has the usual ordered exponential solution

$$p(\sigma,t)=:\exp\int_0^t T(\tau)d\tau : p(\sigma,0) = \sum_{N=0}^{\infty}\int\cdots\int_{t\ge t_{i-1}\ge t_i\ge 0} T(t_1)T(t_2)\cdots T(t_N) \times dt_1 dt_2\cdots dt_N p(\sigma,0) \,. \tag{2.3}$$

The information of particular relevance to us will be expressed by the various lattice multisite densities, and for technical reasons, by the multihole densities

$$\bar{p}_A(t)=\sum_\sigma\left[\prod_{x\in A}\bar{\sigma}_x\right]p(\sigma,t) \tag{2.4}$$

for a given set A. A typical term to be evaluated in (2.3) then takes the form

$$\sum_\sigma\left[\prod_{x\in A}\bar{\sigma}_x\right]\sum_N(-1)^N\prod_{j=1}^N\left[(\bar{\sigma}_{x_j}-a_{x_j}^\dagger)\prod_{\langle y,x_j\rangle}\bar{\sigma}_y\right] \times\prod_{j=1}^N f_{x_j}(t_j)p(\sigma,0) \tag{2.5}$$

for each possible "walk" $x_1,\dots,x_n$ backwards in time. Let us push all the factors $\bar{\sigma}_{x_j}-a_{x_j}^\dagger$ to the left. Since

$$(\bar{\sigma}_x-a_x^\dagger)(\bar{\sigma}_x-a_x^\dagger)=(\bar{\sigma}_x-a_x^\dagger)\,, \quad \bar{\sigma}_x(\bar{\sigma}_x-a_x^\dagger)=\bar{\sigma}_x\,, \quad \text{but } \sum_\sigma(\bar{\sigma}_x-a_x^\dagger)g(\sigma)=0 \text{ for all } g \,, \tag{2.6}$$

such an operation with $\bar{\sigma}_{x_j}-a_{x_j}^\dagger$ gives a value of 0 unless x_j is either in the exclusion zone E_{x_i}, $i<j$, of a previous member of the walk, or belongs to the root set A. The resulting weight of an allowed walk is then $\sum_\sigma\prod_{y\in A\cup_1^N E_{x_j}}\bar{\sigma}_y p(\sigma,0)=\langle\prod_{y\in A\cup_1^N E_{x_j}}\bar{\sigma}_y\rangle^*$, where $*$ will uniformly refer to evaluation at time 0. An internal site y of A (one satisfying $E_y\subset A$) can occur any number of times, interleaving at any times between 0 and t in this walk without affecting any exclusion condition. Such a site hence amasses a total flux weight of

$$1-\int_0^t f_y(\tau)d\tau+\int_0^t\int_0^{\tau'}f_y(\tau)f_y(\tau')d\tau d\tau'+\cdots =\exp\left[-\int_0^t f_y(\tau)d\tau\right] .$$

Any other site can repeat after its initial t' landing any number of times, and at any past times, without affecting exclusion conditions, creating the weight

$$f_y(t')-f_y(t')\int_0^{t'}f_y(\tau)d\tau+f_y(t')\int_0^{t'}\int_0^{\tau'}f_y(\tau)f_y(\tau')d\tau d\tau'+\cdots=f_y(t')\exp\left[-\int_0^{t'}f_y(\tau)d\tau\right] ,$$

a renormalized flux. We conclude that (2.3)–(2.5) reduce to

$$\bar{p}_A(t)\Big/\left[\prod_{y\in\mathrm{int}(A)}G_y(t)\right]=\sum_{N=0}^{\infty}(-1)^N\int\cdots\int_{t\ge t_{i-1}\ge t_i\ge 0}\sum_{\{x_i\in A\cup_1^{i-1}E_{x_j}-\mathrm{int}(A)\}}{}^{\!\!\prime}\;\prod_1^N F_{x_i}(t_i)\left\langle\prod_{y\in A\cup_1^N E_{x_j}}\bar{\sigma}_y\right\rangle^* dt_1\cdots dt_N \,,$$

$$\text{where } F_y(t)=f_y(t)\exp\left[-\int_0^t f_y(\tau)d\tau\right],\quad G_y(t)=\exp\left[-\int_0^t f_y(\tau)d\tau\right] \tag{2.7}$$

and $\sum'$ denotes a sum over walks *without* repetition.

III. 1D RSA FROM INITIAL EQUILIBRIUM

A. Equilibrium inverse format

The simplest nontrivial lattice is a one-dimensional integer grid. We want to examine RSA starting from a (quenched) equilibrium state, and so we must first determine the properties of the equilibrium distribution in the 1D case with NN exclusion interaction among particles,

$$p(\sigma,0)=\frac{1}{Z}\prod_x z_x^{\sigma_x}\prod_{\langle x,x+1\rangle}(1-\sigma_x\sigma_{x+1}) \,. \tag{3.1}$$

Here, z_x stands for a site-dependent activity reflecting the effect of the external potential on a particle at site x and the statistical sum

$$Z=\sum_{\sigma}\prod_{x} z_x^{\sigma_x} \prod_{\langle x,x+1\rangle}(1-\sigma_x\sigma_{x+1}) \tag{3.2}$$

ensures the normalization of probabilities. Its logarithm is the generator for particle densities $\rho_x^*=\langle\sigma_x\rangle^*$ according to

$$\rho_x^*=\frac{\partial}{\partial\ln(z_x)}\ln Z \ . \tag{3.3}$$

The specification of the set of controlling variables, either $\{z_x\}$ or $\{\rho_x^*\}$, automatically determines the form of all correlations in the system within the canonical formalism.

The open chain represents an example of a simply connected lattice characterized by the articulation nature of each of its vertices. For such structures it has been proven [10] that the potential at a given site x is topologically independent of the density profiles inside lattice subsets that can be separated from x via articulation points $\neq x$. In our case, we can thus reduce the original chain to a finite cluster of sites $\{x-1,x,x+1\}$ with the given particle densities $\{\rho_{x-1}^*,\rho_x^*,\rho_{x+1}^*\}$ in which the boundary activities are modified, $z_{x-1}\to\tilde{z}_{x-1}, z_{x+1}\to\tilde{z}_{x+1}$, with no effect on z_x itself. Within this local cluster, the inverse profile equation is readily computed in the form

$$z_x=\frac{\rho_x^*(1-\rho_x^*)}{(1-\rho_x^*-\rho_{x-1}^*)(1-\rho_x^*-\rho_{x+1}^*)} \ . \tag{3.4}$$

Very similar considerations can be applied to the calculation of a multisite hole density $\langle\bar\sigma_x\cdots\bar\sigma_{x+h}\rangle^*$ $(h=1,2,\ldots)$. The separation of the site cluster $\{x,\ldots,x+h\}$ from the chain is accompanied by the modification of boundary activities $z_x\to\tilde{z}_x=\rho_x^*/(1-\rho_x^*-\rho_{x+1}^*)$, $z_{x+h}\to\tilde{z}_{x+h}=\rho_{x+h}^*/(1-\rho_{x+h}^*-\rho_{x+h-1}^*)$ and by the appearance of a prefactor which plays no role in the calculation of the correlation $\langle\bar\sigma_x\cdots\bar\sigma_{x+h}\rangle^*$. The last is evidently given by $1/Z(x,x+h)$ where $Z(x,x+h)$ is the partition function of the reduced cluster $\{x,\ldots,x+h\}$. In order to calculate $Z(x,x+h)$, we will eliminate successively the boundary cluster points. Elimination of, say x, picks up the multiplicative contribution $1+\tilde{z}_x=(1-\rho_{x+1}^*)/(1-\rho_x^*-\rho_{x+1}^*)$ as well as multiplying z_{x+1} by the factor $(1-\rho_x^*-\rho_{x+1}^*)/(1-\rho_{x+1}^*)$, keeping in this way the correct profile relation for site $x+1$ upon the deletion of site x. Performing this procedure at every elimination level down to zero cluster points, the factors generated imply

$$Z(x,x+h)=\prod_{y=x+1}^{x+h-1}(1-\rho_y^*)\Big/\prod_{y=x}^{x+h-1}(1-\rho_y^*-\rho_{y+1}^*) \ ,$$

so that one finds

$$\langle\bar\sigma_x\cdots\bar\sigma_{x+h}\rangle^*=\frac{\prod_{y=x}^{x+h-1}\langle\bar\sigma_y\bar\sigma_{y+1}\rangle^*}{\prod_{y=x+1}^{x+h-1}\langle\bar\sigma_y\rangle^*}=\frac{\prod_{y=x}^{x+h-1}(1-\rho_y^*-\rho_{y+1}^*)}{\prod_{y=x+1}^{x+h-1}(1-\rho_y^*)} \ , \tag{3.5}$$

a Markov field property of the underlying inhomogeneous system. The multisite hole density factorizes into NN two-site probabilities of empty sites; the denominator compensates the "superfluous" presence of internal sites in NN pairs.

Besides the local inverse description in terms of the interior of a cluster taken off the chain, it is useful to develop simultaneously a complementary inverse treatment based on the investigation of the modification of statistical quantities associated with the chain after deleting some of its geometrical elements, one or a sequence of sites or bonds. Let us "delete," e.g., the bond between sites x and $x+1$ as follows. In the statistical sum Z and in every ensemble average we treat the NN exclusion term $(1-\sigma_x\sigma_{x+1})$ as a "correlation term" in the ensemble average of the lattice without $\langle x,x+1\rangle$ bond. The consequent transition from the original chain to a system of disconnected left (L) and right (R) chain fragments corresponds to the replacements $Z\to Z_0$, $\rho_x^*\to\langle\sigma_x\rangle_L^*$, and $\rho_{x+1}^*\to\langle\sigma_{x+1}\rangle_R^*$ (the sites are labeled from left to right and quantities associated with both L and R chain fragments will be denoted by subscript 0), while the NN correlation $\langle\sigma_x\sigma_{x+1}\rangle_0^*$ decouples exactly as $\langle\sigma_x\rangle_L^*\langle\sigma_{x+1}\rangle_R^*$ because of the statistical independence of the chain fragments. This gives rise to a closed system of equations,

$$\begin{aligned} Z&=Z_0(1-\langle\sigma_x\rangle_L^*\langle\sigma_{x+1}\rangle_R^*) \ , \\ \rho_x^*Z&=Z_0(\langle\sigma_x\rangle_L^*-\langle\sigma_x\rangle_L^*\langle\sigma_{x+1}\rangle_R^*) \ , \\ \rho_{x+1}^*Z&=Z_0(\langle\sigma_{x+1}\rangle_R^*-\langle\sigma_x\rangle_L^*\langle\sigma_{x+1}\rangle_R^*) \ , \end{aligned} \tag{3.6}$$

from which the functional dependence of hole densities $\langle\bar\sigma_x\rangle_L^*,\langle\bar\sigma_{x+1}\rangle_R^*$, associated, respectively, with the L and R chain fragments, for densities ρ_x^*,ρ_{x+1}^*, associated with the continuous chain, can be deduced in the form

$$\langle\bar\sigma_x\rangle_L^*=\frac{1-\rho_x^*-\rho_{x+1}^*}{1-\rho_{x+1}^*}, \quad \langle\bar\sigma_{x+1}\rangle_R^*=\frac{1-\rho_x^*-\rho_{x+1}^*}{1-\rho_x^*} \ . \tag{3.7}$$

B. Nonequilibrium inverse format

It will suffice for our purposes to consider A in (2.7) as a connected set, the interval $[a,b]$; in this case, the set $A\cup_1^N E_{x_j}$ will also be an interval, say $[c,d]$, with $c\le a, d\ge b$; the corresponding equilibrium hole probability $\langle\bar\sigma_c\cdots\bar\sigma_d\rangle^*$ is given by (3.5). The factor

$\langle \prod_{a+1}^{b-1} \bar{\sigma}_x \rangle^*$ is common to all terms in (2.7). Furthermore, if $a<b$, the sites $\le a$ and those $\ge b$ impose no restrictions on each other, and their times of occupation can be interleaved arbitrarily. Since the component walks on left sites alone, or right sites alone, must now proceed monotonically along successive sites, we therefore have

$$\bar{p}_{[a,b]}(t)=\prod_{a+1}^{b-1} G_y(t)\bar{p}_{a,L}(t)\left\langle \prod_{a+1}^{b-1} \bar{\sigma}_x \right\rangle^* \bar{p}_{b,R}(t) , \tag{3.8}$$

where

$$\bar{p}_{a,L}(t)=\sum_0^\infty (-1)^N \int \cdots \int_{t\ge t_{i-1}\ge t_i \ge 0} \prod_1^N F_{a+1-i}(t_i) \left\langle \prod_0^{N+1} \bar{\sigma}_{a+1-i} \right\rangle^* \Big/ (1-\rho_{a+1}^*) dt_1 \cdots dt_N ,$$

$$\bar{p}_{b,R}(t)=\sum_0^\infty (-1)^N \int \cdots \int_{t\ge t_{i-1}\ge t_i \ge 0} \prod_1^N F_{b-1+i}(t_i) \left\langle \prod_0^{N+1} \bar{\sigma}_{b-1+i} \right\rangle^* \Big/ (1-\rho_{b-1}^*) dt_1 \cdots dt_N ,$$

a decomposition into unrelated left- and right-hand walks on half lines.

To clarify the physical interpretation of the left- and right-hand quantities $\bar{p}_{x,L}(t)$ and $\bar{p}_{x,R}(t)$ hereby introduced, let us generate a correlation hierarchy by multiplying both sides of master Eq. (2.2) by the corresponding state variables and then summing over all σ, taking advantage of the equality $\sum_{\sigma=0,1}(2\sigma-1)=0$:

$$\frac{\partial}{\partial t}\rho_x(t)=f_x(t)\bar{p}_{[x-1,x+1]}(t) , \tag{3.9a}$$

$$\begin{aligned}\frac{\partial}{\partial t}\bar{p}_{[x,x+h]}(t)=&-f_x(t)\bar{p}_{[x-1,x+h]}(t)\\ &-\sum_{x+1}^{x+h-1} f_y(t)\bar{p}_{[x,x+h]}(t)\\ &-f_{x+h}(t)\bar{p}_{[x,x+h+1]}(t) ,\ h\ge 2 .\end{aligned} \tag{3.9b}$$

Insertion of (3.8) into (3.9b) and use of the factorization property of equilibrium multisite correlations (3.5) then implies the evolution equations for site densities

$$\frac{\partial}{\partial t}\rho_x(t)=F_x(t)\bar{p}_{x-1,L}(t)\bar{p}_{x+1,R}(t) , \tag{3.10}$$

with

$$\begin{aligned}\frac{\partial}{\partial t}\bar{p}_{x,L}(t)&=-F_x(t)\langle\bar{\sigma}_x\rangle_L^*\bar{p}_{x-1,L}(t) ,\\ \frac{\partial}{\partial t}\bar{p}_{x,R}(t)&=-F_x(t)\langle\bar{\sigma}_x\rangle_R^*\bar{p}_{x+1,R}(t) .\end{aligned} \tag{3.11}$$

This is a generalization of the factorization property used by Hemmer and co-workers in their treatment of irreversible filling problems (see, e.g., [18]).

The independence of these boundary expansions on the cluster size $h+1$ tells us that the decomposition (3.8) solves exactly the infinite chain of equations of motion (3.9); the interrelations among different clusters are now realized through local relations (3.11). We emphasize that this property is a direct consequence of the internal structure of equilibrium multisite correlations (3.5). The topological decoupling of the infinite correlation hierarchy differs substantially from that observed in the Glauber dynamics [19] of the zero-field Ising model, or the voter model [20]. In these stochastic dynamics, the reason for the exact decoupling comes from the choice of transition probabilities, which implies the invariance of the correlation space of arbitrary order with respect to the evolution operator (this does not occur in the present case). Finally, writing down the evolution Eqs. (3.10) for $\langle\bar{\sigma}_x\rangle_L$ and $\langle\bar{\sigma}_x\rangle_R$ defined on the edges of L- and R-chain fragments, respectively,

$$\frac{\partial}{\partial t}\langle\bar{\sigma}_x\rangle_L=-F_x(t)\langle\bar{\sigma}_x\rangle_L^*\bar{p}_{x-1,L}(t) , \tag{3.12a}$$

$$\frac{\partial}{\partial t}\langle\bar{\sigma}_x\rangle_R=-F_x(t)\langle\bar{\sigma}_x\rangle_R^*\bar{p}_{x+1,R}(t) , \tag{3.12b}$$

and using the initial conditions $\bar{p}_{x,L}(0)=\langle\bar{\sigma}_x\rangle_L^*$, $\bar{p}_{x,R}(0)=\langle\bar{\sigma}_x\rangle_R^*$ given by (3.8) we conclude at once that

$$\bar{p}_{x,L}(t)=\langle\bar{\sigma}_x\rangle_L,\quad \bar{p}_{x,R}(t)=\langle\bar{\sigma}_x\rangle_R . \tag{3.13}$$

The solution of the nonequilibrium inverse problem is now strictly technical. Combining Eq. (3.10) with (3.12a), we get a formal expression for $\langle\bar{\sigma}_x\rangle_L$,

$$\langle\bar{\sigma}_x\rangle_L=\langle\bar{\sigma}_x\rangle_L^* \exp\left[-\int_0^t dt' \frac{\dot{\rho}_x\langle\bar{\sigma}_x\rangle_L^*}{\langle\bar{\sigma}_x\rangle^*[\langle\bar{\sigma}_x\rangle_L\langle\bar{\sigma}_{x+1}\rangle_R]}\right], \tag{3.14}$$

with the time-dependent quantities in the integrand being functions of t'. Since

$$\begin{aligned}\frac{\partial}{\partial t}[\langle\bar{\sigma}_x\rangle_L\langle\bar{\sigma}_{x+1}\rangle_R]=&-F_x(t)\langle\bar{\sigma}_x\rangle_L^*\langle\bar{\sigma}_{x-1}\rangle_L\langle\bar{\sigma}_{x+1}\rangle_R\\ &-\langle\bar{\sigma}_x\rangle_L F_{x+1}(t)\\ &\times\langle\bar{\sigma}_{x+1}\rangle_R^*\langle\bar{\sigma}_{x+2}\rangle_R\\ =&-\frac{\langle\bar{\sigma}_x\rangle_L^*}{\langle\bar{\sigma}_x\rangle^*}\dot{\rho}_x-\frac{\langle\bar{\sigma}_{x+1}\rangle_R^*}{\langle\bar{\sigma}_{x+1}\rangle^*}\dot{\rho}_{x+1} ,\end{aligned} \tag{3.15}$$

we find

$$\langle\bar{\sigma}_x\rangle_L\langle\bar{\sigma}_{x+1}\rangle_R=\frac{(1-\rho_x^*-\rho_{x+1}^*)(1-\rho_x-\rho_{x+1})}{(1-\rho_x^*)(1-\rho_{x+1}^*)} , \tag{3.16}$$

where the initial conditions (3.7) have been taken into account. Putting (3.16) into (3.14), one readily gets

$$\langle \bar{\sigma}_x \rangle_L = \langle \bar{\sigma}_x \rangle_L^* \exp\left[-\int_0^t dt' \frac{\dot{\rho}_x}{1-\rho_x-\rho_{x+1}} \right], \quad (3.17)$$

with a similar formula for the hole density defined on the R-chain fragment. Consequently, evolution Eq. (3.10) results in the exact inverse relation for the renormalized particle flux

$$F_x(t) = \dot{\rho}_x(t) \frac{(1-\rho_x^*)}{\prod_{\delta=\pm 1} (1-\rho_x^*-\rho_{x+\delta}^*)} \times \exp\left[\sum_{\delta=\pm 1} \int_0^t dt' \frac{\dot{\rho}_{x+\delta}(t')}{1-\rho_x(t')-\rho_{x+\delta}(t')} \right]. \quad (3.18)$$

Note that an implicit dependence on the initial densities occurs in the integrals.

For trivial initial equilibrium conditions $\rho_x^*=0$, Eq. (3.18) reproduces the exact inverse formula (3.16) of Ref. [8]. In the case of spatially homogeneous initial occupation $\rho_x^*=\rho^*$ and adsorption rates $f_x(t)=f(t)$, it reduces to

$$F(t) = \dot{\rho}(t) \frac{1-\rho^*}{(1-2\rho^*)^2} \exp\left[2\int_0^t dt' \frac{\dot{\rho}(t')}{1-2\rho(t')} \right] = \dot{\rho}(t) \frac{1-\rho^*}{(1-2\rho^*)[1-2\rho(t)]}, \quad (3.19)$$

which integrates to the ρ-f interrelation in a direct form

$$\rho(t) = \frac{1}{2} - \frac{(1-2\rho^*)}{2} \exp\left\{ 2\left[\frac{1-2\rho^*}{1-\rho^*} \right][G(t)-1] \right\}. \quad (3.20)$$

The fraction of asymptotically filled sites (the jamming limit) is then

$$\rho(t\to\infty) = \frac{1}{2} - \frac{(1-2\rho^*)}{2} \exp\left[-2\left[\frac{1-2\rho^*}{1-\rho^*} \right] \right]. \quad (3.21)$$

IV. INHOMOGENEOUS RSA ON A BETHE LATTICE

The formalism can be generalized straightforwardly to the RSA process on a Bethe lattice of arbitrary coordination number q. We first introduce some definitions of topological sets on the Bethe structure, allowing us to proceed as in the previous $q=2$ case. Let Ω be a finite sublattice which is a connected cluster of one- and q-coordinated vertices: $\Omega=\Omega_i \cup \partial\Omega$ where $\Omega_i=\{x\in\Omega : |x|=q\}$, $\partial\Omega=\{x\in\Omega : |x|=1\}$ where $|x|\equiv q_x$. The NN coordination of a site $x\in\Omega$ with respect to the interior and exterior of Ω is specified via the sets $\Omega(x)=\{y\in\Omega : y$ is NN of $x\}$ and $\Omega'(x)=\{y\notin\Omega : y$ is NN of $x\}$, constrained by $|\Omega(x)|+|\Omega'(x)|=q$. The introduction of the sets $\Omega(x), \Omega'(x)$ will be of practical interest only for boundary sites $x\in\partial\Omega$ where $|\Omega(x)|=1$; in this case the symbol $\Omega(x)$ will denote the corresponding single site element.

The analog of master Eq. (2.2) describing the dynamics of RSA on the Bethe structure reads

$$\frac{\partial}{\partial t} p(\sigma,t) = \sum_x (2\sigma_x - 1) \times \prod_{a=1}^{q} (1-\sigma_{x+a}) f_x(t) \times \sum_{\tilde{\sigma}_x=0,1} (1-\tilde{\sigma}_x) p(\sigma/\sigma_x \to \tilde{\sigma}_x, t), \quad (4.1)$$

where $\{x+a; a=1,\ldots,q\}$ is the set of all NN's of site x. The consequent infinite chain of evolution equations takes the form

$$\frac{\partial}{\partial t} \rho_x = f_x(t) \left\langle \bar{\sigma}_x \prod_{a=1}^{q} \bar{\sigma}_{x+a} \right\rangle, \quad (4.2a)$$

$$\frac{\partial}{\partial t} \left\langle \prod_{x\in\Omega} \bar{\sigma}_x \right\rangle = -\sum_{x\in\Omega_i} f_x(t) \left\langle \prod_{y\in\Omega} \bar{\sigma}_y \right\rangle - \sum_{x\in\partial\Omega} f_x(t) \left\langle \prod_{y\in\Omega'(x)} \bar{\sigma}_y \prod_{z\in\Omega} \bar{\sigma}_z \right\rangle, \quad (4.2b)$$

where the complete decomposability property of the Ω hierarchy onto sets of one- and q-coordinated vertices is not violated by the time operator. We notice that in every new connected cluster $\Omega\cup\Omega'(x)$ on the right-hand side of (4.2b) site x changes its previous boundary position to an internal one and becomes q coordinated.

The topological simplifications of the inverse equilibrium structure due to the presence of articulation points are of the same kind as in the previous $q=2$ case. Reducibility to a finite cluster of sites implies a local profile equation of the form [11]

$$z_x = \frac{\rho_x^*(1-\rho_x^*)^{q-1}}{\prod_{a=1}^{q} (1-\rho_x^*-\rho_{x+a}^*)} \quad (4.3)$$

and the factorization of cluster hole densities into NN pairs

$$\left\langle \prod_{x\in\Omega} \bar{\sigma}_x \right\rangle^* = \frac{\prod_{\langle x,y\rangle_\Omega} \langle \bar{\sigma}_x \bar{\sigma}_y \rangle^*}{\prod_{x\in\Omega_i} (\langle \bar{\sigma}_x \rangle^*)^{q-1}} = \frac{\prod_{\langle x,y\rangle_\Omega} (1-\rho_x^*-\rho_y^*)}{\prod_{x\in\Omega_i} (1-\rho_x^*)^{q-1}}, \quad (4.4)$$

where the products in numerators run over all NN pairs inside Ω. The elimination of a NN bond $\langle x,y \rangle$ has exactly the same effect as in the 1D case, so that

$$\langle \bar{\sigma}_y \rangle_x^* = \frac{1-\rho_x^*-\rho_y^*}{1-\rho_x^*}. \quad (4.5)$$

The subscript x indicates ensemble averaging with y being topologically decoupled from (and, consequently, statistically independent of) its NN x and all sites connected to y only by a continuous path through point x. The combination of (4.4) with (4.5) results in a useful representation of multisite hole densities,

$$\left\langle \prod_{x\in\Omega} \bar{\sigma}_x \right\rangle^* = \left\langle \prod_{x\in\Omega_i} \bar{\sigma}_x \right\rangle^* \prod_{x\in\partial\Omega} \langle \bar{\sigma}_x \rangle^*_{\Omega(x)} . \tag{4.6}$$

An ansatz for time-dependent multisite correlations $\langle \prod_{x\in\Omega}\bar{\sigma} \rangle$ is now proposed as follows:

$$\left\langle \prod_{x\in\Omega} \bar{\sigma}_x \right\rangle = \prod_{x\in\Omega_i} G_x(t) \left\langle \prod_{x\in\Omega_i} \bar{\sigma}_x \right\rangle^* \prod_{x\in\partial\Omega} \langle \bar{\sigma}_x \rangle_{\Omega(x)} , \tag{4.7a}$$

with initial conditions

$$\langle \bar{\sigma}_x \rangle_{\Omega(x)} \Big|_{t=0} = \langle \bar{\sigma}_x \rangle^*_{\Omega(t)} = \frac{1-\rho_x^* - \rho^*_{\Omega(x)}}{1-\rho^*_{\Omega(x)}} \quad (x\in\partial\Omega) \tag{4.7b}$$

implied by (4.5), (4.6). It solves exactly the infinite hierarchy (4.2) because the resulting equation of motion for the cluster boundary $\{x\in\partial\Omega\}$,

$$\frac{\partial}{\partial t}\langle \bar{\sigma}_x \rangle_{\Omega(x)} = -F_x(t) \prod_{y\in\Omega'(x)} \langle \bar{\sigma}_y \rangle_x \frac{\left\langle \prod_{z\in\Omega_i} \bar{\sigma}_x \bar{\sigma}_z \right\rangle^*}{\left\langle \prod_{z\in\Omega_i} \bar{\sigma}_z \right\rangle^*}$$

$$= -F_x(t) \langle \bar{\sigma}_x \rangle^*_{\Omega(x)} \prod_{y\in\Omega'(x)} \langle \bar{\sigma}_y \rangle_x , \tag{4.8}$$

turns out to be independent of the cluster-Ω shape.

Technical completion of the inverse program comes from the evolution equation for particle densities

$$\frac{\partial}{\partial t}\rho_x = F_x(t) \langle \bar{\sigma}_x \rangle^* \prod_{a=1}^{q} \langle \bar{\sigma}_{x+a} \rangle_x \tag{4.9}$$

and the implicit representation of $\langle \bar{\sigma}_{x+a} \rangle_x$,

$$\langle \bar{\sigma}_{x+a} \rangle_x = \langle \bar{\sigma}_{x+a} \rangle^*_x \exp\left[-\int_0^t dt' \frac{\dot{\rho}_{x+a} \langle \bar{\sigma}_{x+a} \rangle^*_x}{\langle \bar{\sigma}_{x+a} \rangle^* [\langle \bar{\sigma}_x \rangle_{x+a} \langle \bar{\sigma}_{x+a} \rangle_x]} \right] , \tag{4.10}$$

deduced from (4.8) and (4.9). Since

$$\frac{\partial}{\partial t}[\langle \bar{\sigma}_x \rangle_{x+a} \langle \bar{\sigma}_{x+a} \rangle_x] = -\frac{\langle \bar{\sigma}_x \rangle^*_{x+a}}{\langle \bar{\sigma}_x \rangle^*} \dot{\rho}_x - \frac{\langle \bar{\sigma}_{x+a} \rangle^*_x}{\langle \bar{\sigma}_{x+a} \rangle^*} \dot{\rho}_{x+a} \tag{4.11}$$

one readily finds

$$\langle \bar{\sigma}_x \rangle_{x+a} \langle \bar{\sigma}_{x+a} \rangle_x = \frac{(1-\rho_x^* - \rho_{x+a}^*)(1-\rho_x - \rho_{x+a})}{(1-\rho_x^*)(1-\rho_{x+a}^*)} , \tag{4.12}$$

where the initial conditions (4.7b) are satisfied. Thus

$$\langle \bar{\sigma}_{x+a} \rangle_x = \langle \bar{\sigma}_{x+a} \rangle^*_x \times \exp\left[-\int_0^t dt' \frac{\dot{\rho}_{x+a}(t')}{1-\rho_x(t') - \rho_{x+a}(t')} \right] \tag{4.13}$$

and the inverse profile relation follows immediately as

$$F_x(t) = \dot{\rho}_x(t) \frac{(1-\rho_x^*)^{q-1}}{\prod_{a=1}^{q} (1-\rho_x^* - \rho_{x+a}^*)} \times \exp\left[\sum_{a=1}^{q} \int_0^t dt' \frac{\dot{\rho}_{x+a}(t')}{1-\rho_x(t') - \rho_{x+a}(t')} \right] . \tag{4.14}$$

In the homogeneous subspace of initial densities, $\rho_x^* = \rho^*$, and adsorption rates, $f_x(t) = f(t)$, (4.14) integrates at once to

$$\rho(t) = \frac{1}{2} - \frac{(1-2\rho^*)}{2} \left\{ 1 + (q-2) \left[\frac{1-2\rho^*}{1-\rho^*} \right]^{q-1} \times [1-G(t)] \right\}^{2/(2-q)} \tag{4.15}$$

providing in the direct format the exact formula for the jamming coverage

$$\rho(t\to\infty) = \frac{1}{2} - \frac{(1-2\rho^*)}{2} \times \left[1 + (q-2) \left[\frac{1-2\rho^*}{1-\rho^*} \right]^{q-1} \right]^{2/(2-q)} . \tag{4.16}$$

This represents the generalization of $\rho^* = 0$ solutions [21,22].

V. CONTINUUM 1D RSA WITH EQUILIBRIUM INITIAL CONDITIONS

As a by-product of the above treatment we derive in this section the exact solution of the continuous RSA of hard-core particles, known as the random parking problem [16], on a line occupied at initial time by a fluid of equivalent hard rods in thermodynamic equilibrium. The derivation will follow the strategy of continualization [17,18] via the integer $r\to\infty$ limit of the discrete version of RSA for particles with exclusion zone up to the rth neighbor. Although the exact solution will be deducible, for $r>1$, only for an adsorption process spatially homogeneous in both the initial equilibrium state as well as the particle flux, we will start with the most general inhomogeneous case in order to show transparently how the interference of equilibrium and nonequilibrium regimes can be accomplished on the local level, and then we will restrict ourselves to the purely homogeneous case.

Let us consider a 1D lattice gas with core length r, i.e., the presence of a particle at a site excludes r contiguous sites on each side from the occupation by other particles. Topological simplification of equilibrium statistics in the inverse form is similar to that for the previously outlined $r=1$ case (Sec. III A): due to the exclusion character of the particle-particle interaction, the separation of a cluster $\{x-r,\ldots,x,\ldots,x+r\}$ with given particle densities $\{\rho^*_{x-r},\ldots,\rho^*_x,\ldots,\rho^*_{x+r}\}$ from the chain results only in the modification of activities $z_{x-r},\ldots,z_{x-1},z_{x+1},\ldots,z_{x+r}$, but does not affect z_x and the exclusion nature of two-particle interactions. From the local picture, the inverse profile equation

$$z_x=\frac{\rho^*_x\prod_{i=1}^{r}\left[1-\sum_{j=1}^{r+1}\rho^*_{x+i-j}\right]}{\prod_{i=1}^{r+1}\left[1-\sum_{j=1}^{r}\rho^*_{x+i-j}\right]}\,, \tag{5.1}$$

derived using potential distribution theory by Robledo [23], follows immediately. The evident generalization of the factorization form (3.5) of the multisite hole density $\langle\bar\sigma_x\cdots\bar\sigma_{x+h}\rangle^*$ with $h\ge r+1$ now reads

$$\langle\bar\sigma_x\cdots\bar\sigma_{x+h}\rangle^*=\frac{\prod_{y=x}^{x+h-r}\langle\bar\sigma_y\cdots\bar\sigma_{y+r}\rangle^*}{\prod_{y=x+1}^{x+h-r}\langle\bar\sigma_y\cdots\bar\sigma_{y+r-1}\rangle^*}\,. \tag{5.2}$$

The infinite chain of equations of motion for RSA,

$$\frac{\partial}{\partial t}\rho_x=f_x(t)\langle\bar\sigma_{x-r}\cdots\bar\sigma_x\cdots\bar\sigma_{x+r}\rangle\,, \tag{5.3a}$$

$$\begin{aligned}\frac{\partial}{\partial t}\langle\bar\sigma_x\cdots\bar\sigma_{x+h}\rangle=&-\sum_{y=x}^{x+r-1}f_y(t)\langle\bar\sigma_{y-r}\cdots\bar\sigma_{x+h}\rangle\\&-\sum_{y=x+r}^{x+h-r}f_y(t)\langle\bar\sigma_x\cdots\bar\sigma_{x+h}\rangle\\&-\sum_{y=x+h-r+1}^{x+h}f_y(t)\langle\bar\sigma_x\cdots\bar\sigma_{y+r}\rangle\\&(h\ge 2r)\,,\end{aligned} \tag{5.3b}$$

with equilibrium initial conditions $\langle\bar\sigma_x\cdots\bar\sigma_{x+h}\rangle|_{t=0}=\langle\bar\sigma_x\cdots\bar\sigma_{x+h}\rangle^*$ is then solved by the ansatz

$$\begin{aligned}\langle\bar\sigma_x\cdots\bar\sigma_{x+h}\rangle=&\prod_{y=x+r}^{x+h-r}G_y(t)\langle\bar\sigma_{x+1}\cdots\bar\sigma_{x+h-1}\rangle^*\\&\times\langle\bar\sigma_x\cdots\bar\sigma_{x+r-1}\rangle_L\\&\times\langle\bar\sigma_{x+h-r+1}\cdots\bar\sigma_{x+h}\rangle_R\end{aligned} \tag{5.4}$$

providing an h-independent differential recursive scheme for the L quantities

$$\frac{\partial}{\partial t}\langle\bar\sigma_x\cdots\bar\sigma_{x+r-1}\rangle_L=-\sum_{y=x}^{x+r-1}f_y(t)\prod_{z=y}^{x+r-1}G_z(t)\frac{\langle\bar\sigma_{z-r+1}\cdots\bar\sigma_{z+1}\rangle^*}{\langle\bar\sigma_{z-r+2}\cdots\bar\sigma_{z+1}\rangle^*}\langle\bar\sigma_{y-r}\cdots\bar\sigma_{y-1}\rangle_L\,, \tag{5.5a}$$

with initial conditions

$$\langle\bar\sigma_x\cdots\bar\sigma_{x+r-1}\rangle_L\Big|_{t=0}=\frac{\langle\bar\sigma_x\cdots\bar\sigma_{x+r}\rangle^*}{\langle\bar\sigma_{x+1}\cdots\bar\sigma_{x+r}\rangle^*}\,, \tag{5.5b}$$

and an analogous recursion for the R quantities. Together with the evolution equations for particle densities

$$\begin{aligned}\frac{\partial}{\partial t}\rho_x(t)=&F_x(t)\langle\bar\sigma_{x-r}\cdots\bar\sigma_{x-1}\rangle_L\\&\times\langle\bar\sigma_{x-r+1}\cdots\bar\sigma_{x+r-1}\rangle^*\\&\times\langle\bar\sigma_{x+1}\cdots\bar\sigma_{x+r}\rangle_R\,,\end{aligned} \tag{5.6}$$

the recursions represent a complete basis for the profile problem.

Equation (5.6) can be trivially used to eliminate the particle flux from (5.5a) and its R analog, providing in this way, at least in principle, the ρ dependence of auxiliary L and R quantities. Returning to (5.6), the inverse profile relation would then be available. In practice, we succeeded only in the already solved $r=1$ example, while for $r>1$ the ρ dependence of L and R quantities is nonlocal. The situation becomes simpler in the spatially homogeneous case $\rho^*_x=\rho^*$, $f_x(t)=f$ (the time dependence of the particle flux is irrelevant) for which Eqs. (5.6) and (5.5a) take the respective forms

$$\frac{d}{dt}\rho=f\exp(-ft)\frac{[1-(r+1)\rho^*]^{r-1}}{(1-r\rho^*)^{r-2}}s^2\,, \tag{5.7a}$$

$$\frac{1}{s}\frac{d}{dt}s=-f\sum_{i=1}^{r}\left[\frac{1-(r+1)\rho^*}{1-r\rho^*}\right]^i\exp(-ift)\,. \tag{5.7b}$$

The initial conditions for the particle density ρ and the auxiliary L or R variable $s=\langle\bar\sigma_x\cdots\bar\sigma_{x+r-1}\rangle_{L(R)}$ (5.5b) read

$$\rho|_{t=0}=\rho^*,\quad s|_{t=0}=\frac{1-(r+1)\rho^*}{1-r\rho^*}\,. \tag{5.8}$$

The system of Eqs. (5.7) and (5.8) is solvable using simple algebra and we only write down the final result for the time evolution of the particle density in a form convenient for the continualization:

$$(r+1)\rho(t)=(r+1)\rho^*+\frac{[1-(r+1)\rho^*]^r}{(1-r\rho^*)^{r-1}}\int_{(r+1)(1-c)}^{(r+1)(1-ce^{-ft})}du\,\exp\left\{-2\int_{(r+1)(1-c)}^{u}\frac{dv}{v}\left[1-\left[1-\frac{v}{r+1}\right]^r\right]\right\}, \tag{5.9}$$

with $c=[1-(r+1)\rho^*]/(1-r\rho^*)$. Since the maximum density for a given r is equal to $1/(r+1)$, the expressions $\rho_c^*=(r+1)\rho^*$ and $\rho_c(t)=(r+1)\rho(t)$ represent, respectively, the equilibrium and nonequilibrium coverages. In the limit $r\to\infty$ with the appropriately scaled particle flux $f=1/(r+1)$, we finally get

$$\rho_c(t)=\rho_c^*+(1-\rho_c^*)\exp\left[-\frac{\rho_c^*}{1-\rho_c^*}\right]\int_0^t du\,\exp\left\{-2\int_0^u dv\frac{1}{v+\rho_c^*/(1-\rho_c^*)}\left[1-\exp-\left[v+\frac{\rho_c^*}{1-\rho_c^*}\right]\right]\right\}, \tag{5.10}$$

reproducing the $\rho_c^*=0$ solution [16–18].

VI. CONCLUDING REMARKS

In spite of the simplifications having their origin in the simple connectivity of the Bethe lattice, the existence of an exact inverse formula (4.14) for the most general version of RSA, inhomogeneous in renormalized particle flux $\{F_x(t)\}$ as well as in initial site densities $\{\rho_x^*\}$, is surprising. It is the product of a close relationship between equilibrium and nonequilibrium inverse formats, realized coherently at every level of the correlation hierarchy through the factorization property of equilibrium multisite correlations, (3.5) and then (4.4). The aspect of nonuniformity as an inherent characteristic of real physical systems is relevant even in the idealized case of homogeneous external conditions, i.e., constant activity, $z_x=z$, and uniform (renormalized) adsorption rates, $F_x(t)=F(t)$. It is well known [11] that, at equilibrium, the pair of inverse profile relations for homogeneous sublattice occupation,

$$z=\frac{\rho_A^*(1-\rho_A^*)^{q-1}}{(1-\rho_A^*-\rho_B^*)^q},\quad z=\frac{\rho_B^*(1-\rho_B^*)^{q-1}}{(1-\rho_A^*-\rho_B^*)^q} \tag{6.1}$$

following from (4.3), provides, for $q>2$ and activity z exceeding the critical value

$$z_c=\frac{(q-1)^{q-1}}{(q-2)^q},\quad \rho_c^*=\frac{1}{q} \tag{6.2}$$

as its only stable solutions, those with different sublattice densities, $\rho_A^*\neq\rho_B^*$. E.g., for $q=3$, with $z_c=4$, ρ_A^* and ρ_B^* are available in the explicit form

$$\rho_A^*=\frac{(z-2)+\sqrt{z(z-4)}}{2(z-1)},\quad \rho_B^*=\frac{(z-2)-\sqrt{z(z-4)}}{2(z-1)}, \tag{6.3}$$

and their plots versus $1/z$ are represented graphically together with the symmetric solution $\rho_A^*=\rho_B^*$ (stable for $z<4$) by dashed lines in Fig. 1. The corresponding asymptotic values of site coverages under uniform adsorption rates (solid lines) are obtained for the fluid phase region $(z<z_c)$ from asymptotic formula (4.16), while for the crystal phase region $(z>z_c)$ by numerical calculations with the aid of (4.14) adapted to alternating A and B sublattice positions.

The application of the homogeneous asymptotic formula (4.16) to the whole fluid region might be, in principle, incorrect. In the neighborhood of the critical point there could take place, at a specific time or at sufficiently high density, a nonequilibrium phase transition due to an instability leading to the spontaneous sublattice bifurcation. Such a crystallization has been observed for time evolution of the system governed by reversible Glauber dynamics [24] possessing detailed balance, but in that case the bifurcation is inevitable for the attainment of thermal equilibrium at asymptotic time. In what follows we will show that within our irreversible process a fluid phase cannot be transformed into a crystal phase under homogeneous adsorption conditions. The proof starts from the evolution Eq. (4.9) involving the possibility of the sublattice alternation:

$$\frac{d}{dt}\rho_A=F_A(t)\langle\bar\sigma_A\rangle^*(\langle\bar\sigma_B\rangle_A)^q, \tag{6.4a}$$

$$\frac{d}{dt}\rho_B=F_B(t)\langle\bar\sigma_B\rangle^*(\langle\bar\sigma_A\rangle_B)^q. \tag{6.4b}$$

In (4.8), the auxiliary quantities $\langle\bar\sigma_A\rangle_B$, $\langle\bar\sigma_B\rangle_A$ satisfy

$$\frac{d}{dt}\langle\bar\sigma_A\rangle_B=-F_A(t)\langle\bar\sigma_A\rangle_B^*(\langle\bar\sigma_B\rangle_A)^{q-1}, \tag{6.5a}$$

$$\frac{d}{dt}\langle\bar\sigma_B\rangle_A=-F_B(t)\langle\bar\sigma_B\rangle_A^*(\langle\bar\sigma_A\rangle_B)^{q-1}. \tag{6.5b}$$

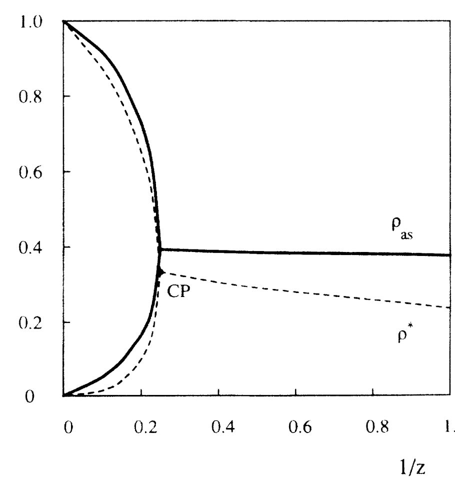

FIG. 1. RSA on the Bethe lattice of coordination number $q=3$: dashed lines represent the dependence of the particle density on the inverse activity $1/z$ at equilibrium (CP denotes the critical point associated with the sublattice symmetry breaking); solid lines represent the corresponding asymptotic values of densities under a homogeneous particle adsorption.

Under homogeneous initial conditions $\langle \bar{\sigma}_A \rangle_B^* = \langle \bar{\sigma}_B \rangle_A^* = (1-2\rho^*)/(1-\rho^*)$ and renormalized adsorption rates $F_A(t) = F_B(t) = F(t)$, the multiplication of Eq. (6.5a) by factor $(\langle \bar{\sigma}_A \rangle_B)^{q-1}$, of Eq. (6.5b) by $(\langle \bar{\sigma}_B \rangle_A)^{q-1}$,and the consequent subtraction of these equations results in

$$\frac{1}{q}\frac{d}{dt}[(\langle \bar{\sigma}_A \rangle_B)^q - (\langle \bar{\sigma}_B \rangle_A)^q] = 0 . \tag{6.6}$$

With homogeneity of the initial regime, we thus have $(\langle \bar{\sigma}_A \rangle_B)^q = (\langle \bar{\sigma}_B \rangle_A)^q$ at arbitrary time. Putting this equality into (6.4a) and (6.4b) with assumed $\langle \bar{\sigma}_A \rangle^* = \langle \bar{\sigma}_B \rangle^* = 1-\rho^*$, we readily find the unique solution $\rho_A(t) = \rho_B(t)$. The lack of a nonequilibrium phase transition is not surprising, and is expected to be a general property of systems whose time evolution is governed by a dynamics of irreversible type.

The locality of the inverse format for simply connected lattices is an important reference when we pass to structures with a certain degree of nonlocality. In the equilibrium statistics of inhomogeneous lattice models, this program has already been started and is successful in overcoming topological nontrivialities of closed circuit [12], and superbond multiconnection [13], type. The nonlocality is reflected through global collective modes of topological or interaction nature permitting one to construct, in a variational way, the exact density functionals which mimic associated lattice structures. The superposition of equilibrium and nonequilibrium formats is a more complicated topic and the presence of a kind of global variable is questionable. If they exist, the present approach could reveal them on the basis of the close relationship between the equilibrium and nonequilibrium forms of multisite correlations. For closed circuits, this procedure has already been started, but in this case the evolution of a cluster boundary is far from transparent. This is the motivation for future study.

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[1] P. J. Flory, J. Am. Chem. Soc. **61**, 1518 (1939).
[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] J. W. Evans, Rev. Mod. Phys. **65**, 1298 (1993).
[5] G. Tarjus, P. Schaaf, and J. Talbot, J. Stat. Phys. **63**, 167 (1991).
[6] J. A. Given, Phys. Rev. A **45**, 816 (1992).
[7] J. A. Given and G. R. Stell, in *Condensed Matter Theories,* edited by L. Blum and F. Malik (Plenum, New York, 1993), Vol. 8, p. 395.
[8] J. K. Percus, J. Stat. Phys. **71**, 1201 (1993).
[9] J. K. Percus, J. Stat. Phys. **55**, 1263 (1989).
[10] L. Šamaj, J. Phys. (Paris) **50**, 273 (1989).
[11] A. Robledo and C. Varea, J. Stat. Phys. **63**, 1163 (1991).
[12] J. K. Percus and M. Q. Zhang, Phys. Rev. B **38**, 11 737 (1988).
[13] L. Šamaj and J. K. Percus, J. Stat. Phys. **73**, 235 (1993).
[14] G. Tarjus, J. Talbot, and P. Schaaf, J. Phys. A **23**, 837 (1990).
[15] N. O. Wolf, J. W. Evans, and D. K. Hoffman, J. Math. Phys. **25**, 2519 (1984).
[16] A. Rényi, Publ. Math. Inst. Hung. Acad. Sci. **3**, 109 (1958).
[17] J. J. González, P. C. Hemmer, and J. C. Hoye, Chem. Phys. **3**, 228 (1974).
[18] P. C. Hemmer, J. Stat. Phys. **57**, 865 (1989).
[19] R. J. Glauber, J. Math. Phys. **4**, 294 (1963).
[20] M. Scheucher and H. Spohn, J. Stat. Phys. **53**, 279 (1988).
[21] J. W. Evans, Phys. Rev. Lett. **62**, 2642 (1989).
[22] Y. Fan and J. K. Percus, Phys. Rev. A **44**, 5099 (1991).
[23] A. Robledo, J. Chem. Phys. **72**, 170 (1979).
[24] L. Šamaj and J. K. Percus, J. Stat. Phys. (to be published).