Exactly solvable irreversible processes on Bethe lattices

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Abstract
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Exactly solvable irreversible processes on Bethe lattices

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We consider the kinetics of processes where the sites of a Bethe lattice are filled irreversibly and, in general, cooperatively by monomers, dimers, or polyatomics. For nearest neighbor and sometimes more general cooperative effects (including random filling as a special case), we show that the infinite hierarchy of rate equations for probabilities of empty subconfigurations can be exactly truncated and solved using a shielding property of empty sites. We indicate, in certain cases, a connection between these Bethe lattice solutions and certain approximate truncation solutions for corresponding processes on "physical" 2-D and 3-D lattices with the same coordination number.

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I. INTRODUCTION

We consider here processes where the sites of a Bethe lattice are irreversibly filled, either randomly or cooperatively, by monomers, dimers, or polyatomics. In the language of graph theory, a Bethe lattice is an infinite, regular (Cayley) tree, i.e., a connected graph without circuits.1 We must also specify the coordination number \( c \) (\( c = 2 \) corresponds to a linear lattice). Thus for \( c > 2 \), Bethe lattices incorporate the connectivity features of a 1-D lattice with some of the combinatorial properties of higher-dimensional lattices. The role of Bethe lattices in the theory of disordered systems, e.g., in Ising model or percolation theory calculations, is well known as providing an artificial mathematical model for which some theoretical techniques give exact results.2 We will show here that the same is true for certain dynamic irreversible processes.

For finite range cooperative effects or random processes, one can immediately write down an infinite hierarchy of rate equations for the probabilities of various subconfigurations3 (which is trivial only for random monomer filling). Using conservation of probability, one can instead deal with the subhierarchy for the probabilities of subconfigurations where all specified sites are empty.4 We show here that it is possible to solve this hierarchy (in closed form) by exact truncation for a variety of processes.

The basic tool used to obtain solutions is the following shielding property of empty sites5: a connected cluster of sites specified empty separates the lattice into several disconnected parts; suppose the cluster is sufficiently large that an event occurring on the lattice is not simultaneously affected by the state of sites in a particular pair of these disconnected parts; then sites in either of these two parts are completely shielded from the effect of those in the other. Just as in the preceding paper,6 those processes amenable to exact solution via truncation are characterized by the existence of a minimal closed subhierarchy for the probabilities of connected clusters of empty sites.

In Sec. II, we consider in detail the exact solution for random dimer filling of a Bethe lattice with arbitrary coordination number \( c \) and, in Sec. III, monomer filling with nearest neighbor (NN) cooperative effects for \( c = 3 \) (the solution for general \( c \) is also discussed here). Explicit examples of the shielding condition are given and its role in exact hierarchy truncation is elucidated. The connection between the solutions for these processes and certain approximate solutions of corresponding processes on "physical" 2-D and 3-D lattices with the same coordination number is also described. In Sec. IV, we discuss more general exactly solvable processes and also describe those not amenable to exact solution. Finally, in Sec. V, we give some concluding remarks, in particular, indicating the utility of exact Bethe lattice solutions as providing a basis for resummation of formal density expansions for solutions of corresponding processes on physical 2-D and 3-D lattices.

The following notation is used here. Empty (filled) sites are denoted by "\( o \)" ("\( a \)"), Let \( f_\sigma \) denote the probability of a subconfiguration \( \sigma \) of sites specified filled or empty, and \( q_\sigma = f_\sigma / f_\emptyset \), the conditional probability of \( \sigma \) given \( \emptyset \) (for typographic convenience, empty conditioning sites \( \emptyset \) are denoted \( \emptyset \) here). All processes considered have translational invariance (as well as other natural symmetries) so \( f_\emptyset \)'s and \( q_\emptyset \)'s are position independent.

II. RANDOM DIMER FILLING

Here we consider the random dimer filling of NN pairs of sites on a Bethe lattice with coordination number \( c \) (arbitrary, but fixed). The rate of decrease of the probability of some subconfiguration of sites specified empty consists simply of a sum of terms corresponding to all possible ways a dimer can land completely within or partly overlapping this configuration (cf. Ref. 5). To provide some examples we need the following notation. Let \( o_n \) denote a connected string of \( n \) empty sites, e.g., \( o, o_o, oo, ooo, ... \), and \( o_oo \) denote a string of \( i + j \) empty sites where, in addition, one of the remaining \( c - 2 \) NN of a non-end site of the string is also specified empty. Then if \( \tau \) denotes the single filling rate, one has

\[
-\tau^{-1} \frac{df_\emptyset}{dt} = cf_{oo}, \tag{2.1a}
\]

\[
-\tau^{-1} \frac{df_oo}{dt} = f_{oo} + 2(c - 1)f_{oo}, \tag{2.1b}
\]

\[
-\tau^{-1} \frac{df_{oo}}{dt} = (n - 1)f_{oo} + 2(c - 1)f_{oo_o} + (c - 2) \sum_{i+j=n-1} f_{o_o, o}, \tag{2.1c}
\]

...
The key to exact solution of this problem is the shielding property of empty sites (as stated in the introduction) which is embedded in (2.1) in a rather subtle way. Since each site of a Bethe lattice is an articulation point (i.e., separates the lattice into disconnected parts), for random dimer filling, one has that a single site specified empty shields. Thus, for example,

$$q_m = q_{1000} = f_{000},$$

(2.2)

The shielding property can most easily be proved by writing down the q-hierarchy and observing self-consistency (assuming compatible initial conditions). Because of notational difficulties, we only give the following examples of the general q-equations \( \frac{d}{dt} \ln q_{1000} = S(\sigma + \alpha') - S(\sigma) \), where \( S(\sigma') \) is (2.3a)

\[
- \frac{1}{\tau} \frac{d}{dt} \ln q_{1000} = 1 + (c - 1)(q_{1000} - q_{000}),
\]

(2.3a)

(2.3b)

\[
\sum_{n=0}^{\infty} (-1)^n \binom{c}{n} q^n f_0 = (1 - q)f_0.
\]

The expressions (2.6a) and (2.7a) for \( c = 2 \) are of course exact results for dimer filling on a linear lattice. In particular, (2.7a) was first obtained by Flory in 1939 (see Ref. 6). For \( c > 2 \), some of these expressions have appeared previously in the context of "first shell truncation approximations" for random dimer filling on higher-dimensional lattices, in particular, \( c = 3 \) for the 2-D hexagonal and \( c = 4 \) for the 2-D square lattice. For \( c = 6 \), these correspond to the "first shell truncation approximation" on the 3-D cubic but not the 2-D triangular lattice.

### III. MONOMER FILLING WITH NN COOPERATIVE EFFECTS

We first illustrate in detail this process for a Bethe lattice with coordination number 3. Here the rates for filling a site with 0,1,2,3 occupied NN are denoted by \( \tau_{a, a} = \tau_{a, o}, \tau_{o, o}, \tau_{o, a} \), respectively. The rate equations for the probability of a single empty site \( f_o \), of an adjacent pair of empty sites \( f_{oo} \) of a string of three empty sites \( f_{ooo} \) of a site and all three of its NN empty \( f_{oo} = f_{oo} \) are

\[
- \frac{d}{dt} f_o = \tau_{a, o} f_o + 3 \tau_{o, o} f_{oo} + 3 \tau_{o, o} f_{oo} + \tau_{o, o} f_{oo} + \tau_{o, o} f_{oo},
\]

(3.1a)

\[
- \frac{d}{dt} f_{oo} = 2 \left[ \tau_{a, o} f_{oo} + 2 \tau_{o, o} f_{oo} + \tau_{a, a} f_{oo} \right] \]

(3.1b)

\[
- \frac{d}{dt} f_{ooo} = - \frac{d}{dt} f_{oo} = \tau_{a, a} f_{ooo} + \tau_{a, o} f_{oo} + \tau_{a, o} f_{oo} + 2 \left[ \tau_{o, o} f_{oo} + 2 \tau_{o, o} f_{oo} + \tau_{a, a} f_{oo} \right] \]

(3.1c)
\[
- \frac{d}{dt} f_{oo} = \tau_{oo} f_{oo} + 3 \left[ \tau_{oo} f_{oo} + 2 \tau_{a} f_{oo} + \tau_{a} f_{aa} \right] \\
= \ldots , \quad (3.1d)
\]

Examples of shielding used here to obtain an exact truncation of this hierarchy are shown in Fig. 1. These include such identities as
\[
q_{aaao} = q_{aaao} = q_{aaao} = q_{aaao} = \ldots , \quad (3.2a)
\]
and thus also imply such quantities as
\[
q_{aaao} = q_{aaao} = q_{aaao} = q_{aaao} = q_{aaao} = q_{aaao} = q_{aaao} = q_{aaao} = \ldots , \quad (3.3)
\]
are equal. Further identities such as \( q_{aaao} = q_{aaao} = q_{aaao} = q_{aaao} = \ldots \), all compatible with the statement of shielding given in the Introduction, then follow from conservation of probability.

Again these properties are imbedded in the hierarchy equations in a rather subtle way. The simplest demonstration of the results required here comes from writing down the closed \( q \)-subhierarchy for conditional probabilities of a single empty site \( q \)-given various adjacent connected clusters of sites are empty (factorization [cf. (3.3)] and conservation of probability must be used to obtain this reduced form). When suitably arranged, consistency with the shielding condition becomes obvious [cf. (2.3) and Ref. 4] assuming compatible initial conditions. This procedure is straightforward though notationally complex. Thus instead of presenting these general \( q \)-equations, we consider in detail only the particularly important cases for \( q_{aaao} \) and \( q_{aaao} \). These equations are first simply obtained from (3.1b)–(3.1d) and then closed applying shielding and factorization [cf. (3.2) and (3.3)] as shown below:

\[
\frac{d}{dt} \ln q_{aaao} = \left( \frac{d}{dt} \ln f_{oo} \right)_{oo} - \left( \frac{d}{dt} \ln f_{oo} \right)_{oo} \\
= - \tau_{oo} q_{aaao} - \tau_{aaao} q_{aaao} - 2 \tau_{a} q_{aaao} - q_{aaao} \\
= - 4 \tau_{a} q_{aaao} - q_{aaao} - 2 \tau_{a} q_{aaao} - q_{aaao} \\
= - \tau_{a} q_{aaao} - q_{aaao} \\
= - \tau_{a} \left( \tau_{a} q_{aaao} + \tau_{a} q_{aaao} \right) \\
= \ldots , \quad (3.4a)
\]

Equations (3.1a), (3.1b), and (3.4), together with the identities \( f_{oo} = f_{oo} q_{aaao} \), \( f_{oo} = f_{oo} q_{aaao} \), yield an exact closed set of equations for \( f_{oo} \), \( q_{aaao} \), and \( q_{aaao} \). From these quantities, together with the shielding condition, we can readily calculate, e.g., the probability for any empty, connected cluster, e.g., \( f_{oo} = \left( q_{aaao} \right)^{2} \left( q_{aaao} \right)^{2} f_{oo} \).

We now analyze in detail the filling of an initially empty lattice for various choices of rates. All numerical results presented come from simultaneous integration of the closed set of equations described above [using a variable-order Adams predictor-corrector (Gear's) method]. First we consider the case where each filled NN changes the rate from \( \tau_{o} \) by a factor of \( \alpha \), i.e., \( \tau_{a} = \tau_{a} \alpha^{2} \). The behavior \( f_{oo} \) as a function of the coverage \( \theta = 1 - f_{o} \) is shown in Fig. 2 for various \( \alpha \).

(a) Highly autoinhibitory rates \( \alpha < 1 \): Here the process effectively takes place in four stages, namely, filling of sites with \( m = 0,1,2,3 \) occupied NN (consecutively). In the \( m \)th stage,

\[
\frac{d}{d \theta} f_{oo} = - \frac{3}{2} (3 - m) , \quad (3.5)
\]

since \( 3 - m \) NN pairs are destroyed by each monomer adsorbing and there are \( 3 \) as many sites as there are (adjacent) pairs of sites (cf. Ref. 3). We denote the \( \theta \) values of the ends of these stages by \( \theta^{*} \). Then \( f_{oo} \rightarrow 0, f_{oo} \rightarrow 0.2500, \) and \( f_{oo} \rightarrow 0.0767 \) as \( \theta \rightarrow \theta^{*} = 0.3750 \) [these are saturation values for filling with NN blocking and are determined (numerically) after setting \( \alpha = 0 \); \( f_{oo} \rightarrow 0 \) and \( f_{oo} \rightarrow 0.0732 \) as \( \theta \rightarrow \theta^{*} = 0.5076 \) [numerical determination of these values actually comes from a modified choice of rates (ii), described below, when \( \alpha = 0 \); \( f_{oo} \rightarrow 0 \) as \( \theta \rightarrow \theta^{*} = 0.6174 \) and \( \theta^{*} = 1 \)].

(b) Random filling \( \alpha = 1 \): Here, trivially, \( f_{oo} \rightarrow f_{oo}^{3}, f_{oo} \rightarrow f_{oo}^{3} \), etc.

(c) Highly autocatalytic rates \( \alpha > 1 \): Here the process involves competition between filling of a small fraction of sites with empty NN (island birth) and the rapid formation of
contiguous clusters of filled sites around these (island growth). We now consider the $\alpha \to \infty$ limit which we show cannot be thought of as a single island growing. For this were the case, except for the nucleating atom, every later atom adsorbed would destroy two $\omega\omega$ pairs so $(d/d\theta)\varphi_\infty = -\frac{3}{4}\times 2 = -\frac{3}{2}$. This is clearly not correct for all $\theta$ since $\varphi_\infty \to 0$ only when $\theta \to 1$. However, we do anticipate that $\lim_{\alpha \to \infty} (d/d\theta)\varphi_\infty |_{\omega = 0} = -\frac{3}{2}$. Deviation of $(d/d\theta)\varphi_\infty$ from $-\frac{3}{2}$ for $\theta \neq 0$ (see Fig. 2) must be due to infinitely large islands merging. In contrast, the $\alpha \to \infty$ behavior of $f_\infty$ for corresponding processes on 1-D and “physical” 2-D or 3-D lattices can be determined by regarding these as occurring by a single island growing.

Next we consider the modified choices for the rates $\tau_\omega^\alpha$: $\tau_{\omega\omega}^\alpha = T \omega^\alpha \tau_{\omega\omega}^\omega$, of (i) $1:0:0:0$, (ii) $1:0:0:0$, and (ii)' $1:0:0:0$. Here we focus our attention on the saturation coverage $\theta^\ast(\alpha) < 1$ for finite $\alpha$ (see Fig. 3). Of course $\lim_{\alpha \to \infty} \theta^\ast(\alpha) = \theta^\ast$ for (i) and $\theta_i^\ast$ for (ii) where $\theta_i^\ast$, defined above, are associated with filling in stages. For (ii) and (ii)', when $\alpha = 1$, filling occurs randomly except that it is blocked when all $\omega$ are occupied. It has been shown previously that the saturation coverage for this sort of “almost random” filling at an infinite, uniform lattice with coordination number $c$ is given by $c/(c + 1)$, i.e., $\theta^\ast(1) = \frac{1}{3}$ for (ii) and (ii)' (for $\theta_i^\ast$). The $\alpha \to \infty$ limit for (ii) and (ii)' provides an unequivocal demonstration of the anomalous feature of irreversible island growth on Bethe lattices described above, namely, that the highly autocatalytic/clustering limit cannot be thought of as a single island growing. From Fig. 3 we see that $\lim_{\alpha \to \infty} \theta^\ast(\alpha) < 1$ for (i) and (ii). Our numerical verification could be supplemented by a detailed asymptotic analysis of the appropriate solutions of (3.1) and (3.2) of Ref. 3. The deficit from unity is associated with imperfect filling at the boundaries between coalesced infinite contiguous islands. The fact that these boundaries must be associated with a finite fraction of the lattice sites is not that surprising when one realizes that, for a single island on this Bethe lattice containing all atoms $<N\omega$ lattice vectors from the nucleating atom, $\frac{1}{2}(1 - 2^{1-N/3})$ the atoms are boundary atoms.

One final point of significance pertaining to this process is its connection with approximate solution for monomer filling, with $\omega\omega$ cooperative effects, at the sites of a 2-D hexagonal lattice (also having coordination number 3). In previous work we have developed schemes to approximately truncate the infinite hierarchies of rate equations for such processes. These deal with conditional probabilities involving only empty sites (and a single conditioned $\omega$-site) and attempt to exploit a shielding propensity of empty (cf. filled) sites. Two such schemes both start with the empty $q$-hierarchy and either (i) factorize $q$’s involving several $\omega$-sites in terms of those with a single $\omega$-site then truncate $\phi$-sites further than $n$ lattice vectors from the $\omega$-site ($FT.n$), or (ii) truncate $\phi$-sites further than $n$ lattice vectors from any $\omega$-site then factorize ($T^\ast.n$). Reasonable results are expected for $n \geq 2$ which avoids explicit truncation of the $f_\infty$ equation.

It is easy to show that both the $FT.2$ and $T.2F$ equations for the above mentioned monomer filling of a hexagonal lattice are identical to (3.1a), (3.1b), and (3.4) for $f_\infty = q_\omega f_\infty$ (or $q_\omega$, $q_{\omega\omega}$, $q_{\omega\omega\omega}$) and, consequently, we are guaranteed “well-behaved” solutions of these. We thus anticipate that the above Bethe lattice solutions bear some similarity to the corresponding exact quantities for the hexagonal lattice. It is however also clear that the $FT.2$ and $T.2F$ solutions have “lost knowledge” of the closed loops in the hexagonal lattice. Now consider the case where, again, each filled $\omega$ changes the rate from $\tau_{\omega\omega}$ by a factor of $\alpha$. As $\alpha \to 0$, filling on the hexagonal lattice also occurs in four stages and (3.5) holds (cf. Ref. 3). No doubt, the ends of these stages will vary from the Bethe lattice values given above. One can think of the $\alpha \to \infty$ limit as a single contiguous island growing and since, at nonzero coverages, an infinitesimal fraction of the lattice sites is not that “physical” 2-D or 3-D, having coordination number 3). In previous work we have developed schemes to approximately truncate the infinite hierarchies of rate equations for such processes. These deal with conditional probabilities involving only empty sites (and a single conditioned $\omega$-site) and attempt to exploit a shielding propensity of empty (cf. filled) sites. Two such schemes both start with the empty $q$-hierarchy and either (i) factorize $q$’s involving several $\omega$-sites in terms of those with a single $\omega$-site then truncate $\phi$-sites further than $n$ lattice vectors from the $\omega$-site ($FT.n$), or (ii) truncate $\phi$-sites further than $n$ lattice vectors from any $\omega$-site then factorize ($T^\ast.n$). Reasonable results are expected for $n \geq 2$ which avoids explicit truncation of the $f_\infty$ equation.

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We can also consider a choice of rates analogous to (i) and (ii) described above for the Bethe lattice. The qualitative behavior of the saturation coverage $\theta^\ast(\alpha)$ for these cases is deceptively similar to the Bethe lattice behavior. In particular $\lim_{\alpha \to \infty} \theta^\ast(\alpha) < 1$, but this corresponds to the coverage of a single infinitely large noncontiguous island [which contains isolated empty sites for (i) and (ii) and also isolated empty $\omega$-site pairs for (ii)].

Finally, in this section, we consider the extension of the above results to monomer filling with $\omega\omega$ cooperative effects on a Bethe lattice with arbitrary coordination number $c$. A detailed treatment would require the development of an
elaborate notational scheme to describe subconfigurations on such a lattice. However since the procedure leading to exact solution parallels the case described above, we just sketch the steps. The input to the hierarchical equations here is a set of rates \( \tau_i, i = 0, 1, \ldots, c \), for filling a site with exactly \( i \) already filled NN. As for \( c = 3 \), one starts by straightforwardly writing down rate equations for \( f_0, f_1, f_2, \ldots \) and using shielding to truncate these, the latter quantities are obtainable for NN, but not longer-range, cooperative effects, unless they incorporate a suitable range of blocking. It is quite straightforward to extend the analysis to treat adsorption of different types of monomers, e.g., in the case of NN cooperative effects, also obtain exact solutions. The analogous 1-D calculations can be found in Ref. 11.

We note that the preceding discussions of exact solutions for certain processes have concentrated on obtaining probabilities of empty, connected clusters of sites. We cannot obtain probabilities of, e.g., filled, connected clusters or two-point correlations from these. However by adding rate equations for suitable disconnected empty configurations and using shielding to truncate these, the latter quantities can also be calculated exactly. Reference 10 illustrates the analogous procedures for the 1-D monomer filling process with NN cooperative effects. Finally we remark that one could also continue to examine competitive irreversible adsorption of different types of monomers "a," "b,"..., and, e.g., in the case of NN cooperative effects, also obtain exact solutions. The analogous 1-D calculations can be found in Ref. 11.

V. DISCUSSION

Here we have shown how to obtain exact solutions via hierarchy truncation for a variety of nontrivial irreversible, random and cooperative, monomer, dimer, and polyatomic filling processes on Bethe lattices. These, being exact solutions, have intrinsic interest in enhancing our understanding of the kinetic behavior of irreversible processes on lattices. In some cases we have commented on the agreement between Bethe lattices solutions and certain natural low-order approximate truncation solutions for corresponding processes on 2-D and 3-D "physical" lattices with the same coordination number. Roughly speaking, this will be the case if the shortest closed loops on the physical lattice are sufficiently

![FIG. 4. Monomer filling with NN cooperative effects on a Bethe lattice with coordination number \( c \); the \( c + 1 \) empty \( q \)'s satisfying a closed set of equations (after applying shielding) are shown.](image)

![FIG. 5. Dimer filling with NN cooperative effects of a Bethe lattice with coordination number \( c = 3 \); \( q \)'s satisfying a closed set of equations (after applying shielding) here \( \ldots \) indicate that the \( q \)'s are independent of these conditioning sites.](image)
large compared to the size of the polyatomic adsorbing, the cooperative range, and thus the (low-order) truncation range. Then these low-order truncations “cannot tell” that the loops close. This agreement was observed, for example, with random dimer filling on lattices with closed loops of length $>3$ (so excluding the triangular lattice) for one lattice vector truncations; and for monomer filling with NN cooperative effects on a hexagonal, but not on a square or triangular lattice, for two lattice vector truncations.

For cases where agreement exists, this suggests some similarity between the Bethe lattice and exact “physical” lattice processes at least with regard to the probabilities of corresponding small configurations (not containing closed loops). We can always obtain formal “density” expansions for the $f$’s for physical lattice processes no matter how complex the cooperative effects. The above observation suggests the following simple resummation procedure for expansion of $f$’s for small configurations. First we write the physical lattice probability $f_\text{phys}$ as the sum of the Bethe lattice one, $f_\text{Bethe}$, and a residual power series density expansion. Then coefficients in the residual expansion can be obtained from a knowledge of $f_\text{Bethe}$ and the density expansion of $f_\text{phys}$. This procedure has proved quite successful for some random dimer filling processes.

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