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Concentration for one and two-species one-dimensional reaction–diffusion systems

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Abstract. We look for similarity transformations which yield mappings between different one-dimensional reaction–diffusion processes. In this way results obtained for special systems can be generalized to equivalent reaction–diffusion models. The coagulation ($A + A \to A$) or the annihilation ($A + A \to \emptyset$) models can be mapped onto systems in which both processes are allowed. With the help of the coagulation–decoagulation model results for some death–decoagulation and annihilation–creation systems are given. We also find a reaction–diffusion system which is equivalent to the two-species annihilation model ($A + B \to \emptyset$).

Besides we present numerical results of Monte Carlo simulations. An accurate description of the effects of the reaction rates on the concentration in one-species diffusion–annihilation model is made. The asymptotic behaviour of the concentration in the two-species annihilation system ($A + B \to \emptyset$) with symmetric initial conditions is studied.

1. Introduction

Diffusion controlled reactions in one-dimensional systems have attracted much interest in the last few years. They are non-equilibrium statistical systems which exhibit the property of self-organization. Mean field rate equations fail to reproduce the dynamics of these models which are characterized by non-trivial correlations. Theoretical descriptions of such systems have to take into account local fluctuations in the particle density.

Exact results have been obtained for some one-dimensional models in the continuum from diffusion-like equations [1] and on a one-dimensional lattice [2–4]. In the latter case the time evolution of the system is determined by a master equation [5]. It is useful to rewrite it as a Euclidian Schrödinger equation [3].

Interesting experimental measurements have been reported recently [6–9]. With the help of these experiments the exciton propagation is studied. The conclusion drawn from the measurements is that this propagation takes place in one dimension along structures like molecular 'wires' and chains. The luminescence decay at early times is algebraic:

$$c(t) \propto \frac{1}{t^{\alpha}} \qquad \text{with} \quad \alpha = \tfrac{1}{2}. \tag{1.1}$$

This observation yields the conjecture that exciton–exciton annihilation processes take place [9]. However, it is not clear whether exciton–exciton coagulation (fusion) reactions happen. In both the annihilation and coagulation chemical models the long-time behaviour of the concentration obeys (equation (1.1)). Moreover, we will prove that this asymptotic behaviour of the concentration is common to all models in which both reactions take place (3.10). We call them annihilation–coagulation systems. The coefficient of the leading term depends on the ratio of the rates of these two processes.

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We show in this paper that all exciton reaction rates can be determined from experimental data. In our study we use a numerical and an analytical approach. First, we want to determine the next-to-leading term in the long-time expansion of the concentration in the coagulation model. Three results are given in the literature concerning this regime. An approximative equation for the particle concentration derived from empty interval probabilities is obtained in [10]. A approximation scheme applied to a diffusion-type equations is used in [11]. These two approaches lead to almost the same conjecture:

$$c(t) = \sqrt{\frac{1}{2\pi t}} + \frac{R}{\pi t}\frac{\Delta'}{1-\Delta'} + O(t^{-x}) \tag{1.2}$$

where $R = 1$ in [11] and $R = \pi/4 \simeq 0.786$ in [10]. Here $1 - \Delta'$ is the coagulation rate. The time-scale in which the diffusion rate is equal to one is chosen. The coefficient of the leading term was exactly computed for $\Delta' = \emptyset$ [12]. In this case the next-to-leading term is of order $t^{-3/2}$ [4] and the operator which appears in the master equation is the Hamiltonian of a supersymmetric system [3].

We verify the conjecture (1.2) by determining the value of the coefficient of the next-to-leading term in the $\Delta' \neq \emptyset$ case. It can be computed by solving the Bethe ansatz equations for the XXZ chain in an external field [3]. However, this computation is a mathematical challenge because one needs to know both the spectra and the eigenvectors of the Hamiltonian. Here we investigate this problem with the help of Monte Carlo simulations. The numerical data presented in section 4 strongly support the $R = 1$ conjecture.

Secondly, we want to find all chemical models which can be described with the help of (1.2) and other results concerning the coagulation model. We do this by looking for similarity transformations which map the coagulation model onto other reaction–diffusion systems. In [12–14] it was shown that the one-species annihilation and coagulation systems are connected. A general proof is given within the framework of the 'Hamiltonian' formalism. The operators which appear in the master equations describing the two models are the Hamiltonians of quantum chains [3] and are equivalent. We say that two reaction–diffusion systems are equivalent when the corresponding Hamiltonians can be obtained one from the other through a similarity transformation [4]. In this paper we apply this method and look for further transformations which connect chemical models and lead to linear relations among their observables. Thus one can extend results concerning one reaction–diffusion system to all its equivalent ones.

We prove that the one-species coagulation–annihilation models can be divided into equivalence classes with respect to one parameter. Each class also contains a pure coagulation and a pure annihilation model.

Another reaction–diffusion system which has attracted much attention is the two-species annihilation model ($A + B \longrightarrow \emptyset + \emptyset$). Scaling considerations indicate that, for equal initial site occupation probabilities of the two species, the concentration decay is algebraic with a time exponent equal to $\frac{1}{4}$ [15]. This has been proven in [16]. In [17] it was conjectured that the asymptotic behaviour of the concentration depends on its initial value:

$$c_A(t) = c_B(t) = \frac{K'\sqrt{c(0)}}{t^{1/4}} + O(t^{-x}) \qquad \text{if} \quad c_A(0) = c_B(0) = c(0) \tag{1.3}$$

where $K' = (2\pi)^{-3/4} \simeq 0.252$. A similar result was obtained in [18] through a renormalization group analysis for spatial dimensions higher than two. In this work the conjectured dependence is validated for small values of $c(0)$. A general closure scheme for truncating the hierarchies of the joint density function equations leads to the same formula but with $K' = (32\pi)^{-1/4} \simeq 0.316$ [19]. A value of $K' = 0.28$ has been numerically

determined [17]. We report results of Monte Carlo simulations started at low values of the initial concentration. A better estimate for the constant K' is obtained. It is in good agreement with the values conjectured in [17, 18].

We also look for the chemical models equivalent to the two-species annihilation reaction–diffusion system.

The paper is structured as follows. In section 2 a brief review of the 'Hamiltonian' formalism is given. A general condition satisfied by the transformations which map reaction–diffusion systems among themselves is derived.

In section 3 we look for chemical models equivalent to systems previously studied. We concentrate on systems for which the particle concentration was computed.

Two models which the procedure is applied to were mentioned before: the one-species coagulation and the two-species annihilation reaction–diffusion systems. Another model which is exactly solved in some special case is the coagulation–decoagulation one [1, 4]. We find two reaction–diffusion systems which are equivalent to it: the annihilation–creation and the death–decoagulation models. Thus we can give exact solutions for the particle number in a probabilistic cellular automaton in which decoagulation and death processes are permitted, but particles do not diffuse.

In the last section new results obtained from Monte Carlo simulations are presented.

2. The master equation and the similarity transformation

We begin with a brief review of the 'Hamiltonian' description of reaction–diffusion systems. We will use the same notations and conventions as in [4, 20] where a comprehensive description of this formalism is given.

First, one has to construct the configuration space. Hard-core interactions among particles moving on the one-dimensional lattice are considered. This also means that multiple occupation of sites is not allowed. If the system is populated with one species of particles, each site can be either empty or occupied by one particle. If there are two-species of particles each site can be in one of three states. The number of states is denoted by N and the lattice length by L. To each site i we attach a N-dimensional vector space V_i. In the two state models ($N = 2$) the vector $(1, 0)$ corresponds to a vacancy while the vector $(0, 1)$ corresponds to an occupation of the site by an A particle. In the three-state models ($N = 3$) the vectors $(1, 0, 0)$, $(0, 1, 0)$ and $(0, 0, 1)$ correspond to an empty site and to occupation of the site by a particle A or B, respectively.

The configuration space has the structure of an L-fold tensor product $V = V_1 \otimes \cdots \otimes V_L$. Each of the N^L vectors of the tensor product base corresponds to a possible configuration of the lattice. A natural convention used to denote these vectors is obtained by associating to each site a variable β_i which takes integer values between 0 and $N - 1$. In the two-state models we use $\beta_i = 0$ to index a vacancy while $\beta_i = 1$ corresponds to the presence of an A particle. In addition, $\beta_i = 2$ is used to describe a site that is occupied by a B particle, in the three-state models. In this 'spin basis' [5]

$$|\{\beta\}\rangle = |\beta_1, \ldots, \beta_L\rangle \tag{2.1}$$

we define the ket vector

$$|P\rangle = \sum_{\{\beta\}} P(\{\beta\}; t)|\{\beta\}\rangle \tag{2.2}$$

which describes the state of the system. $P(\{\beta\}; t)$ stands for the realization probability of the configuration $\{\beta\} = \{\beta_1, \ldots, \beta_L\}$ at time t.

The state of a system can also be described by the set of empty intervals probabilities [21]. They are defined as the probabilities to find one or more sequences of consecutive vacant sites. This description sometimes simplifies exact computations.

The dynamics of reaction–diffusion systems are determined by the rates of the allowed processes. We consider only reactions which can take place when two hopping particles collide. So the set of parameters is given by the probabilities that a state (α, β) on two adjacent sites will change into the state (γ, δ) after one unit of time, denoted by

$$\Gamma_{\gamma,\delta}^{\alpha,\beta} \qquad \text{with the convention} \quad \Gamma_{\alpha,\beta}^{\alpha,\beta} = 0 . \tag{2.3}$$

Here α, β, γ and δ take values between 0 and $N-1$. The sum of all reaction rates which modify the state (α, β) of two neighbouring sites is denoted by

$$\Gamma_{\alpha,\beta} = \sum_{\gamma,\delta=0}^{N-1} \Gamma_{\gamma,\delta}^{\alpha,\beta} . \tag{2.4}$$

All rates are non-negative, constant and real. We discuss only the left–right symmetric case. That is, all the rates satisfy the condition

$$\Gamma_{\gamma,\delta}^{\alpha,\beta} = \Gamma_{\delta,\gamma}^{\beta,\alpha} . \tag{2.5}$$

For one-species systems a list of reactions and corresponding rates is given below:

diffusion	$A + \emptyset \to \emptyset + A$	rate	Γ_{01}^{10}
annihilation	$A + A \to \emptyset + \emptyset$	rate	Γ_{00}^{11}
creation	$\emptyset + \emptyset \to A + A$	rate	Γ_{11}^{00}
coagulation	$A + A \to A + \emptyset$	rate	Γ_{10}^{11}
decoagulation	$A + \emptyset \to A + A$	rate	Γ_{11}^{10}
death	$A + \emptyset \to \emptyset + \emptyset$	rate	Γ_{00}^{10}
birth	$\emptyset + \emptyset \to A + \emptyset$	rate	Γ_{10}^{00}.

The master equation [5] describing the time evolution of the probability distribution $P(\{\beta\}; t)$ can be written in the form of a Euclidian Schrödinger equation [3]

$$\frac{\partial}{\partial t}|P\rangle = -H|P\rangle \tag{2.6}$$

where H operates on V. We consider periodic boundaries. Due to the fact that the reactions take place only between particles placed on two neighbouring sites this Hamiltonian can be written as a sum:

$$H = \sum_{i=1}^{L} H_i \tag{2.7}$$

where H_i acts locally on $V_i \otimes V_{i+1}$. This operator is defined by

$$H_i = \sum_{\alpha,\beta=0}^{N-1} \left[\Gamma_{\alpha,\beta}\, E_i^{\alpha,\alpha} E_{i+1}^{\beta,\beta} - \sum_{\gamma,\delta=0}^{1} \Gamma_{\gamma,\delta}^{\alpha,\beta}\, E_i^{\gamma,\alpha} E_{i+1}^{\delta,\beta} \right] \tag{2.8}$$

where E^{kl} are $N \times N$ matrices with the entries $(E^{kl})_{nm} = \delta_{k,n}\delta_{l,m}$. This Hamiltonian is not necessarily Hermitian. It ensures the conservation of probability and thus has a bra ground state

$$\langle 0|H = 0 \tag{2.9}$$

which is the sum of all basis vectors

$$\langle 0| = \sum_{\{\beta\}} \langle\{\beta\}| \,. \tag{2.10}$$

In other words, if we consider H_i as a matrix, equation (2.8) with (2.4) states that the sum of the entries from each column is 0. H has the same property (2.9).

The formal solution of (2.6) is

$$|P(t)) = \exp(-Ht)|P_0\rangle \tag{2.11}$$

where the initial state is denoted by $|P_0\rangle$ [3].

The expectation value of an observable X is given by the matrix element

$$\langle X\rangle(|P_0\rangle, t) = \sum_{\{\beta\}} X(\{\beta\})P(\{\beta\}; t) = \langle 0|X|P(t)\rangle \,. \tag{2.12}$$

The advantage of this Hamiltonian formalism is that we can introduce similarity transformations. Our interest is to find those B for which the transformed Hamiltonian

$$\hat{H} = \sum_{i=1}^{L} \hat{H}_i = B\,H\,B^{-1} \tag{2.13}$$

also describes the dynamic of a reaction–diffusion system. (The quantities in the transformed model are denoted with a ^.) Therefore we have to concentrate on local transformations:

$$B = b \otimes b \otimes \cdots \otimes b \tag{2.14}$$

defined by the L-fold tensor product of a real $N \times N$ matrix b. The mapping can easily be extended to the computation of expectation values. They can be computed [4] from the corresponding expectation values in the 'original' model:

$$\begin{aligned} \widehat{\langle X\rangle}(|P_0\rangle, t) &= \langle 0|X \exp(-\hat{H}t)|P_0\rangle \\ &= \langle 0|XB \exp(-Ht)B^{-1}|P_0\rangle = \langle XB\rangle(B^{-1}|P_0\rangle, t) \,. \end{aligned} \tag{2.15}$$

Some restrictions have to be imposed on b. First, the sum of the entries from each column of $\hat{H}_i$ must be 0:

$$\langle 0|BH_i = 0 \,. \tag{2.16}$$

A general solution of (2.16) is

$$\langle 0|B = \rho\langle 0| \,. \tag{2.17}$$

Here ρ is real. We have checked explicitly for every model considered in this paper that transformations obeying (2.16) but not (2.17) do not lead to new mappings. Using equation (2.10) and (2.14) one gets the restriction

$$\sum_{i=0}^{N-1} b_{ij} = \rho^{1/L} \,. \tag{2.18}$$

We can eliminate an overall scaling parameter by choosing $\rho = 1$. Thus the transformations are represented by real $N \times N$ matrices with the property that the sum of the entries on each column is equal to 1.

Not all these $N \times (N-1)$-parameter transformations lead to another reaction–diffusion system. The matrix elements of the two sites ($L = 2$) Hamiltonian $\hat{H}$ have to satisfy the following sign conditions:

$$\begin{aligned} &\hat{H}_{mm} \geqslant 0 \qquad m = 0; 1 \ldots N^2 - 1 \\ &\hat{H}_{mn} \leqslant 0 \qquad m, n = 0, 1 \ldots N^2 - 1 \qquad m \neq n \,. \end{aligned} \tag{2.19}$$

The remaining transformations map reaction–diffusion system one onto the other. The program REDUCE was used to compute the transformed Hamiltonians.

3. Mappings between reaction–diffusion systems

We will start with the Hamiltonians of those chemical models for which the behaviour of the particle concentration is known and look for local transformations which map them onto other reaction–diffusion systems.

There are always the $N! - 1$ permutation transformations which interchange (if $N = 3$ different types of) particles and vacancies. We will treat everything 'modulo' these permutations.

As we mentioned before, we restrict ourselves to the left–right symmetric case. The similarity transformation preserves this property. The generalization to the non-symmetric case is straightforward.

In the three-state model we take equal diffusion rates for the two kinds of particles ($\Gamma_{10}^{01} = \Gamma_{20}^{02}$). One parameter of the problem can be eliminated by choosing the time-scale in which the diffusion rates are equal to 1.

3.1. Two-state models

Exact results are available for the one-species annihilation, coagulation and coagulation–decoagulation models. For the last two models the system of linear differential equations for the empty interval probabilities was written and solved. These equations decouple from those corresponding to many intervals probabilities [20, 22]. In the transformed models identical systems of equations can be written but for other observables, namely the probabilities of having interpolating sequences of vacancies and particles on consecutive sites [20].

The general form of the local transformation is given by (2.18):

$$b = \begin{pmatrix} 1-\alpha & \beta \\ \alpha & 1-\beta \end{pmatrix}. \tag{3.1}$$

We will start with the annihilation model and not with the equivalent coagulation one. For simplicity we will use the notation a, instead of Γ_{00}^{11}, for the annihilation rate and f for the coagulation rate (Γ_{10}^{11}).

The annihilation model. In this case particles diffuse and disappear pairwise when two of them try to occupy the same site. We denote the corresponding Hamiltonian with $H_{\text{ann.}}$ and introduce, as in [3, 4, 28], the parameter Δ' defined by

$$\Delta' = 1 - \frac{a}{2}. \tag{3.2}$$

If $a < 2$, Δ' is equal to the probability that no reaction takes place at a two-particles encounter. If the parameter Δ' is zero exact solutions can be obtained because $H_{\text{ann.}}$ can be expressed in terms of free fermions. In [2] the time dependence of the particle concentration is derived in the thermodynamical limit with a full lattice as initial configuration. The finite chain is treated exactly in [3] and the two-point correlation functions for the infinite chain are obtained. The particle concentration for random homogeneous initial conditions is deduced in [4].

It can easily be seen that the transformed Hamiltonian $\hat{H}$ will only fulfil the conditions (2.19) if at least one of the entries of b is zero. We take $\alpha = 0$. The case $\beta = 0$ can be obtained by applying a permutation transformation on $\hat{H}$.

It is a well known fact [12–14] that the annihilation and coagulation models are equivalent. We reobtain this result by choosing $\beta = -1$. In this case $\hat{H}$ is identical with the Hamiltonian of the coagulation model [4].

This is, however, only one of the possible choices for the transformation parameter. For $-1 \leqslant \beta \leqslant 0$ we get the Hamiltonian of a system in which apart from diffusion with the same rate $\hat{\Gamma}_{01}^{10} = 1$, the following processes are permitted:

- annihilation with rate

$$\hat{\Gamma}_{00}^{11} = \hat{a} = 2\frac{\beta+1}{1-\beta}(1-\Delta')$$

- coagulation with rate

$$\hat{\Gamma}_{01}^{11} = \hat{f} = 2\frac{\beta}{\beta-1}(1-\Delta').$$

The matrix b and Δ' can be written as functions of $\hat{a}$ and $\hat{f}$:

$$\Delta' = 1 - \hat{f} - \hat{a}/2 \tag{3.3}$$

or equivalently $a = \hat{a} + 2\hat{f}$. One gets the following result:

$$\hat{H}(\hat{a}, \hat{f}) = B H_{\text{ann.}} B^{-1}$$

where

$$b = \begin{pmatrix} 1 & \dfrac{-\hat{f}}{\hat{a}+\hat{f}} \\ 0 & \dfrac{\hat{a}+2\hat{f}}{\hat{a}+\hat{f}} \end{pmatrix} \tag{3.4}$$

and with the parameter Δ' (equation (3.3)).

We have obtained a one-parameter group of similarity transformations which act between annihilation–coagulation models ($(a, f) \longrightarrow (\hat{a}, \hat{f})$) corresponding to the same value of Δ' (i.e. $a/2 + f = \hat{a}/2 + \hat{f}$). It is straightforward to write the transformation which connects the coagulation model ($a = \emptyset$) and one described by $\hat{H}(\hat{a}, \hat{f})$.

Some explicit results concerning the particle concentration can be given now by applying transformations (2.15) on formulae obtained for the annihilation ($f = 0$) (equation (3.4)) or coagulation ($a = 0$) models.

Uncorrelated initial conditions are considered: at time $t = 0$ every site is occupied with the same probability p. In this case

$$B^{-1}|\vec{P}_0\rangle = \begin{pmatrix} 1 - \dfrac{\hat{a}+\hat{f}}{\hat{a}+2\hat{f}}p \\ \dfrac{\hat{a}+\hat{f}}{\hat{a}+2\hat{f}}p \end{pmatrix}_1 \otimes \cdots \otimes \begin{pmatrix} 1 - \dfrac{\hat{a}+\hat{f}}{\hat{a}+2\hat{f}}p \\ \dfrac{\hat{a}+\hat{f}}{\hat{a}+2\hat{f}}p \end{pmatrix}_L$$

which corresponds to an initial site occupation probability in the equivalent annihilation model equal to $\frac{\hat{a}+\hat{f}}{\hat{a}+2\hat{f}}p$.

The operator of the occupation number of the site i is given by

$$n_i = \begin{pmatrix} 0 & 0 \\ 0 & 1 \end{pmatrix}_i . \tag{3.5}$$

The expectation value of the product of k such operators in the coagulation–annihilation model is (2.15)

$$\langle \widehat{n_{i_1} \cdots n_{i_k}} \rangle (p, t) = \left(\frac{\hat{a}+2\hat{f}}{\hat{a}+\hat{f}}\right)^k \langle n_{i_1} \cdots n_{i_k} \rangle_{\text{ann}} \left(\frac{\hat{a}+\hat{f}}{\hat{a}+2\hat{f}}p, t\right). \tag{3.6}$$

We now drop the superscript.

The exactly solvable case $\Delta' = 1 - f - \frac{a}{2} = 0$ corresponds to the physical situation that whenever two particles meet at least one of them disappears from the system. We mentioned in the introduction that the operators which appear in the master equation of these models are the Hamiltonians of supersymmetric systems. These reaction–diffusion models are mentioned in [23] in connection with the Glauber dynamic of the q state Potts model. The results given in [4] for $f = 1$ or $a = 2$ can be generalized to these annihilation–coagulation models. The particle concentration on a finite chain is

$$c(t, p, L) = \frac{1 - [1 - (2 - f)p]^L}{(2 - f)L} - \frac{1}{(2 - f)L} \sum_{k=1}^{L-1} \left\{ \left[\frac{1 - (-1)^k [1 - (2 - f)p]^L}{\frac{1 + [1 - (2-f)p]^2}{2[1 - (2-f)p]} - \cos\frac{\pi k}{L}} \right. \right.$$
$$\left. - \frac{1 - (-1)^k [1 - (2 - f)p]^L}{1 - \cos\frac{\pi k}{L}} \right]$$
$$\left. \times \sin^2\left(\frac{\pi k}{L}\right) \exp\left[-4t\left(1 - \cos\frac{\pi k}{L}\right)\right] \right\}. \tag{3.7}$$

The behaviour of the concentration (3.7) in the finite-size scaling limit [3]

$$L \to \infty \qquad t \to \infty \qquad \text{with } z = \frac{4t}{L^2} \text{ fixed} \tag{3.8}$$

can be determined [4]. Exact expressions can be written for the scaling and the first correction functions in terms of Jacobi theta functions. We get the following scaling relation:

$$L\, c(z, p, L) = \frac{1}{2 - f} \left\{ \theta_3\left(0, \frac{i\pi z}{2}\right) + \frac{1}{L^2} \left[\frac{z}{6} \frac{\partial^2}{\partial z^2} \theta_3\left(0, \frac{i\pi z}{2}\right) \right.\right.$$
$$\left.\left. + \frac{1}{2}\left(1 - \frac{2}{(2 - f)p}\right)^2 \frac{\partial}{\partial z} \theta_3\left(0, \frac{i\pi z}{2}\right) \right] \right\} + O\left(\frac{1}{L^4}\right). \tag{3.9}$$

Another interesting result concerns the large-time behaviour of the particle concentration in the thermodynamic limit. In the $\Delta' \neq \emptyset$ case we start from the conjectured expression (1.2). Applying (2.15) we get

$$c(t) = \frac{a + 2f}{a + f} \left[\sqrt{\frac{1}{8\pi t}} + \frac{R}{2\pi} \frac{\Delta'}{1 - \Delta'} \frac{1}{t} \right] + O(t^{-x}). \tag{3.10}$$

The coefficient of the leading term depends only on the ratio of the annihilation and the coagulation rates. The next-to-leading term ($\sim 1/t$) is dependent on Δ'. Both of them are independent of the initial concentration. We will give strong numerical evidence which supports the conjecture presented in [11] (i.e. $R = 1$) in section 4.

This asymptotic expression is an exact result in the supersymmetric ($\Delta' = 0$) case [2]. The next-to-leading term is of order $t^{-3/2}$. For uncorrelated homogeneous initial conditions, in the $z \to \emptyset$ limit [4] of (3.9) one gets

$$c(t, p) = \frac{1}{2 - f} \left\{ \sqrt{\frac{1}{2\pi t}} - \frac{1}{\sqrt{2\pi}} \frac{1}{16} \left[\left(1 - \frac{2}{(2 - f)p}\right)^2 - \frac{1}{2} \right] \frac{1}{t^{3/2}} \right\} + O(t^{-5/2}). \tag{3.11}$$

During the course of the publication of this paper, two other works concerning the the annihilation–coagulation model appeared. In one of them the method of similarity transformations is used [24], the other one is based on a field-theoretical approach [25].

The coagulation–decoagulation model. In this system, apart from diffusion, coagulation and its backward reaction are permitted. We will use, as in [4], the variable η defined through the decoagulation rate: $\eta^2 = \Gamma_{11}^{10} + 1$.

Exact solutions of this model are available in the continuum limit [1] and for the finite lattice in the case in which the coagulation and diffusion rates are equal ($f = 1$) [4]. In the latter case the steady state concentration is $c_s = 1 - \eta^{-2}$, independently of L and p.

We found that the coagulation–decoagulation model is equivalent with two other reaction–diffusion systems. Only two transformations, which correspond to special choices for α and β in (3.1), connect this model with others.

(i) One of them is

$$b = \begin{pmatrix} 1 & -\frac{1}{f}\eta^2 - 1 \\ 0 & 1 + \frac{1}{f}\eta^2 - 1 \end{pmatrix}. \tag{3.12}$$

If $f \leqslant 1$, the transformed Hamiltonian $\hat{H}$ corresponds to a reaction–diffusion system with the following processes:

- diffusion with rate $\hat{\Gamma}_{01}^{10} = 1 - f$
- decoagulation with rate $\hat{\Gamma}_{11}^{10} = f + \eta^2 - 1$
- death with rate $\hat{\Gamma}_{00}^{10} = f$.

If $f = 1$ there is no diffusion in the new model. The motion of the particles is realized by successive processes of decoagulation (with rate $\eta^2 > 1$) and death (with rate 1) on neighbouring pairs of lattice sites. In this case we can use the results from [4]. For the finite chain and homogeneous random initial conditions the particle concentration is

$$c(t, p, L) = \frac{1 - [1 - p(1 - \eta^{-2})]^L}{1 - \eta^{-2L}} - \frac{1}{\eta L} \sum_{k=1}^{L-1} \left\{ \left[\frac{1 + (-1)^{k+1} \eta^L [1 - p(1 - \eta^{-2})]^L}{\frac{1+\eta^2[1-p(1-\eta^{-2})]^2}{2\eta[1-p(1-\eta^{-2})]} - \cos\frac{\pi k}{L}} \right. \right.$$
$$\left. \left. - \frac{1 + (-1)^{k+1} \eta^L [1 - p(1 - \eta^{-2})]^L}{\frac{\eta^2+1}{2\eta} - \cos\frac{\pi k}{L}} \right] \sin^2\left(\frac{\pi k}{L}\right) \exp\left(-\Lambda_k^0 t\right) \right\} \tag{3.13}$$

where Λ_k^0 is

$$\Lambda_k^0 = \eta\left(2(\eta + \eta^{-1}) - 4\cos\frac{\pi k}{L}\right). \tag{3.14}$$

The full lattice is a stationary state:

$$c(0, 1, L) = c(t, 1, L) = 1.$$

Interchanging A and $\emptyset$ a system with a death rate η^2 greater than the decoagulation rate 1 and zero steady state concentration is obtained. For this choice of rates (3.13) gives the concentration of vacancies if p is replaced by $1 - p$.

(ii) The other transformation which gives a mapping of the coagulation–decoagulation model is:

$$b = \begin{pmatrix} \frac{1}{2}\left[1 + \sqrt{\frac{\eta^2 - 1}{f} + 1}\right] & \frac{1}{2} \\ \frac{1}{2}\left[1 - \sqrt{\frac{\eta^2 - 1}{f} + 1} - 1\right] & \frac{1}{2} \end{pmatrix}. \tag{3.15}$$

One obtains an annihilation–creation reaction–diffusion system with rates:

- diffusion $\hat{\Gamma}_{01}^{10} = \frac{\eta^2+1}{2}$
- annihilation $\hat{\Gamma}_{00}^{11} = \hat{a} = \frac{1}{2}\left[\sqrt{\frac{\eta^2-1}{f}+1}+1\right]^2 f^2$
- creation $\hat{\Gamma}_{11}^{00} = \frac{1}{2}\left[\sqrt{\frac{\eta^2-1}{f}+1}-1\right]^2 f^2$.

The annihilation rate is greater than the creation rate. The time unit can be adjusted in such a way that the new diffusion rate $\hat{\Gamma}_{01}^{10}$ becomes equal to 1.

In the case $f = 1$ the sum of the annihilation and creation rates is equal to 2. We can redefine the variable η:

$$\eta = 1 + \frac{2}{\left(\frac{\hat{a}}{2-\hat{a}}\right)^y - 1}$$

where $y = \frac{1}{2}$. The concentration for the finite chain is

$$c(t, p, L) = \frac{1-\eta}{2} + \frac{(\eta-\eta^{-1})[1-(1-2p)^L\eta^{-L}]}{2(1-\eta^{-2L})} - \frac{1}{2L}\sum_{k=1}^{L-1}\left\{\left[\frac{1+(-1)^{k+1}(1-2p)^L}{\frac{1+(1-2p)^2}{2(1-2p)} - \cos\frac{\pi k}{L}} - \frac{1+(-1)^{k+1}(1-2p)^L}{\frac{\eta^2+1}{2\eta} - \cos\frac{\pi k}{L}}\right] \times \sin^2\left(\frac{\pi k}{L}\right)\exp\left(-\Lambda_k^0 t\right)\right\} \tag{3.16}$$

where Λ_k^0 is

$$\Lambda_k^0 = \frac{2\eta}{\eta^2+1}\left(2(\eta+\eta^{-1}) - 4\cos\frac{\pi k}{L}\right). \tag{3.17}$$

The steady-state concentration is $c_s = (1-\eta^{-1})/2 < 0.5$.

After a permutation of the two lines in (3.15) a system with interchanged annihilation and creation rates is obtained. The redefinition of η in the exactly solved case is the same as the one given above but with $y = -\frac{1}{2}$. Formula (3.16) gives the vacancies concentration if p is again replaced with $1-p$. The steady-state concentration is $c_s = (1+\eta^{-1})/2 > 0.5$.

In the limit $\eta \to \infty$ the reaction–diffusion system in which all three rates are equal is obtained. The operator which appears in the corresponding master equation is the Hamiltonian of the Ising model [3].

3.2. Three-state models

The starting point in this section is the two-species annihilation model. The particles diffuse and react when an A and a B try to occupy the same site:

$$A + B \longrightarrow \emptyset + \emptyset.$$

We denote the annihilation rate with a ($\Gamma_{00}^{12} = a$), as in the preceding section. All diffusion rates are equal to 1 and $\Gamma_{12}^{21} = 0$ modelling hard-core interactions between particles.

The two sites Hamiltonian for this model is given by

$$H = 2\times\begin{pmatrix} 0 & 0 & 0 & 0 & 0 & -a & 0 & -a & 0 \\ 0 & 1 & 0 & -1 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 1 & 0 & 0 & 0 & -1 & 0 & 0 \\ 0 & -1 & 0 & 1 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & a & 0 & 0 & 0 \\ 0 & 0 & -1 & 0 & 0 & 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & a & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \end{pmatrix} \tag{3.18}$$

in the $\{|00\rangle, |01\rangle, |02\rangle, \ldots |22\rangle\}$ base.

A general transformation has the form (2.18):

$$b = \begin{pmatrix} 1-\alpha-\beta & \gamma & \epsilon \\ \alpha & 1-\gamma-\delta & \zeta \\ \beta & \delta & 1-\zeta-\epsilon \end{pmatrix}. \tag{3.19}$$

We have to ensure that the transformed Hamiltonian satisfies the condition (2.19). The first, fifth and ninth columns of $\hat{H}$ are identical up to a factor. So are line one, five and nine. The matrix elements of $\hat{H}$ can be interpreted in terms of reaction rates only if the entries from at least one of these lines or from its corresponding column are zero. The other two lines $i, j \in \{1, 5, 9\}$ have at most two non-zero entries in the two corresponding columns i, j. By imposing this condition we are left with one transformation

$$b = \begin{pmatrix} 1 & 1-\alpha & 1-\beta \\ 0 & \alpha & 0 \\ 0 & 0 & \beta \end{pmatrix} \tag{3.20}$$

which leads to a new reaction–diffusion system if a, α and β are greater than one and if $\alpha^{-1} + \beta^{-1} \geqslant 1$. The transformed two-sites Hamiltonian is

$$\hat{H} = 2\times\begin{pmatrix} 0 & 0 & 0 & 0 & 0 & -a(-1+\alpha^{-1}+\beta^{-1}) & 0 & -a(-1+\alpha^{-1}+\beta^{-1} & 0 \\ 0 & 1 & 0 & -1 & 0 & -(1-\alpha^{-1}) & 0 & -(a-1)(1-\alpha^{-1}) & 0 \\ 0 & 0 & 1 & 0 & 0 & -(a-1)(1-\beta^{-1}) & -1 & -(1-\beta^{-1}) & 0 \\ 0 & -1 & 0 & 1 & 0 & -(a-1)(1-\alpha^{-1}) & 0 & -(1-\alpha^{-1}) & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & a & 0 & 0 & 0 \\ 0 & 0 & -1 & 0 & 0 & -(1-\beta^{-1}) & 1 & -(a-1)(1-\beta^{-1}) & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & a & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \end{pmatrix}. \tag{3.21}$$

$\hat{H}$ has the same diffusion part and is left–right symmetric. All processes which reduce the number of particles start from an $A\,B$ pair. Apart from annihilation we have 'two-species coagulation' reactions:

- $A + B \to \emptyset + \emptyset$ with rate $\hat{\Gamma}^{12}_{00} = a\,(\alpha^{-1} + \beta^{-1} - 1)$
- $A + B \to \emptyset + A$ with rate $\hat{\Gamma}^{12}_{01} = 1 - \alpha^{-1}$
- $A + B \to A + \emptyset$ with rate $\hat{\Gamma}^{12}_{10} = (a-1)(1-\alpha^{-1})$
- $A + B \to B + \emptyset$ with rate $\hat{\Gamma}^{12}_{20} = 1 - \beta^{-1}$
- $A + B \to \emptyset + B$ with rate $\hat{\Gamma}^{12}_{02} = (a-1)(1-\beta^{-1})$.

Note that the diagonal terms of the Hamiltonian are invariant under the transformation ($\hat{\Gamma}_{12} = \Gamma_{12} = a$). The coagulation rates depend on the type of the particle which survives the process and they are $(a-1)$ times smaller if the particle which survives is the one which jumps. There are only two coagulation rates, independent of the initial position of the surviving particle if $a = 2$. This case corresponds to the physical situation in which at any A B encounter at least one particle leaves the system.

From equation (2.15) and (1.3) it is easy to see that the concentration will have an algebraic fall-off if

$$\frac{c_A(0)}{c_B(0)} = \frac{\alpha}{\beta}.$$

In this case the particle concentrations are

$$c_A(t) = \alpha^{1/2} \frac{K' \sqrt{c_A(0)}}{t^{1/4}} \tag{3.22}$$

$$c_B(t) = \beta^{1/2} \frac{K' \sqrt{c_B(0)}}{t^{1/4}} = \frac{\beta}{\alpha} c_A(t). \tag{3.23}$$

4. Monte Carlo simulations

Only a few chemical models have been solved exactly. For some others, quantitative estimates have been made with the help of approximation schemes. This is the reason why numerical methods have been extensively used in the study of reaction–diffusion systems. The dynamics of the particle density and spatial distribution can be determined with the help of Monte Carlo simulations [26, 27].

The Hamiltonian formalism opens new possibilities in this field. For small chains numerical data can be obtained with a high accuracy by using diagonalization techniques or simulations. The study of the finite-size scaling behaviour (3.8) in the limit $z \to \emptyset$ permits the determination of the particle concentration for infinite chains at very large times [28].

In this section we present our numerical results concerning the asymptotic behaviour of the particle concentration in the one- and two-species annihilation models. We simulate directly the thermodynamical limit by using large lattices.

We consider chemical systems in which no reaction is permitted which creates particles on a pair of empty sites. This is why the so-called 'direct' Monte Carlo method [26, 29] can be used. The way we implement this method is described in detail in [28].

We will first present our results for the two-state models and continue with the three-state models.

4.1. The two-state annihilation–coagulation model

The influence of the reaction rate in annihilation and coagulation models is a subject which has received a lot of attention in recent years (see [10, 11] and references therein). We mentioned in the introduction the conjectured expression (1.2) for the concentration decay in the $\Delta' \neq \emptyset$ coagulation model. This result is in agreement with the conclusions of a numerical study we carried out in our previous work. Using a finite-size scaling analysis we obtained the same value for the time exponents in the leading $(-\frac{1}{2})$ and the next to leading term (-1) and we found that the coefficient of the leading term is independent of Δ' [28].

As mentioned there, a better test of (1.2) (or equivalently of (3.10)) can be made through a direct simulation of the thermodynamic limit. Results of such computations are presented in [11]. They are, in some cases, in semi-quantitative agreement with the

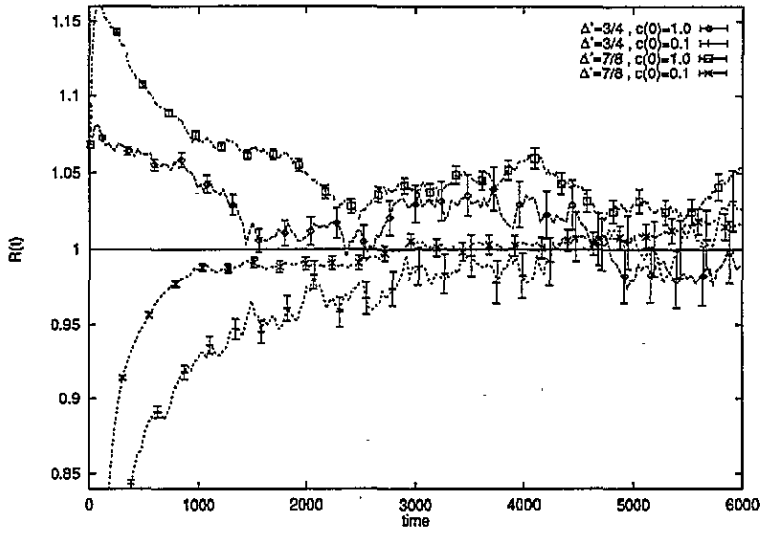

Figure 1. The Monte Carlo data are represented by dashed and chain curves. Error bars are given only for a few points. Effects of the annihilation rate on the asymptotic behaviour of the particle concentration for the $A + A \longrightarrow \emptyset + \emptyset$ reaction $-R(t)$ as defined by (4.1), for $\Delta' = \frac{3}{4}$ and $\frac{7}{8}$. The straight line is $R(t) = 1$.

conjecture presented there. We concentrate on the long-time behaviour and use smaller lattices as the one mentioned in [11]. Thus we are able to obtain very good statistics in a reasonable CPU time.

We tested the conjecture (1.2) in two different ways. Simulations of the annihilation model were performed on a lattice of length $L = 2000$ for times up to a value of $t_{max} = 1.5 \times 10^4$.

In the first set of simulations two values for the initial concentrations, 1.0 and 0.1, were used. A decrease of three, respectively, two orders of magnitude of the concentration enables us to reach the asymptotic regime. 300 000 runs were performed in order to ensure a relative error of the particle concentration of less then 10^{-3}. In figure 1 the following quantity (see equation (3.10) with $f = 0$):

$$R(t) = 2\pi\left(c(t) - \sqrt{\frac{1}{8\pi t}}\right)\frac{1-\Delta'}{\Delta'}t \tag{4.1}$$

is represented for $\Delta' = 0.875$ and 0.75. We see that our data converge to the value $R(t) = 1$ for which the straight line is drawn.

Simulations were also performed for negative values of Δ'. The convergence is worse, $R(t)$ seems to oscillate. In this case the absolute value of the next to leading term of (3.10) is smaller than for positive Δ's, so $R(t)$ approaches faster the order of magnitude of the numerical errors.

A second set of simulations of the annihilation model were performed in which we averaged only over 20 000 runs. We applied a χ^2 test in two steps to the data. A linear combination of time powers was presumed:

$$y = K_x t^{-x} + K_{x+1/2} t^{-x-1/2} + K_{x+1} t^{-x-1}. \tag{4.2}$$

The first fit was made for the concentration $y = c(t)$ taking $x = \frac{1}{2}$. The coefficient of the leading term was determined with a confidence level greater then 99%. The first three

significant digits of $K_{1/2}$ are identical with those of $(8\pi)^{-1/2} \simeq 0.1995$.

The next step is to determine the coefficient of the first correction. The leading term is subtracted from the numerical data:

$$y = c(t) - \frac{1}{\sqrt{8\pi t}}$$

and a value $x = 1$ is used in the fit (4.2). The confidence level was good (greater then 50%). The values for R are listed in table 1.

Table 1. Values of R (1.2) obtained from Monte Carlo simulations of the annihilation model. The two different conjectured values are 1 [11] and 0.786 [10].

Δ'	Initial concentration c(0)	Values of R from MC data
−7	1.00	1.090 ± 0.020
−7	0.10	1.060 ± 0.010
−3/2	1.00	0.940 ± 0.010
−27/28	1.00	0.950 ± 0.020
−1	1.00	1.000 ± 0.040
−1	0.25	1.180 ± 0.080
−1	0.10	1.060 ± 0.080
−3/7	1.00	1.010 ± 0.030
−1/9	1.00	0.940 ± 0.070
1/2	1.00	1.060 ± 0.040
1/2	0.25	0.990 ± 0.040
3/4	1.00	1.004 ± 0.005
3/4	0.10	1.030 ± 0.020
7/8	1.00	0.998 ± 0.005
7/8	0.10	1.005 ± 0.005

For most values of Δ' simulations of the same model with different initial concentrations were performed, as can be seen in table 1. The best fits were obtained for the choice of reaction rates corresponding to a value of Δ' near 1. This is easy to understand because for such Δ's the contribution of the next to leading term in (3.10) is maximal. The simulations corresponding to $\Delta' = \frac{3}{4}$, and $\frac{7}{8}$ are the ones used to make figure 1. For this data the statistic was made by averaging over 300 000 runs.

The conclusion we draw from the results of the two tests is that the conjecture presented in [11], predicting a value of $R = 1$ in (1.2) is correct.

These results are valid if the data we considered before are not affected by the finite size of the lattice on which the simulations were performed. We check this by performing the simulations described in the following.

In the asymptotic regime the system contains a low number of particles which are spatially separated. A particle which was not involved in a reaction represents a random walker. This means that at large times, of the order of the square of lattice length, finite-size effects should be dominant.

We obtain a more precise quantitative picture from simulations of the annihilation model for times up to $t_{max} = 10^6$. We use a lattice of the same length ($L = 2000$) and averaged over 20 000 runs. Three values for the parameter Δ' were chosen: $\frac{7}{8}$, 0 and −7. Each simulation started with a full lattice as initial configuration.

For an infinite system the long-time behaviour of the concentration is algebraic.

For a finite system the concentration decay is exponential. We are interested to study the transition between the two types of decay, i.e. the onset of finite size effects at large

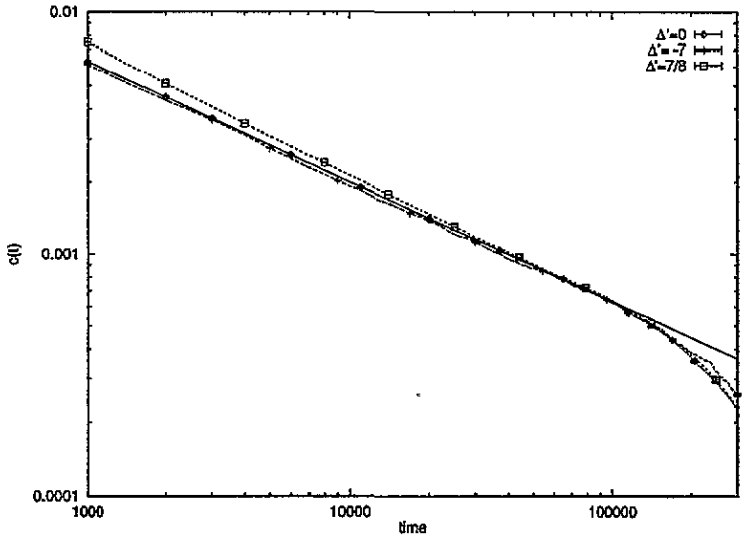

Figure 2. Onset of finite size effects—the algebraic decay. Double logarithmical plot of the concentration for the one-species annihilation reaction.

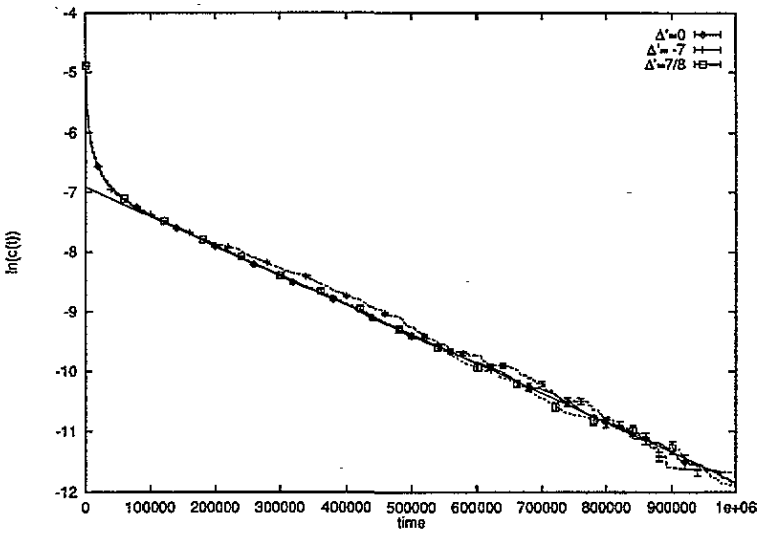

Figure 3. Onset of finite size effects—the exponential decay. Plot of the logarithm of the concentration for the one-species annihilation reaction.

times.

Figure 2 gives a log–log plot of the concentration. The straight line corresponds to $c(t) = (8\pi t)^{-1/2}$. We see that for times $t > 10^5$ the broken curves strongly deviate from the straight line.

In figure 3 which is a plot of the logarithm of the concentration we can see that starting from the same value of time ($t = 10^5$) the curves fit nicely with another straight line

described by

$$c(t) = \frac{2}{L} \exp\left(-2\frac{\pi^2}{L^2}t\right). \tag{4.3}$$

The formula is obtained in the $t \to \infty$ limit of (3.7), for the $\Delta' = 0$ annihilation model ($c = 0$).

From figures 2 and 3 we conclude that the onset of the finite-size effects takes place in a narrow vicinity of $t = 10^5$. This value is considerably smaller than L^2 but much larger than the t_{max} we used. It is now clear that the results we presented in the first part of this section refer to the thermodynamic limit of the annihilation model.

The slope of the line from figure 3 is equal to the energy of the first excited state of H_{ann}. We can conclude that this energy has the value $4(1 - \cos(\pi/L))$ independent of the parameter Δ'. This confirms previous results obtained from numerical diagonalization of this Hamiltonian [30].

4.2. Numerical results for the three-state models

The simulation of the thermodynamic limit of the two-species annihilation reaction requires a very large amount of CPU time.

This has two reasons. A reliable study of the long-time decay of the concentration presumes simulations in which the total number of particles reduces by at least two orders of magnitude. The algebraic fall-off is much slower than in the $A + A \longrightarrow \emptyset + \emptyset$ case. This means that we have to perform the simulations up to much higher values of t_{max} than we did in the one-species case.

On the other hand, if we start at $t = 0$ with a random distribution, there will be local fluctuations of the particle densities. The decay of the initial fluctuations plays an essential role in the dynamics of the system [17]. Their length scale extends in time. So one is forced to use longer lattices than in the one-species case where such domains do not appear.

A lattice of length $L = 10^5$ was used to simulate three state models. In the case of the two-species annihilation we were able to average only over 100 runs. We have chosen small values of the initial density, as suggested by the results obtained for the $d = 2$ case [18]. Our aim is to see if the particle concentration obeys the algebraic decay law (1.3) for the $A-B$ symmetric case and to compute K'. Results from similar simulations were reported in [17] which lead to $K' = 0.280$. This is in equally good agreement with the two conjectured values 0.252 and 0.316.

Figure 4 shows the curve

$$K(t) = c_A(t)\frac{t^{1/4}}{\sqrt{c(0)}} \tag{4.4}$$

for various choices of the annihilation rate and initial site occupation probabilities. The simulations were stopped at values of t_{max} of the order of 10^6. These values of t_{max} are a hundred times larger as the one used in [17]. At very large times the asymptotic regime is reached with the three curves converging to slightly different values. However, the difference between the limit values is less than the sum of their standard deviations.

Our data show a weak dependence of $K(t)$ on a and $c(0)$. This suggests that K' in (1.3) is a function of these parameters. This may well not be the case because the next to leading terms in the asymptotic development of the concentration also contribute to $K(t)$ and can determine the difference between the three curves.

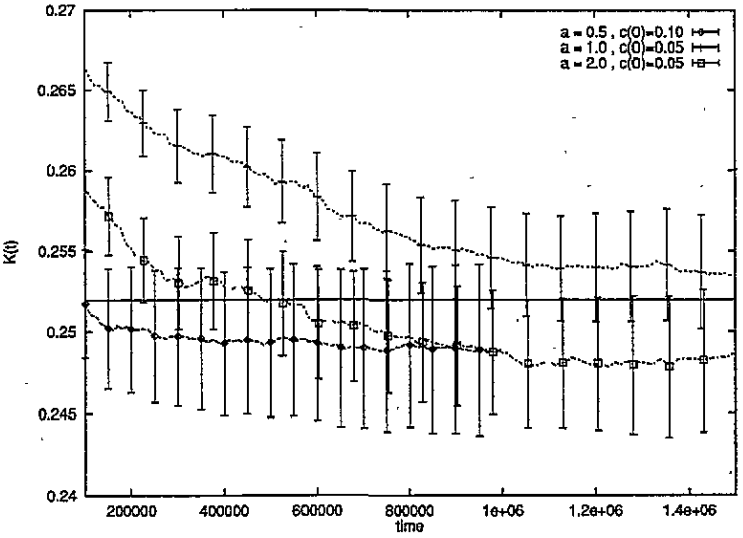

Figure 4. Asymptotical behaviour of the particle concentration in the $A + B \longrightarrow \emptyset + \emptyset$ reaction. Plot of $K(t)$ as defined by (4.4). The straight line corresponds to $K(t) = (2\pi)^{-3/4}$.

We get the estimate $K' = 0.251 \pm 0.003$ by averaging over the three limit values. Our result is important because it shows that the conjectured dependence of the concentration on its initial value [17] is most probably correct.

The weak dependence of $K(t)$ on a suggests that the asymptotic behaviour is universal in the sense it is independent of the annihilation rate. For a better numerical confirmation one should apply the procedure described in the previous section for the $A + A \longrightarrow \emptyset + \emptyset$ case. The first step would be to determine the exponent x in (1.3) [28] and then apply χ^2 tests. This would require better statistics in the determination of the asymptotic behaviour of the concentration. This is an aim which we cannot achieve with the computer facilities which are currently at our disposal.

5. Conclusions

We found a general condition satisfied by local transformations which realize mappings between reaction–diffusion systems. They are $N \times (N-1)$-parameter real matrices which have the sum of entries in each column equal to one.

Some new results concerning two state models are derived:

All annihilation–coagulation systems corresponding to the same parameter Δ' (equation (3.3)) are equivalent. The continuous similarity transformation which acts between these models is given. Results previously derived for the coagulation and annihilation models are generalized to the chemical systems in which both reactions are permitted.

At this point we return to the exciton decay measurements mentioned in the introduction. Further experimental determinations are very desirable for two reasons:

(i) It would be interesting to have accurate measurements of the exciton hopping time which is related to their diffusion constant. The coefficient of the leading term ($\sim t^{-1/2}$) in the exciton concentration decay can then be extracted from the experimental data [9]. Thus one can determinate the ratio of the exciton–exciton annihilation and coagulation rates (3.10).

(ii) Lower errors in the experimental measurements of the luminescence decay would permit the computation of the next to leading term (its time exponent and coefficient) which appears in an expansion of the exciton concentration.

If this term is of order $t^{-3/2}$ (equation (3.11)) the dynamic of exciton propagation and reactions is described by a system which is supersymmetric [3]. This would be the first such system measured experimentally. The other possibility is that the next to leading term is of order t^{-1} (equation (3.10)). In both cases the determination of the leading and next-to-leading term and of the hopping time would permit the computation of the rates of the processes which take place between excitons.

The two-states coagulation–decoagulation, annihilation–creation and death–decoagulation models are equivalent. This enables us to give an exact expression for the particle number of a probabilistic cellular automaton in which modifications of the configuration occur only by starting from particle-vacancy pairs (3.13). The same observable is computed for the model in which the sum of the annihilation and creation probabilities is one (3.16).

A reaction–diffusion system equivalent to the two-species annihilation ($A+B \rightarrow \emptyset+\emptyset$) is found.

We have accurately determined the effect of the reaction rate on the long-time decay of the concentration in the one-species annihilation model. Our results are in excellent agreement with a theoretical conjecture previously made in [11].

We also present quantitative analysis of the asymptotic behaviour of concentration in the three-states annihilation model. Results of simulations performed for different values of the rate of the $A + B \longrightarrow \emptyset + \emptyset$ reaction are presented. They are fully compatible with previous results.

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