Steady-state density in annihilation of immobile reactants with input of particles

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The annihilation reaction $A + A \to 0$ and the coagulation reaction $A + A \to A$ with reaction probability p and the input probability of particles ϵ in a one-dimensional lattice is studied. The behavior of the steady-state density of particles as a function of the parameter $a \equiv \epsilon/p$ is obtained. The limit $a \to 0$ is solved exactly. The solution is derived from a system of linear differential equations for the probabilities of finding n correlative particles Γ_n. The analytical results were confirmed by numerical simulations.

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I. INTRODUCTION

In the past decade much effort has been dedicated to the study of reaction-diffusion systems [1]. These systems show a breakdown of the mean-field approximation and of the standard chemical rate equations in low dimensions (i.e., fractals, multifractals, or one-dimensional systems).

The models of immobile reactants [2–4] have received less attention because generally, unless longer-range reactions are allowed [3], the time dependence of the particle density involves exponential relaxation rather than anomalous power-law behaviors. Nevertheless, besides the dynamics, there is another point of interest which is the steady-state value of the density when there is input of particles.

In the present work the steady-state of annihilation ($A + A \to 0$) and coagulation ($A + A \to A$) reactions with immobile reactants in a one-dimensional lattice are studied. It is considered that when two particles are at first neighbors they react with probability p. The model represents a system surrounded by a gas of particles at constant pressure. So there is a constant input rate of particles adsorbed into the system.

Very recently [4] analytical results have been obtained for annihilation and coagulation reactions between immobile reactants with probability p of reaction. The motivation for the present paper is to introduce the input of particles into the model and to study the steady-state regime.

The input changes the behavior of the system, even when the input rate per lattice site tends to zero. As we will see later, the results obtained for this case are quite different from the case with no input.

II. THE MODEL AND THE EQUATION SYSTEM

We will describe the calculation for the case $A + A \to A$. The method for case $A + A \to 0$ is very similar.

In the model the system evolves with a discretized time t. The one-dimensional lattice has L sites with periodic boundary conditions. Each site of the lattice can be occupied by only one particle or it can be empty. At each time step, one of the L sites is randomly chosen with equal probability $1/L$ and time is increased by $1/L$. If the site is empty, it is occupied by an A particle with probability ϵ. If the chosen site is already occupied by a particle, this particle "looks" at either of the first neighbors with equal probability 1/2; if the neighbor is occupied by another particle, the first one reacts and evaporates with probability p; if not, nothing happens, and the particle remains at its site.

Let us denote the occupation number of a generic site i by s_i. If site i is occupied then $s_i = 1$; otherwise $s_i = 0$. If we have at time t any given configuration $\{s\}$, the occupation probability of site i at time $t + \delta t$ (with $\delta t = 1/L$) is

$$\begin{aligned} P_{i\{s\}}(t+\delta t) = {} & (1-s_{i-1})(1-s_i)(1-s_{i+1})\epsilon/L \circ\circ\circ \\ & +(1-s_{i-1})(1-s_i)s_{i+1}\epsilon/L \circ\circ\bullet \\ & +(1-s_{i-1})s_i(1-s_{i+1}) \circ\bullet\circ \\ & +(1-s_{i-1})s_i s_{i+1}(1-p/2L) \circ\bullet\bullet \\ & +s_{i-1}(1-s_i)(1-s_{i+1})\epsilon/L \bullet\circ\circ \\ & +s_{i-1}(1-s_i)s_{i+1}\epsilon/L \bullet\circ\bullet \\ & +s_{i-1}s_i(1-s_{i+1})(1-p/2L) \bullet\bullet\circ \\ & +s_{i-1}s_i s_{i+1}(1-p/L) \bullet\bullet\bullet . \end{aligned} \tag{1}$$

On the right-hand side we show the configuration of sites $(i-1, i, i+1)$, which corresponds to each term of $P_{i\{s\}}(t+\delta t)$. The symbol $\bullet$ ($\circ$) denotes an occupied (empty) site at time t. Simplifying Eq. (1) and averaging over configurations $\{s\}$ we obtain

$$P_i(t+\delta t) = P_i(t) + \frac{\epsilon}{L} - \frac{\epsilon}{L}P_i(t) - \frac{p}{2L}\langle s_i(s_{i-1}+s_{i+1})\rangle, \tag{2}$$

where $P_i(t) = \langle s_i \rangle$. If a random initial distribution of particles and periodic boundary conditions are used, there

are no privileged sites in the lattice. On average the particle distribution will be uniform for all times. Therefore $P_i = P_j = P$ and $\langle s_i s_{i+1}\rangle = \langle s_j s_{j+1}\rangle$ for all i, j. Let us denote the two-particle correlation $\langle s_i s_{i+1}\rangle$ by Γ_2. Then

$$P(t+\delta t) - P(t) = \frac{\epsilon}{L} - \frac{\epsilon}{L}P(t) - \frac{p}{L}\Gamma_2(t). \tag{3}$$

The occupation probability per lattice site $P(t)$, now independent of i, is equivalent to the global density of particles, which we will denote by $\Gamma_1(t)$. Knowing that $\delta t = 1/L$, in the continuous limit, when $L \to \infty$, we have

$$\frac{d\Gamma_1(t)}{dt} = \epsilon[1 - \Gamma_1(t)] - p\Gamma_2(t). \tag{4}$$

The first term represents the input of particles: the input probability ϵ times the probability of finding an empty site. The second term represents the decrease of $\Gamma_1(t)$ due to the reaction.

In the same manner we can obtain less obvious linear differential equations for the n-particle correlations $\Gamma_n = \langle s_1\, s_2 \cdots s_n\rangle$,

$$\begin{aligned}\frac{d\Gamma_n(t)}{dt} &= \epsilon \sum_{j=1}^{n} \langle s_1 \cdots s_{j-1}(1 - s_j) s_{j+1} \cdots s_n\rangle \\ &\quad -p[(n-1)\Gamma_n(t) + \Gamma_{n+1}(t)] \\ &= \epsilon \sum_{j=1}^{n} (\langle s_1 \cdots s_{j-1}\, s_{j+1} \cdots s_n\rangle - \Gamma_n) \\ &\quad -p[(n-1)\Gamma_n(t) + \Gamma_{n+1}(t)] \end{aligned} \tag{5}$$

for $n \geq 2$. The first term is the input probability ϵ times the probability of finding an incomplete chain of n sites with an empty site. The second term is the reaction term. The first part $[-p(n-1)\Gamma_n(t)]$ is due to the probability of a reaction of one of the $n-1$ bonds of an n chain. If there is a particle in a site adjacent to the n chain, there is an additional bond to consider in the reaction. This case is taken into account by the second part $[-p\Gamma_{n+1}(t)]$. Chains larger than $n+1$ sites are included in this equation and no additional terms with Γ_{n+j} (with $j > 1$) are required. If we consider no input of particles, i.e., $\epsilon = 0$, then Eq. (5) is equal to Eq. (3) in Ref. [4] for a one-dimensional lattice.

For $n = 2$ we have $d\Gamma_2(t)/dt = 2\epsilon[\Gamma_1(t) - \Gamma_2(t)] - p[\Gamma_2(t) + \Gamma_3(t)]$. For $n \geq 3$ we have to make an approximation for $\langle s_1 \cdots s_{j-1}\, s_{j+1} \cdots s_n\rangle$. We can propose as a first step a decorrelation of the product, i.e., $\langle s_1 \cdots s_{j-1}\, s_{j+1} \cdots s_n\rangle \simeq \langle s_1 \cdots s_{j-1}\rangle\langle s_{j+1} \cdots s_n\rangle = \Gamma_{j-1}\Gamma_{n-j}$. This approximation becomes valid when the chain of $j-1$ sites is far from the chain of $n-j$ sites. We consider that it could also be a good approximation to take both chains together instead of one far from the other, i.e., $\langle s_1 \cdots s_{j-1}\, s_{j+1} \cdots s_n\rangle \simeq \langle s_1 \cdots s_{j-1}\, s_j \cdots s_{n-1}\rangle = \Gamma_{n-1}$. This approach has the advantage of producing linear differential equations, so the steady state can be easily obtained. Finally we have

$$\begin{aligned}\frac{d\Gamma_n(t)}{dt} &= n\,\epsilon[\Gamma_{n-1}(t) - \Gamma_n(t)] \\ &\quad - p[(n-1)\Gamma_n(t) + \Gamma_{n+1}(t)] \quad \text{for } n \geq 3. \end{aligned} \tag{6}$$

In the next section we show numerical simulations that support this approximation.

III. THE STEADY STATE

To evaluate the steady-state density Γ_1 we have to solve the equation system

$$\begin{aligned} &a\Gamma_1 + \Gamma_2 = a, \\ &na\Gamma_{n-1} - (na + n - 1)\Gamma_n - \Gamma_{n+1} = 0 \quad \text{for } n \geq 2, \end{aligned} \tag{7}$$

where parameter a is equal to ϵ/p. From Eqs. (7) it can be probed by induction that

$$\Gamma_n = \frac{n!\, a^{n-1}}{y_n} - \frac{y_{n-1}}{y_n}\Gamma_{n+1} \quad \text{for } n \geq 1, \tag{8}$$

where y_n are polynomials in a (except for $n = 0$), defined by

$$y_{n+1} = (n+1)a y_{n-1} + [(n+1)a + n]y_n, \tag{9}$$

with $y_0 = 1/a$ and $y_1 = 1$. Iterating Eq. (8) we have

$$\begin{aligned}\Gamma_n = \frac{n!\, a^{n-1}}{y_n} &+ \sum_{i=n}^{\infty} \frac{(i+1)!\, a^i}{y_{i+1}} \prod_{j=n}^{i} \left(-\frac{y_{j-1}}{y_j}\right) \\ &+ \Gamma_\infty \prod_{i=n}^{\infty} \left(-\frac{y_{i-1}}{y_i}\right). \end{aligned} \tag{10}$$

Due to the annihilation, the correlation Γ_n vanishes when $n \to \infty$. Simplifying Eq. (10) one finally gets the steady-state correlations for the coagulation reaction $A + A \to A$

$$\Gamma_n = y_{n-1} \sum_{i=n-1}^{\infty} (-1)^{i-n+1} \frac{(i+1)!\, a^i}{y_i y_{i+1}}. \tag{11}$$

For $n = 1$ the steady-state density is

$$\Gamma_1 = \frac{1}{a} \sum_{i=0}^{\infty} (-1)^i \frac{(i+1)!\, a^i}{y_i y_{i+1}}. \tag{12}$$

In Fig. 1 we plot Γ_n as a function of a for different values of n. Although Eqs. (4) and (5) are valid only for uniform initial particle distributions, the steady state derived from these equations is valid for all kinds of initial conditions since the input and the annihilation make the particle distribution homogeneous. Therefore, a universal behavior of the Γ_n's, in the sense that they are independent of initial conditions, is obtained. Numerical data are also plotted in Fig. 1 to confirm Eq. (11). The agreement between numerical and analytical data supports the approximation made in Eq. (6) for $n \geq 3$.

The case with no input of particles, i.e., $a = 0$, was solved in Refs. [2] and [4]. If random initial distributions are used, the density behaves as $\Gamma_1(t) = \rho_0 \exp[-\rho_0(1 - e^{-pt})]$, where ρ_0 is the initial density. For long times $\Gamma_1 = \rho_0 e^{-\rho_0}$. In our case, when $a \to 0$, $\Gamma_1 \to 1/3$. For $a \to 0$ the result is completely different from the one for $a = 0$. This means that a very small input of particles is sufficient for the system to forget initial conditions and to reach the steady-state density value which only depends

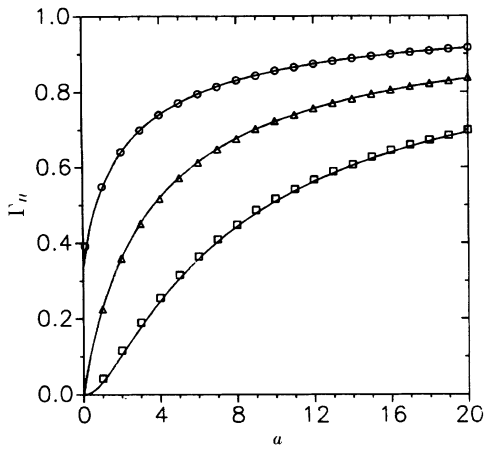

FIG. 1. Plot of Γ_n versus a for the reaction $A + A \to A$. From top to bottom $n = 1$ (the density), 2, and 4. The lines are evaluated from Eq. (11). The points were obtained via numerical simulations. In the simulations a time $t_{\max} = 200$ was enough to reach the steady state; random initial distributions were used with initial density $\rho_0 = 0.5$; a lattice with periodic boundary conditions and size $L = 30\,000$ was used and the results were obtained averaging over ten samples.

on the parameter a. The value $\Gamma_1 = 1/3$ is an exact result independent of the approximation we made for $n \geq 3$ because when $a \to 0$, $\Gamma_n \to 0$ for $n \geq 2$, and the result can be derived from the two equations of (7) (for $n = 1$ and $n = 2$), which are exact.

Repeating the same process for the annihilation reaction $A + A \to 0$, the result for the steady-state density is

$$\Gamma_1' = \frac{1}{a}\sum_{i=0}^{\infty}(-2)^i \frac{(i+1)!\,a^i}{y_i' y_{i+1}'}, \tag{13}$$

where the polynomials y_n' are defined by

$$y_{n+1}' = 2a(n+1)y_{n-1}' + [(n+1)a + n]y_n', \tag{14}$$

with $y_0' = 2/a$ and $y_1' = 1$. Now, in the limit when $a \to 0$, $\Gamma_1' \to 1/5$. As before, the value $1/5$ is exact. As expected, this value is lower than the one corresponding to coagulation reaction.

IV. CONCLUSIONS

We studied the steady-state regime for the coagulation ($A+A \to A$) and annihilation ($A+A \to 0$) reactions with input of particles ($0 \to A$). For both cases approximate results for the n-particle correlations Γ_n were obtained. In particular, when $n = 1$ we have the density of particles. The results were derived from Eqs. (7), a nonhomogeneous equation system. A universal behavior of the Γ_n's, in the sense that they are independent of initial conditions, was obtained. In the limit of small input, $a \to 0$, we obtained the exact value of the steady-state density for both types of reactions.

With the help of a symbolic manipulator computer program such as MATHEMATICA, the method described in Sec. II, for deriving linear differential equations, may be useful for other cases. The method was also used in Ref. [5].

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