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Dynamical disorder in diffusion-limited reactions

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Abstract

We consider a simple model of annihilation reaction, when particles are located on a regular lattice, one of them presenting a fluctuating activity. Time-correlated fluctuations are addressed. The reaction probability and the reaction function are determined exactly. It is shown that the usual, mean-field theory of chemical kinetics does not hold. In particular, the classical law of chemical kinetics does not apply in low dimensions. In three dimensions, a chemical constant exists and it increases with the relaxation frequency of the fluctuations λ. Previously known results are recovered in the limit case of uncorrelated fluctuations, when $\lambda \to \infty$. Different related models, which cannot be solved exactly, are discussed in this limit case. © 2002 Published by Elsevier Science B.V.

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1. Introduction

Natural media are often subject to stochastic time fluctuations, due to their internal evolution or due to their interaction with a changing environment. These fluctuations can modify their reactivity. For instance:

- the microscopic description of an elementary reaction can involve a fluctuating interaction potential, taking into account the stochastic interaction of the reacting molecules with the surrounding fluid;
- random activation or deactivation of a reagent can be caused by external factors (photons, solvent molecules...);
- because of the complexity of biomolecule structures, some geometrical configurations may inhibit a reaction, whereas stochastic changes in the molecule geometries can

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allow it to take place. Such fluctuations often exist for reactions in biomembranes, ligand binding to proteins, or molecular transport in complex molecules, among many examples.

In the present paper, we consider the effects of an activation–deactivation process on the kinetics of a model diffusion-limited annihilation reaction. It is shown that these effects strongly depend on the dimension of the system, and that, generally, they cannot be described by a mean-field theory.

At first, we recall some results concerning a model, ungated diffusion-limited reaction, known from a long time, which are necessary for the following sections. Section 3 addresses the general problem of time-correlated activity fluctuations of one of the reagents. Conclusions and perspectives are discussed in Section 4.

2. Ungated diffusion-limited reactions

2.1. Target annihilation by scavengers randomly moving on a lattice

After the pioneering work of Smoluchowski [1], diffusion-limited reactions have been studied by many authors (see Refs. [2–6], for instance) using diffusion calculations as well as random walks on lattices [6–13], both formalisms giving essentially the same results. Here, we consider a simple annihilation reaction on a lattice, in the absence of a fluctuating reactivity.

2.1.1. Model

A particle A (target) and N_B particles B (scavengers) are located on the N sites of a d-dimensional regular lattice. Each site can contain several particles. Particle A is immobile, whereas particles B perform independent, homogeneous discrete-time random walks on the lattice sites (including sites occupied by other particles): in the simplest model, at integer times $t = 0, 1, \ldots n, \ldots$ each particle B jumps with equal probability on one of its nearest neighbours, but more complex laws could be considered.

Particle A is destroyed as soon as a particle B reaches it.

2.1.2. Survival probability $\Psi(n)$

The probability $\Psi(n)$ that particle A survives up to time n can be computed in the thermodynamic limit N_B, $N \to \infty$ $N_B/N \to b$ concentration of scavengers. It can be shown [7–11] that

$$\Psi(n) = \exp(-bD(n)) , \tag{1}$$

where $D(n)$ is the expectation value of the number of distinct sites $\mathsf{S}(n)$ visited by an n-step random walk (the average being taken on trajectories)

$$D(n) = \langle \mathsf{S}(n) \rangle_\Gamma . \tag{2}$$

2.1.3. Expectation value of the number of distinct sites visited

The expectation value $D(n)$ of the number of distinct sites visited by an n-step random walk depends on the type of the lattice and of the dimensionality. For a d-dimensional Polya random walk, it is known [14] that when $n \to \infty$

$$\text{if } d = 1, \quad D(n) = (8n/\pi)^{1/2} + O(n^{-1/2}), \tag{3.1}$$

$$\text{if } d = 2, \quad D(n) = \pi C n / \log n + O(n/\log^2 n), \tag{3.2}$$

$$\text{if } d = 3, \quad D(n) = nS + O(n^{-1/2}). \tag{3.3}$$

Here C is a constant depending on the lattice. For instance, $C = 4 \times 3^{-3/2}$, 1 and $2 \times 3^{-1/2}$ for hexagonal, square and triangular lattices, respectively.

S is the probability that a B particle never returns to its initial position

S has a well-known, finite value in three dimensions, depending on the lattice, whereas it vanishes in 1 or 2 dimensions. Thus, it can be concluded that

- the ordinary, mean-field kinetics only applies in 3 (or more) dimensions:

  $$\Psi'(n) = \exp(-Sbn), \tag{4}$$

  which corresponds to the chemical reaction constant

  $$k = S. \tag{5}$$

- On the contrary, $\Psi(n)$ does not decay exponentially if $d = 1$ or 2 dimensions: the mean-field theory breaks down for low dimensions.

2.2. Rosenstock trapping model

2.2.1. Model

Following Rosenstock [12], we now consider one single particle A performing a random walk on a lattice containing N_B immobile, randomly placed traps B. Particle A disappears as soon as it reaches one trap B.

2.2.2. Survival probability $\Psi(n)$ in thermodynamic limit

If $N, N_B \to \infty$, $N_B/N = b$, the survival probability of A at time n is given by

$$\Psi(n) = \langle \exp(-b\mathsf{S}(n)) \rangle_\Gamma. \tag{6}$$

Here, $\mathsf{S}(n)$ is the number of distinct sites visited during the n steps trajectory Γ, and the average is taken on trajectories.

The average contained in formula (6) cannot be calculated exactly. The simplest approximation is the Rosenstock approximation:

$$\langle \exp(-b\mathsf{S}(n)) \rangle_\Gamma \approx \exp(-b\langle \mathsf{S}(n) \rangle_\Gamma) \tag{7}$$

or with previous notations

$$\Psi(n) \approx \exp(-bD(n)) \tag{8}$$

which is the same result as for the target annihilation problem. In fact, the right-hand side of (7) is a rigorous lower bound of $\Psi(n)$. It has been shown that approximation (8) is fairly good for intermediate values of N_B [9], because the logarithm is a slowly varying function.

For large values of N_B, however, this approximation is no longer valid. Donsker and Varadhan [15] have shown that if $N_B \to \infty$

$$\Psi(n) \approx \exp(-a_d b^{2/d+2} N_B^{d/d+2}) . \tag{9}$$

3. Stochastically gated reaction with memory

We now consider particles with fluctuating activity, according to the motivations mentioned previously. In the model reactions described in Sections 2.1 and 2.2, either particle A or particles B, or both, can fluctuate between an inert state and an active internal state, which leads to a number of problems corresponding to different physical situations. In the present section, we focus on the case of immobile targets exposed to mobile, fluctuating scavengers, which can be solved explicitly.

3.1. Immobile target and mobile, fluctuating scavengers

Particles: N_A particles A (targets) and N_B particles B (scavengers) are located on the N sites of a d-dimensional regular lattice, each site can contain several particle. Particles A are immobile; particles B perform an independent, homogeneous discrete-time random walk on sites of the lattice (including sites occupied by other particles).

Fluctuations. Particles B fluctuate independently between two internal states: an inert form B^0 (state 0) and an activated form B^1 (state 1)

$$\mathrm{B}^0 \rightleftharpoons \mathrm{B}^1 \tag{10}$$

(which may, for instance, represent the thermal excitation–deexcitation of molecules, or any of the processes mentioned in the introduction).

The waiting times T_0 in state 0 and T_1 in state 1 are independent, stochastic variables obeying the exponential laws

$$P(T_i > t) = \exp(-\lambda_i t) \quad \text{for } i = 0, 1 . \tag{11}$$

For this random telegraph process, the conditional probability $p(j, t+\tau/i, t)$ to find a scavenger B in state j at time $t+\tau$, knowing that it is in state i at time t is

$$p(j, t+\tau/i, t) = p_{ji}(\tau) = (\delta_{ji} - \alpha_i)\mathrm{e}^{-\lambda\tau} + \alpha_j , \tag{12}$$

where δ is the Kronecker symbol, and $\lambda = \lambda_0 + \lambda_1$, $\alpha_j = 1 - \lambda_j/\lambda$.

Interactions. Particles B do not interact with each other. Particles B^0 do not interact with particles A, but if an active particle B^1 meets a A, particle A is destroyed instantaneously, whereas B^1 remains unchanged

$$A + \mathrm{B}^1 \to \mathrm{B}^1 \, . \tag{13}$$

However, it results from Section 2 that the kinetics of this "reaction" is not correctly described by the usual, mean-field reaction kinetics. In fact, because the fluctuations of each B particle are correlated along its trajectory, a complete stochastic theory is needed to compute $\langle N_A(t)\rangle$, average number of A particles at time t.

$\langle N_A(t)\rangle$ is the sum of the probability for each particle A to survive from time 0 up to time t:

$$\langle N_A(t)\rangle = N_A(0)\psi(t) \, , \tag{14}$$

$\psi(t)$ being the probability for a given A to survive up to time t.

3.2. Survival probability of a target with N_B scavengers

From now on, we consider a given target A, and N_B scavengers. If a scavenger B reaches target A in state 0 at time t, and returns to A at a further time $t+\tau$, then the probability that B is then in state i ($i = 0$ or 1) is p_{00} (τ). Thus, to compute the probability that A is annihilated by B at time n, it is necessary to consider the complete trajectory of B from $t = 0$ to n.

Let us label the scavengers by the integers from 1 to N_B and denote them as B_i, $i = 1, 2, \ldots N_B$. Given their trajectories $\{\Gamma_i\}$ $i = 1, 2, \ldots N_B$, the conditional probability that particle B_i is in state 0 at all its visits to A between $t = 0$ and n is

$$P_i(n/\Gamma_i) = \alpha_0 \prod_{h=1}^{m-1} p_{00}(\tau_i^h) \quad \text{if } m > 0 \ (=1 \text{ if } m = 0) \tag{15}$$

with

$$p_{00}(\tau) = \alpha_0 + \alpha_1 \mathrm{e}^{-\lambda\tau} \, . \tag{16}$$

The conditional probability that particle A survives at time n is

$$\psi(n/\{\Gamma_j\}) = \prod_{i=1}^{N_B} P_i(n/\Gamma_i)$$

and since particles B are independent, the survival probability of A at time n, averaged on trajectories, is

$$\psi(n) = \langle \psi(n/\{\Gamma_j\})\rangle = \langle P_1(n/\Gamma)\rangle^{N_B} \tag{17}$$

which permits to compute the survival probability of A from the conditional probability $P_1(n/\Gamma)$ concerning a given particle B.

3.2.1. Thermodynamic limit of survival probability

Let M be the initial position of particle B, and Γ_M a trajectory starting from M at time 0. If the probability of the initial position of B is uniformly distributed among the N sites, then we have

$$\langle P_1(n/\Gamma)\rangle = N^{-1}\sum_M \langle P_1(n/\Gamma_M)\rangle_{\Gamma_M} , \tag{18}$$

where $\langle P_1(n/\Gamma_M)\rangle_{\Gamma_M}$ denotes the average on all trajectories starting from M. Writing

$$\psi(n) = \left(1 - N^{-1}\sum_M (1 - \langle P_1(n/\Gamma_M)\rangle_{\Gamma_M}\rangle)\right)^{N_B} . \tag{19}$$

we obtain, in the thermodynamic limit $N, N_B \to \infty$ and $N_B/N = b$

$$\psi(n) \approx \exp\left(-b\sum_M (1 - \langle P_1(n/\Gamma_M)\rangle_{\Gamma_M}\rangle)\right) , \tag{20}$$

where $1 - \langle P_1(n/\Gamma_M)\rangle$ is the probability that B, starting from M, destroys A at some time $t \leqslant n$.

In the case where particle B is always activated ($\alpha_1 = 1$) $\langle P_1(n/\Gamma_M)\rangle_{\Gamma_M}\rangle$ is the probability $L_M(n)$ that B, starting from M, reaches A at $t \leqslant n$, and $D(n) = \sum_M L_M(n)$ is the average number of distinct sites visited by a B particle during the n-steps trajectory.

3.3. First reaction time and reaction probability

We define the following:

- the probability $Q_M(n)$ that B has never reached A at time n;
- the probability $P_M(n)$ that B reaches A for the first time at time n;

  $$P_M(n) = Q_M(n-1) - Q_M(n) \quad (n > 0)$$

- the similar probabilities $\underline{Q}_M(n)$ and $\underline{P}_M(n)$ for arrival at A of B under activated form, with

  $$\underline{Q}_M(n) = \langle P_1(n/\Gamma_M)\rangle_{\Gamma_M} . \tag{21}$$

As mentioned in Section 2.1, the probabilities $P_M(n)$ and $Q_M(n)$ are known for most random walks and networks in d dimensions. They permit to compute $\underline{Q}_M(n)$ and $\underline{P}_M(n)$ easily. In fact, consider a trajectory Γ_M starting from M at time 0 and reaching A at the successive times $t_i^1, t_i^2, \ldots, t_i^m$ with $\tau_i^h = t_i^{h+1} - t_i^h \geqslant 1$ $(i = 0, \ldots, m-1)$, $\tau_i^m = n - t_i^{m-1} \geqslant 0$.

The probability that B never reaches A under its activated form up to $t = n$ is

$$\underline{Q}_M(n) = Q_M(n) + \sum_{m\geqslant 1}\ \sum_{\tau_0+\tau_1+\cdots+\tau_m=n} \alpha_0 P_M(\tau_0)\, p_{00}(\tau_1) P_A(\tau_1)\ldots \\ \times p_{00}(\tau_{m-1}) P_A(\tau_{m-1}) Q_A(\tau_m)$$

with

$$p_{00}(\tau) = \alpha_0 + \alpha_1 \mathrm{e}^{-\lambda\tau} .$$

The Laplace transform $\hat{\underline{Q}}_M(s)$ of $\underline{Q}_M(n)$ is found to be

$$\hat{\underline{Q}}_M(s) = \hat{Q}_M(s) + \alpha_0 \hat{P}_M(s)[1 - \alpha_0 \hat{P}_A(s) - \alpha_1 \hat{P}_A(s+\lambda)]^{-1} \hat{Q}_A(s) , \tag{22}$$

where $\hat{Q}_A$ and $\hat{P}_A$ refer to the return of particle B to A, starting from A: assuming that the medium is homogeneous we drop index A and obtain one of our main results

$$\hat{\underline{P}}_M(s) = \hat{\underline{P}}_M(s) = \hat{P}_M(s) \left[1 + \frac{\alpha_0}{\alpha_1} \frac{1 - \hat{P}(s)}{1 - \hat{P}(s+\lambda)} \right]^{-1} . \tag{23}$$

3.3.1. Reaction probability

$\underline{P}_M \equiv \hat{\underline{P}}_M(0)$ is the reaction (or annihilation) probability, i.e. the probability that B will ever destroy A. On the other hand, we notice that

$P_M \equiv \hat{P}_M(0)$ is the probability that B, starting from M, will ever reach A, and $\hat{P}(0) = \sum P(n) \equiv R \equiv 1 - S$ is the probability that a B particle will ever return to its initial position, whereas S is the probability that it will never returns.

Taking $s = 0$ in the previous formula, we see that

(i) *in one and two dimensions*, where $R = P_M = 1$, we have $\underline{P}_M = 1$: the reaction occurs with probability 1, independent of the internal fluctuations of particle B.

(ii) *in three* (*or more*) *dimensions*, $P_M = R = 1 - S$ and

$$\frac{P_M}{\underline{P}_M} = 1 + \frac{\alpha_0}{\alpha_1} \frac{S}{1 - \hat{P}(\lambda)} . \tag{24}$$

As a result, $\hat{P}(\lambda)$ decreases with $\lambda, \underline{P}_M$ is an increasing function of the internal relaxation frequency: when λ increases from 0 to ∞, the reaction probability $\underline{P}_M$ increases from $\alpha_1 P_M$ to $\alpha_1(\alpha_1 + \alpha_0 S)^{-1} P_M$ independent of the dimension $d \geqslant 3$ and of the type of the lattice.

A similar conclusion was obtained [16] in a very different model of reaction with a fluctuating reaction potential (infinite dichotomic barriers).

3.3.2. Limit cases

The limit cases of 0 and infinite relaxation frequency λ are important in practice. They permit to recover results obtained previously in simpler theories:

Frozen disorder: If the relaxation frequency λ tends to 0, the internal state of particle B does not change with time. Then

$$\hat{P}(\lambda) \to S \quad \text{and} \quad \underline{P}_M = \alpha_1 P_M$$

which is obvious: if B is activated, then annihilation occurs at the first encounter of B with A, whereas it never occurs if B is inert.

Fluctuations without memory: If the relaxation frequencies λ_0 and λ_1 become infinite, then $\hat{P}(\lambda) \to 0$ and we recover the case of fluctuations without memory [17], further discussed in Section 4, for which

$$\frac{P_M}{\underline{P}_M} = 1 + \frac{\alpha_0}{\alpha_1} S \,.$$

3.4. Reaction kinetics

The kinetics of the annihilation reaction is defined by the survival probability $\psi(n)$ and formula (20)

$$\psi(n) = \exp(-b\underline{D}(n)) \,, \tag{25}$$

$\underline{D}(n)$ being the "integrated reaction function"

$$\underline{D}(n) = \sum_M (1 - \underline{Q}_M(n)) \,. \tag{26}$$

The usual reaction function $\underline{k}(n)$ can be defined by

$$\underline{k}(n) = -\frac{1}{b} \frac{\partial}{\partial n} \ln \psi(n) = \underline{D}(n) - \underline{D}(n-1) \tag{27}$$

and is related to the reaction probability by

$$\underline{k}(n) = \sum_M (\underline{Q}_M(n-1) - \underline{Q}_M(n)) = \sum_M \underline{P}_M(n) \,. \tag{28}$$

Its Laplace transform is

$$\hat{\underline{k}}(s) = \hat{k}(s) \left[1 + \frac{\alpha_0}{\alpha_1} \frac{1 - \hat{P}(s)}{1 - P(s+\lambda)} \right]^{-1} \,, \tag{29}$$

$\hat{k}(s)$ being the Laplace transform of the reaction constant $k(n)$ in the absence of internal fluctuations ($\alpha_0 = 1$). It can be noticed that $k(n)$ is the probability that at its nth step the B particle visits a site which was never visited previously.

3.4.1. Results

- *in one or two dimensions*, $\hat{\underline{k}}(s) \approx \hat{k}(s)$ if $s \to 0$: thus, $\underline{k}(n)$ has the same asymptotic behavior as $k(n)$ when $n \to \infty$: neither k nor $\underline{k}$ tends to a finite limit when $n \to \infty$: there is no reaction constant, and the ordinary law of chemical kinetics does not apply.
- *in three* (*or more*) *dimensions*,

  $$\hat{\underline{k}}(s) \sim S s^{-1} \left[1 + \frac{\alpha_0}{\alpha_1} \frac{S}{1 - \hat{P}(\lambda)} \right]^{-1} \tag{30}$$

  when

  $$s \to 0 \,,$$

which implies that if $n \to \infty$, $\underline{k}(n)$ tends to a constant $\underline{k}$, which is the usual reaction constant

$$\underline{k}(n) \to \underline{k} = S\left[1 + \frac{\alpha_0}{\alpha_1}\frac{S}{1 - \hat{P}(\lambda)}\right]^{-1}$$

or

$$\frac{1}{\underline{k}} = \frac{1}{S} + \frac{\alpha_0}{\alpha_1}\frac{1}{1 - \hat{P}(\lambda)}\,, \tag{31}$$

where $\hat{P}(s)$ is the Laplace-transformed probability of first return of scavenger B to its initial position at step n, $S = 1 - \hat{P}(0)$ is the probability that B never returns to its initial position.

Thus, like the reaction probability, the reaction constant $\underline{k}$ increases with the relaxation frequency λ: if λ increases from 0 to ∞, then $\underline{k}$ increases from $\alpha_1 S$ to $\alpha_1 S(\alpha_1 + \alpha_0 S)^{-1}$.

This result can be understood intuitively. In fact, if a scavenger B reaches A under inert form, then it has a large probability to return to A after a few steps: if the relaxation frequency of B is low, then it has a high probability to be still inactive at these returns and the reaction rate will be low.

3.4.2. Limit cases

- *Frozen disorder*: when $\lambda \to 0$, $\underline{k} \to \alpha_1 k = \alpha_1 S$, which amounts to multiplying the concentration of B by α_1, as it should be.
- *Uncorrelated time fluctuations*: when $\lambda \to \infty$, $\hat{P}(\lambda) \to 0$ and we obtain an inverse addition law for the rate constants

$$\frac{1}{\underline{k}} = \frac{1}{S} + \frac{\alpha_0}{\alpha_1}\,. \tag{32}$$

Such "inverse addition laws" for rate constants are frequently obtained for diffusion-limited reactions, or more generally for chain processes (reaction can only be completed by performing a number of successive steps).

4. Other models of stochastically gated reactions: uncorrelated time fluctuations

It has been observed that other fluctuation schemes can be studied in the model annihilation reaction of a particle A by particles B. For instance, either A or B can move, and either A or B can fluctuate. Only the case treated in Section 3 can be solved analytically, even in the limit of uncorrelated time fluctuations. However, a number of approximate results or exact bounds have been obtained in this limit case [17], and we now focus on this situation. It has been shown in particular that:

at moderate times n, the Rosenstock approximation can be used, then all the models have the same behaviour, and they are insensitive to the fact as to which species is mobile. This behaviour is essentially the same as for ungated reactions, but corrections do depend on the probability α_0 that an encounter is reactive;

at large n, different models have different behaviours:

- the integral reaction rate $\underline{D}(n)$, defined by (25), is smaller when A is fluctuating and B is ungated, than in the contrary situation, either when A is immobile or when A is mobile (but only one kind of particle moves, while another kind fluctuates);
- exact lower and upper bounds for $\underline{D}(n)$ have been determined for each model;
- they show that the asymptotic dependence $\underline{D}(n)$ on n is the same as for ungated case, but the prefactors depend on the precise model.

These points remain to be considered in the case of correlated fluctuations.

5. Perspectives

The previous studies should be extended in many directions. In particular, correlated fluctuations should be addressed for all models mentioned in the previous section, and more general ones, considering for instance that both chemical species can move and fluctuate. Obviously, time correlations make it necessary to consider the trajectories of all moving particles, and calculations can become very intricate.

On the other hand, space correlations between particles may have a very important role, and they should also be considered.

It should be pointed out that the simple processes studied previously can also represent

- other elementary reactions, for instance

  $$\mathrm{B}^0 + C \rightleftharpoons \mathrm{B}^1 ,$$

  $$A + \mathrm{B}^1 \to \mathrm{B}^1 + D ,$$

  where B^1 is a catalyst for the production of D, whereas B^1 is produced reversibly by random interactions of the inert molecules B^0 with particles C, maintained at a fixed concentration.
- interaction processes in different fields, such as population dynamics (as implied by the name “scavengers” used for particle B).

Thus, although these processes are rather simple, they can be used, in a first approximation, for modelling some complex phenomena, which would be untreatable in a realistic description. This may be the case, in particular, in biophysics, where media are likely to fluctuate and, on the other hand, reactions can often occur in low dimension media. Thus a stochastic theory of their reactivity is necessary, and simple models like the models mentioned previously, or more elaborate ones, can be especially useful.

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