Pascal principle for diffusion-controlled trapping reactions

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In this paper, we analyze the long-time behavior of the survival probability $P_A(t)$ of an A particle, that performs lattice random walk in the presence of randomly moving traps B. We show that for both perfect and imperfect trapping reactions, for arbitrary spatial dimension d and for a rather general class of random walks, $P_A(t)$ is less than or equal to the survival probability of an *immobile* target A in the presence of randomly moving traps.

Pascal has once asserted that all misfortune of man comes from the fact that he does not stay peacefully in his room [1]. Taking this statement out of its philosophical context, it might be tempting to evoke it as the "Pascal principle" in regard to the problem of survival of an A particle, which moves randomly in a sea of randomly moving traps B (presented at mean density "b"), and is annihilated upon the first encounter with any of them. This complex problem, which is intimately related to many fundamental problems of statistical physics, has been attracting a great deal of attention within the last three decades (see, e.g., Refs. [2–4], and references therein). Many important results have been obtained but an exact solution is lacking as yet.

In a recent paper [5], which focused on the behavior in the particular case when both A and B's perform conventional diffusive motion, it has been claimed that it is intuitively clear that when the traps are initially (statistically) symmetrically placed with respect to an A particle, the latter will on average survive longer if it stays still than if it diffuses. This represents, if true, a clear illustration of the Pascal principle, and implies that the A particle's survival probability $P_A(t)$ obeys the inequality

$$P_A(t) \leqslant P_A'(t), \tag{1}$$

where $P_A'(t)$ stands for the corresponding survival probability of an *immobile* target A in the presence of diffusive traps. The latter can be evaluated exactly [2,6]. Furthermore, following the reasonings of earlier works [2,7], the authors of Ref. [5] constructed a lower bound on $P_A(t)$ and showed that these bonds converge as $t \to \infty$ for systems of spatial dimension $d \leqslant 2$, defining thus the large-t asymptotic form of $P_A(t)$ exactly. Subsequently, in Ref. [9], the arguments of Ref. [5] have been generalized to arbitrary symmetric random motion with particles' and traps' trajectories characterized by a fractal dimension d_ω (not necessarily equal to 2, which is specific for conventional diffusive motion), and here exact large-t asymptotic forms of $P_A(t)$ have been obtained for systems of spatial dimension $d \leqslant d_\omega$.

We note that the inequality in Eq. (1) has been derived previously for the process of hopping transport of an excitation on a disordered array of immobile donor centers in the presence of randomly placed, immobile quenchers [10]. On the other hand, Eq. (1) is compatible with recent results on

ballistic $A + A \to 0$ annihilation process [11]. For trapping $A + B \to B$ reactions (TR) involving *diffusive* species the authors of Ref. [5] were unable, however, to prove the inequality in Eq. (1), but furnished only some arguments in favor of it. Consequently, results on *exact* asymptotic behavior of $P_A(t)$ depend crucially on whether the inequality in Eq. (1) is indeed correct.

In this paper, we analyze, in the lattice formulation of the model, the Pascal-principle-like inequality in Eq. (1). Following the line of argument of Refs. [6,8], we show that for both instantaneous (perfect) and imperfect TRs, for arbitrary spatial dimension d, and for a rather general class of random walks (not necessarily conventional diffusion), $P_A(t)$ of a mobile A particle in the presence of mobile traps is less than the survival probability of an immobile target A in the presence of mobile Bs. Our proof assumes essentially that B's perform a space and time homogeneous, unbiased jump process on the lattice sites. Behavior in the continuous-space systems, which requires a somewhat more delicate analyzis (especially for $d \geqslant 2$), will be presented elsewhere [12].

Consider a d-dimensional hypercubic lattice containing M sites. A single A particle is initially located at the origin, while N traps B are placed on the lattice at positions $\mathbf{Y}_0^{(i)} \neq 0$, where the superscript i here and henceforth numerates the traps, $i = 1, \ldots, N$.

In regard to particles dynamics, we suppose that the A particle performs a continuous-time jump process on the lattice sites and that the time interval between the consecutive jumps is a random variable. We denote then as Γ_A the A particle trajectory recorded at integer time moments $k = 0, \ldots, n$, such that $\Gamma_A = \{\mathbf{X}_0 = 0, \mathbf{X}_1, \ldots, \mathbf{X}_n\}$, where $\mathbf{X}_k$ is the vector of a lattice site at which the A particle resides at time moment k. Note that since the time interval between the consecutive jumps is a random variable, two successive positions are not necessarily different and not necessarily nearest neighbors.

Now, we suppose that the B's perform identical and independent *discrete time random walks*: that is, at each tick of the clock each B can jump with a given probability from a lattice site $\mathbf{Y}$ to another (not necessarily neighboring) site $\mathbf{Y}'$, or it may also remain at $\mathbf{Y}$. We define then as $\Gamma_B^{(i)}$ the trajectory of the ith B particle, $\Gamma_B^{(i)} = \{\mathbf{Y}_0^{(i)}, \mathbf{Y}_1^{(i)}, \ldots, \mathbf{Y}_n^{(i)}\}$, where $\mathbf{Y}_k^{(i)}$ denotes the position of the ith trap at time moment k, $k = 0, 1, \ldots, n$.

Next, let $P(\mathbf{Y}_n^{(i)}|\mathbf{Y}_0^{(i)})$ be the conditional probability of finding the ith trap B at site $\mathbf{Y}_n^{(i)}$ at time moment n, knowing that it started its random walk at $\mathbf{Y}_0^{(i)}$. We assume now that random walks executed by the B particles satisfy the following, quite general conditions.

(i) the random walk is space and time homogeneous:

$$P(\mathbf{Y}_n^{(i)}|\mathbf{Y}_0^{(i)}) = P(\mathbf{Y}_n^{(i)} - \mathbf{Y}_0^{(i)}|0), \qquad (2)$$

(ii) at any time moment n and for any i [13],

$$P(\mathbf{Y}_n^{(i)} \neq \mathbf{Y}_0^{(i)}|\mathbf{Y}_0^{(i)}) \leq P(\mathbf{Y}_n^{(i)} = \mathbf{Y}_0^{(i)}|\mathbf{Y}_0^{(i)}) \equiv R_n, \qquad (3)$$

i.e., the conditional probability $P(\mathbf{Y}_n^{(i)} \neq \mathbf{Y}_0^{(i)}|\mathbf{Y}_0^{(i)})$ of finding at time moment n the ith trap at site $\mathbf{Y}_n^{(i)}$ different from its starting point $\mathbf{Y}_0^{(i)}$, is less than or equal to the probability $P(\mathbf{Y}_n^{(i)} = \mathbf{Y}_0^{(i)}|\mathbf{Y}_0^{(i)})$ of finding it at time moment n exactly at the starting point; here, R_n denotes the return probability of the random walk executed by the traps. By convention, $R_0 \equiv 1$.

Finally, we consider two situations with respect to reaction; namely, when (a) the A particle gets annihilated with probability $p = 1$ upon the first encounter with any of B's (perfect reaction) and (b) when the annihilation of the A particle takes place with probability $p < 1$ when an A and any of B's occur at the same site (imperfect reaction). For computational convenience, we stipulate that for both situations reaction can take place only at integer time moments; that is, if at a noninteger time A jumps on a site which is occupied by any B, A survives till the departure of this particle or an arrival of another B. The probability that both A and B particles jump on the same site simultaneously is clearly equal to 0.

Let $P_n^{(i)}(\Gamma_A|\mathbf{Y}_0^{(i)})$ denote *the conditional probability* that for a given realization of the A particle trajectory Γ_A, the ith B particle starting its walk from the point $\mathbf{Y}_0^{(i)}$ *does not destroy* (encounter) A *up to time n*. Since B's move and act independently of each other, the conditional probability $\Psi_n(\Gamma_A|\{\mathbf{Y}_0^{(i)}\})$ that, for a given realization of the A particle trajectory Γ_A and a given set of the starting points $\{\mathbf{Y}_0^{(i)}\}$, the A particle survives up to time n, is determined by

$$\Psi_n(\Gamma_A|\{\mathbf{Y}_0^{(i)}\}) = \prod_{i=1}^{N} P_n^{(i)}(\Gamma_A|\mathbf{Y}_0^{(i)}), \qquad (4)$$

and hence, the A particle survival probability obeys

$$P_A(n) = \langle\langle\Psi_n(\Gamma_A|\{\mathbf{Y}_0^{(i)}\})\rangle_{\{\mathbf{Y}_0^{(i)}\}}\rangle_{\Gamma_A}, \qquad (5)$$

the average being taken first over the starting points of B's and then over all possible trajectories Γ_A.

Now, supposing that B's were initially uniformly distributed on the lattice (excluding the origin) and dropping the superscript "i," one finds from Eqs. (4) and (5) that

$$P_A(n) = \left\langle \left\{ \frac{1}{M} \sum_{\mathbf{Y}_0 \neq 0} P_n(\Gamma_A|\mathbf{Y}_0) \right\}^N \right\rangle_{\Gamma_A}$$

$$= \left\langle \left\{ 1 - \frac{1}{M} \sum_{\mathbf{Y}_0 \neq 0} [1 - P_n(\Gamma_A|\mathbf{Y}_0)] \right\}^N \right\rangle_{\Gamma_A}. \qquad (6)$$

Turning next to the thermodynamic limit, i.e., setting $N, M = \infty$ with a fixed ratio $b = N/M$, we find

$$P_A(n) = \left\langle \exp\left\{ -b \sum_{\mathbf{Y}_0 \neq 0} [1 - P_n(\Gamma_A|\mathbf{Y}_0)] \right\} \right\rangle_{\Gamma_A}. \qquad (7)$$

Consequently, the survival probability $P_A(n)$ can be thought of as the generating function of the probability

$$Q_n(\Gamma_A|\mathbf{Y}_0) = 1 - P_n(\Gamma_A|\mathbf{Y}_0) \qquad (8)$$

that for a given Γ_A, a single B, being at $\mathbf{Y}_0 \neq 0$ at $n = 0$, destroys the A at some time moment $\leq n$. Note also that when the A particle is immobile, Eq. (7) reduces to

$$P_A'(n) = \exp\left\{ -b \sum_{\mathbf{Y}_0 \neq 0} [1 - P_n(0|\mathbf{Y}_0)] \right\}, \qquad (9)$$

which can be evaluated explicitly [2,6].

We seek now an upper bound on the survival probability in Eq. (7). Let $F_k(\Gamma_A|\mathbf{Y}_0)$ denote *the conditional probability that* a single B particle, being at $\mathbf{Y}_0$ at $k = 0$, encounters A for the first time at time moment k, given the A particle trajectory Γ_A is fixed. Then, the conditional probability $Q_n(\Gamma_A|\mathbf{Y}_0)$ that a single B particle, starting from $\mathbf{Y}_0$, destroys A at or before $k = n$ obeys

$$Q_n(\Gamma_A|\mathbf{Y}_0) = \sum_{0 \leq k \leq n} F_k(\Gamma_A|\mathbf{Y}_0). \qquad (10)$$

Now, the conditional probability that the trajectory of B (extended after the possible annihilation of A) meets Γ_A at time n (not necessarily for the first time) is clearly

$$P(\mathbf{Y}_n = \mathbf{X}_n|\mathbf{Y}_0) = F_n(\Gamma_A|\mathbf{Y}_0)$$

$$+ \sum_{0 \leq k < n} P(\mathbf{Y}_n = \mathbf{X}_n|\mathbf{Y}_k) F_k(\Gamma_A|\mathbf{Y}_0). \qquad (11)$$

Summing both sides of the last equation over all initial positions $\mathbf{Y}_0 \neq 0$, we obtain

$$\sum_{\mathbf{Y}_0 \neq 0} P(\mathbf{Y}_n = \mathbf{X}_n|\mathbf{Y}_0) = K_n(\Gamma_A)$$

$$+ \sum_{0 \leq k < n} P(\mathbf{Y}_n = \mathbf{X}_n|\mathbf{Y}_k) K_k(\Gamma_A), \qquad (12)$$

where $K_n(\Gamma_A)$ is defined as

$$K_n(\Gamma_A) = \sum_{\mathbf{Y}_0 \neq 0} F_n(\Gamma_A | \mathbf{Y}_0). \tag{13}$$

We note that $K_n(\Gamma_A)$ has a meaning of a time-dependent reaction rate; using Eq. (13), we rewrite Eq. (7) as

$$P_A(n) = \left\langle \exp\left\{ -b \sum_{0 \leq k \leq n} K_k(\Gamma_A) \right\} \right\rangle_{\Gamma_A}. \tag{14}$$

On the other hand, the survival probability $P'_A(n)$ of an *immobile A* particle, Eq. (9), can be written as

$$P'_A(n) = \exp\left\{ -b \sum_{0 \leq k \leq n} K_k \right\} = \exp\{-b(S_n - 1)\}, \tag{15}$$

where $K_k \equiv K_k(\Gamma_A \equiv 0)$, while S_n is the expected number of distinct sites visited by a single B up to time moment n (see, e.g., Ref. [14] for more details). The last quantity is obtained directly by inversion of its generating function $\hat{S} = \sum_{n=0}^{\infty} S_n \xi^n$, which can be evaluated explicitly, $\hat{S} = (1 - \xi)^{-2} \hat{R}^{-1}$, $\hat{R} = \sum_{n=0}^{\infty} R_n \xi^n$ being the generating function of the return probability R_n.

We turn now to the comparison of $K_k(\Gamma_A)$ and K_k. Using the normalization $\sum_{\mathbf{Y}_0} P(\mathbf{Y}_n = \mathbf{X}_n | \mathbf{Y}_0) = 1$, and the condition (i), we have $\sum_{\mathbf{Y}_0 \neq 0} P(\mathbf{Y}_n = \mathbf{X}_n | \mathbf{Y}_0) = 1 - P(\mathbf{Y}_n = \mathbf{X}_n | 0)$. Consequently, by virtue of (ii),

$$\sum_{\mathbf{Y}_0 \neq 0} P(\mathbf{Y}_n = \mathbf{X}_n | \mathbf{Y}_0) \geq 1 - R_n. \tag{16}$$

Now, from Eqs. (12) and (16), we get

$$K_n(\Gamma_A) + \sum_{0 \leq k < n} P(\mathbf{Y}_n = \mathbf{X}_n | \mathbf{Y}_k) K_k(\Gamma_A) \geq 1 - R_n. \tag{17}$$

On the other hand, the inequality in Eq. (3) implies that

$$P(\mathbf{Y}_n = \mathbf{X}_n | \mathbf{Y}_k) \leq R_{n-k}. \tag{18}$$

Recollecting that $R_0 = 1$ and making use of Eq. (18), we thus enhance the inequality in Eq. (17), which now reads

$$\sum_{0 \leq k \leq n} R_{n-k} K_k(\Gamma_A) \geq 1 - R_n, \tag{19}$$

and also becomes an equality when A is *immobile*.

Further on, multiplying both sides of Eq. (19) by ξ^n and performing summation over n, we get

$$\hat{K}(\Gamma_A) = \sum_{n=0}^{\infty} K_n(\Gamma_A) \xi^n \geq \frac{1}{(1 - \xi)\hat{R}} - 1. \tag{20}$$

Next, taking into account that

$$\hat{S}(\Gamma_A) = \sum_{n=0}^{\infty} \xi^n \left(\sum_{0 \leq k \leq n} K_k(\Gamma_A) \right) = \frac{\hat{K}(\Gamma_A)}{1 - \xi}, \tag{21}$$

we find from Eq. (20) the following inequality:

$$\hat{S}(\Gamma_A) \geq \frac{1}{(1 - \xi)^2 \hat{R}} - \frac{1}{1 - \xi} \equiv \sum_{n=0}^{\infty} \xi^n \left(\sum_{0 \leq k \leq n} K_k \right), \tag{22}$$

which implies that the generating function of the expression in the exponent in Eq. (14), describing the A particle survival probability in case when it "leaves the room" and changes its position with time, is always greater than or equal to the generating function of the expression in the exponent in Eq. (15), which applies to the case when the A particle stays peacefully at its initial position.

Hence, turning to the limit $\xi \to 1^-$ ($n \to \infty$) and making use of the Tauberian theorems [15], we arrive at the conclusion that the desired inequality in Eq. (1) holds in the limit $n \to \infty$. As a matter of fact, it can be shown that this inequality holds generally for arbitrary finite n; the proof in this statement is, however, rather cumbersome and will be presented elsewhere [12].

Finally, we briefly outline the steps involved in the derivation of Eq. (1) in the general case when reaction between an A and any of B's is not instantaneous, but takes place with some finite probability p. Following Ref. [14], we suppose that here each trap bears "a gate," which may be either open or closed; in the former case the trap is reactive and annihilates the A particle upon the encounter, while in the latter case it is inert with respect to reaction. The state of the gate on the ith trap is characterized by a random variable ζ_i such that $\zeta_i = 1$ (open gate) with probability p, and $\zeta_i = 0$ (closed gate) with the probability $1 - p$, respectively. Each ζ_i updates its state at each tick of the clock; the updating process proceeds completely at random, without memory in time and without correlations with the gates imposed on other B particles. As shown in Ref. [14], such a model with stochastic, two-state gates corresponds to situations in which the elementary reaction act is characterized by a finite intrinsic reaction constant $K_{el} = p/(1 - p)$.

Now, we notice that $P_A(n)$ in this case can be still written in the form of Eq. (7) with $Q_n(\Gamma_A | \mathbf{Y}_0)$ defined by Eqs. (8) and (10), but here $F_k(\Gamma_A | \mathbf{Y}_0) = F_k^{(p)}(\Gamma_A | \mathbf{Y}_0)$ should be interpreted as the conditional probability that the B particle encounters the A particle for the first time at time moment k exactly and moreover, that at this moment of time the B particle is in reactive state; the superscript "(p)" will signify that here we deal with imperfect TR. Further on, let $P^{(p)}(\mathbf{Y}_n = \mathbf{X}_n | \mathbf{Y}_0)$ be the conditional probability that the trajectory of B meets Γ_A at time n (not necessarily for the first time) and at this time moment B is in reactive state. For the model under study, such a probability obeys

$$P^{(p)}(\mathbf{Y}_n = \mathbf{X}_n | \mathbf{Y}_0) = p P(\mathbf{Y}_n = \mathbf{X}_n | \mathbf{Y}_0) \tag{23}$$

and

$$P^{(p)}(\mathbf{Y}_n = \mathbf{X}_n | \mathbf{Y}_0) = F_n(\Gamma_A | \mathbf{Y}_0)$$
$$+ \sum_{0 \leq k < n} P^{(p)}(\mathbf{Y}_n = \mathbf{X}_n | \mathbf{Y}_k) F_k(\Gamma_A | \mathbf{Y}_0). \tag{24}$$

Summing both sides of Eq. (24) over $\mathbf{Y}_0 \neq 0$, we get

$$\sum_{\mathbf{Y}_0 \neq 0} P^{(p)}(\mathbf{Y}_n = \mathbf{X}_n | \mathbf{Y}_0) = 0) \geq p(1 - R_n), \qquad (25)$$

and hence, we find

$$(1 - R_n) \leq K_n^{(p)}(\Gamma_A) + p \sum_{0 \leq k < n} R_{n-k} K_k^{(p)}(\Gamma_A), \qquad (26)$$

where $K_n^{(p)} = \Sigma_{\mathbf{Y}_0 \neq 0} F_n^{(p)}(\Gamma_A | \mathbf{Y}_0)$. Multiplying both sides of Eq. (26) by ξ^n and then summing it over n, $n = 0, \dots, \infty$, we arrive at the following inequality:

$$\hat{S}^{(p)}(\Gamma_0) \geq \frac{p[1 - (1-\xi)\hat{R}]}{(1-\xi)^2(1 - p + p\hat{R})}, \qquad (27)$$

where $\hat{S}^{(p)}(\Gamma_A)$ is the generating function of the sum $\Sigma_{0 \leq k \leq n} K^{(p)}(\Gamma_A) = b^{-1} \ln[1/P_A^{(p)}(n)]$, $P_A^{(p)}(n)$ being the A particle survival probability for imperfect TR.

Turning to the limit $\xi \to 1^-$ ($n \to \infty$), we notice that here $(1-\xi)\hat{R} \ll 1$ and hence, in this limit only the first term in the square bracket matters. On the other hand, this leading term $\hat{S}^{(p)} = p(1-\xi)^{-2}/(1 - p + p\hat{R})$ coincides *exactly* with the expression obtained earlier [14] for the generating function of the exponent of the survival probability of an immobile target A in the presence of stochastically gated traps. The Tauberian theorem [15] then insures that also in this general case of imperfect TR the inequality in Eq. (1) holds as $n \to \infty$.

To conclude, we have proven here that in the long-time limit the survival probability of an A particle performing random walk on the sites of a d-dimensional lattice in the presence of randomly moving traps is less than or equal to the survival probability of an immobile A particle in the presence of randomly moving traps. This result holds for quite a general class of random walks as well as for perfect and imperfect trapping reactions.

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[13] We remark that Eq. (3) is not exactly satisfied for the discrete time, strictly nearest-neighbor Polya random walks, since in this case $R_n \equiv 0$ for odd values of n and $\mathbf{Y}_n$. On the other hand, one easily obtains random walks satisfying the inequality in Eq. (3) by considering, for instance, a modified Polya walk in which at each step the particle has a probability p_0 to remain at its position; Eq. (3) holds here if $p_0 \geq 1/2$. More general arguments including the case of standard Polya walks will be presented in Ref. [12].

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