Survival probability and order statistics of diffusion on disordered media

L. Acedo† and S. B. Yuste†
Departamento de Física, Universidad de Extremadura, E-06071 Badajoz, Spain
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We investigate the first passage time \( t_{j,N} \) to a given chemical or Euclidean distance of the first \( j \) of a set of \( N \gg 1 \) independent random walkers all initially placed on a site of a disordered medium. To solve this order-statistics problem we assume that, for short times, the survival probability (the probability that a single random walker is not absorbed by a hyperspherical surface during some time interval) decays for disordered media in the same way as for Euclidean and some class of deterministic fractal lattices. This conjecture is checked by simulation on the incipient percolation aggregate embedded in two dimensions. Arbitrary moments of \( t_{j,N} \) are expressed in terms of an asymptotic series in powers of \( 1/\ln N \) which is formally identical to those found for Euclidean and (some class of) deterministic fractal lattices. The agreement of the asymptotic expressions with simulation results for the two-dimensional percolation aggregate is good when the boundary is defined in terms of the chemical distance. The agreement worsens slightly when the Euclidean distance is used.

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I. INTRODUCTION

Diffusion in disordered media has been an area of intensive research during the last two decades [1,3]. Transport in non-crystalline, disordered materials cannot be explained by the classical theories of diffusion since anomalous behaviors (relative to what happens in the Euclidean domain) are the rule here. In particular, the mean-square displacement of a random walker \( \langle r^2(t) \rangle \) is no longer proportional to the time \( t \) as occurs in uniform Euclidean systems of any dimension (Fick’s Law), but the more general anomalous diffusion law \( \langle r^2 \rangle \sim 2Dt^{2/d_w} \) holds for large times, \( r \) being the Euclidean distance from the position of the random walker at \( t = 0 \), \( d_w \) the anomalous diffusion exponent, and \( D \) the diffusion constant. As the geometrical structure of real disordered media is very complex and varied, it is usually modeled by stochastic fractal lattices. Of these, the incipient percolation aggregate embedded in either two or three dimensions is the most widely used [1,2,4,5,6].

Statistical problems related to a single random walker have traditionally been the subject of more intensive research than those corresponding to \( N > 1 \) interacting or independent random walkers [1,7]. Of course, problems in which the walkers interact cannot be analyzed in terms of the single-walker theory. However, there also exist other multiparticle problems that can not be analyzed in terms of the single-walker theory even though the walkers are independent. These problems have begun to be the target of in-depth studies during the last decades [5,7,8,9,10,11,12,13,14,15,16]. Particular attention has been paid to (i) the evaluation of the average number of distinct sites visited up to time \( t \) by \( N \) independent random walkers all starting from the same origin in both Euclidean and fractal lattices [5,7,8,9,10,11,12,13,14,15,16], and (ii) the description of the order statistic of the diffusion processes. This is the subject we address in this paper and can be stated as follows. A set of \( N \) independent random walkers all initially placed at a given site (the origin) of a medium start to diffuse at time \( t = 0 \). Eventually, at time \( t_{1,N}(z) \) a random walker of this set reaches for the first time a site separated from the origin by the distance \( z \). Next, a second random walker reaches at time \( t_{2,N} \) a site at the same distance \( z \) from the origin, and so on [14]. Equivalently, we can understand \( t_{j,N}(z) \) as the time taken (escape time or lifetime) by the \( j \)th particle out of a set of \( N \gg 1 \) to escape from a “spherical” region of radius \( z \) centered at the starting site of diffusion. Our goal in this paper is to calculate the escape-time moments \( \langle t_{j,N}^p(z) \rangle \) when \( N \gg 1 \) random walkers diffuse in a disordered medium.

This order-statistic problem was solved for the one-dimensional lattice in Ref. [13,14] and for some class of deterministic fractal substrates in Ref. [14]. For \( d \)-dimensional Euclidean lattices, the form of the first moment of \( t_{1,N} \) was guessed in Ref. [13] and checked using simulations results for \( d = 2 \) and \( d = 3 \). The full solution of this problem for Euclidean media and for arbitrary \( p \) and \( j \) has been obtained recently [15,16]. For all these media, the \( p \)th moment of the time \( t_{j,N} \) spend by the \( j \)th random walker in reaching the Euclidean distance \( r \) was given in terms of an asymptotic series for large \( N \) of the form

\[
\langle t_{j,N}^p(z) \rangle \sim \left( \frac{r}{\sqrt{2D}} \right)^{pd_w} (\ln N)^{p(1-d_w)} \sum_{n=0}^{\infty} \sum_{m=0}^{n} \tau_{nm} (\ln \ln N)^m (\ln N)^n,
\]

where the coefficients \( \tau_{nm} \) depend on \( p, j \) and the substrate. The question we want to answer is whether this is also true for disordered media.

The prior knowledge of the short-time asymptotic ex-
expression of the survival probability $\Gamma(z, t)$, which is defined as the probability that a random walker who starts at an origin site has not arrived at a spherical boundary of radius $z$ in the time interval $(0, t)$, was a key that allowed to find rigorously the full asymptotic approximation \( (t_{j,N}^m) \) for Euclidean and some deterministic fractal media \( [20] \) in Refs. \([13, 14, 16]\). (Here, and henceforth, we use the term “short time” or “short-time regime” to mean that $z/(z^2(t))^{1/2} \gg 1$, $(z^2(t))$ being the mean square distance $z$ travelled by a single random walker by time $t$.) However, the use of the asymptotic procedures of Refs. \([13, 14, 16]\) for estimating $(t_{j,N}^m)$ for disordered media is impeded by the fact that the value of the short-time survival probability is unknown for these substrates. In spite of this, in Ref. \([14]\) it was conjectured that Eq. (1) is valid for disordered media too \([21]\). Unfortunately, the scaling approach of Dr"ager and Klafter \([15]\), using a scaling approach, have shown recently that Eq. (1) is not valid for disordered media too \([21]\). For the deterministic fractals discussed in Ref. \([14]\), so that, for these cases, Eqs. (1) and (2) agree. Unfortunately, the scaling approach of Dr"ager and Klafter is not precise enough to lead to a fully correct asymptotic expression of the form of Eq. (1). This can be seen, for example, because the asymptotic series for $(t_{1,N}^j)$ given by these authors in Ref. \([13]\) does not agree with the rigorous asymptotic series reported in Refs. \([13, 14]\) for the one-dimensional lattice.

In this paper we deal with the order statistic of the survival probability $\Gamma(z, t)$, which is defined as the probability that a random walker who starts at the site $z$ by time $t$, follows Eq. (3) for the two-dimensional incipient percolation aggregate. We check this conjecture in the two-dimensional incipient percolation aggregate and, on the way, estimate for this case the unknown parameters appearing in the proposed relationship. In Sec. III we give the asymptotic expressions of the moments of $t_{N,j}$ for stochastic fractals assuming that the survival probability for the short-time regime is given by the expression proposed in Sec. I. These theoretical results are compared with simulation data for the two-dimensional percolation aggregate. The paper ends with some conclusions and remarks.

II. SHORT-TIME SURVIVAL PROBABILITY

From now on, we will denote by $z$ either the chemical distance $\ell$ or the Euclidean distance $r$. As discussed in Sec. I we have to know the survival probability $\Gamma(z, t)$, or equivalently the (boundary) mortality function $h(z, t) = 1 - \Gamma(z, t)$, for short times \( [i.e., \; z/(z^2(t))^{1/2} \gg 1] \) in order to calculate the escape-time moments $(m_{j,N}^m)$. This mortality function is the probability that a given random walker starting at the site $z = 0$ will have been trapped by an absorbing boundary of radius $z$ by time $t$. In this paper we will consider media for which this mortality function (averaged over all realizations of the lattice if the lattice is stochastic) grows for $\xi \equiv z/t^{1/d_w^c} \gg 1$ as a stretched exponential with power-law corrective terms:

$$h(z, t) = h(\xi) \sim A\xi^{-\mu v} \exp[-c\xi^v] \{1 + h_1\xi^{-v} + \ldots\}$$

(3)

where $A, \mu, v, h_1$ are characteristic parameters of the lattice. The anomalous diffusion coefficient $d_w^c$ is replaced by $d_w$ if the Euclidean distance is used ($z = r$) and by $d_w^c$ if $z = \ell$. There are good reasons to propose the functional form of Eq. (3). To start with, this relation holds for Euclidean lattices. Then, the mortality function was obtained in Ref. \([14, 22]\) for some class of deterministic fractals by using a renormalization procedure which involved only boundaries containing the nearest neighbors of the origin after successive decimations, and the result is in agreement with Eq. (3) too. In these media $z \equiv r$, $\mu = 1/2$ and $v = d_w/(d_w - 1)$. (Although the results of renormalization cannot be directly applied to any origin and any arbitrary boundary, there are again good reasons to suspect that the functional form is the same \([23]\).) Also, a closely related quantity to $h(z, t)$, the (site) mortality function $h_1(r)$ defined as the probability that a specific site $r$ has been visited by a single random walker by time $t$, follows Eq. (3) for the two-dimensional percolation aggregate \([23]\). Finally, one expects that the mortality function and the propagator $P(z, t)$ decay for $\xi \gg 1$ in the same way \([23, 24]\), and it is known that $P(z, t)$ decays as $\exp(-c\xi^v)$ for stochastic fractals, where $v = d_w^c/(d_w^c - 1)$ \([1,2,3]\). We will devote the remaining part of this section to verify that $h(\ell, t)$ and $h(r, t)$ behave as conjectured in Eq. (3) for the two-dimensional percolation aggregate.
A. Mortality function for the two-dimensional percolation aggregate: Chemical distance

We start by estimating the mortality function \( h(\ell, t) \) when the “circular” absorbing boundary is placed at the chemical distance \( \ell \) from the starting site. The numerical evaluation of this quantity is performed by the Chapman-Kolmogorov method (also called the exact enumeration method \([1, 2]\)). The circular boundary of absorbing traps is simulated by a set of special sites belonging to the cluster that absorbs all the probability density that enters them without giving back any probability to their neighbors. In the simulation we locate these boundaries at distances \( \ell = 40, 100, 160 \), and perform three experiments of absorption for \( t = 0, 1, \ldots, t_{\text{max}} \) with \( t_{\text{max}} = 1000 \) on every aggregate built, one for each boundary. The resulting mortality function is averaged over 2000 realizations of the percolation aggregate, which are generated by the Leath method \([3, 24]\).

In Fig. 1 we plot \( \ln[-\ln h(\ell, t)] \) versus \( \ln \hat{\xi} \) with \( \hat{\xi} = \ell/t^1/d_w^\ell \) and \( d_w^\ell = 2.4 \). [Hereafter we will put the symbol \( \sim \) over quantities corresponding to the chemical (Euclidean) distance.] The value of \( d_w^\ell \) was taken from one of our previous works \([12]\) and is in agreement with the values reported by other authors \([1, 2, 23, 29]\). If the conjecture in Eq. (3) is right we should observe the linear behavior \( \ln[-\ln h(\ell, t)] \sim \ln \hat{\xi} + \hat{v} \ln \xi \). Certainly the plots seem linear in Fig. 1 except for a portion in the range of large \( \xi \) where the curves deviate upwards. This is a finite size effect (already analyzed in the case of the two-dimensional Sierpinski gasket in Ref. \([23]\)) associated with the existence of a minimum arrival time corresponding to a random walker who travels “ballistically” along a chemical path from the origin to the absorbing boundary, which in turn implies a maximum available value of \( \hat{\xi} \) in the simulations: \( \hat{\xi}_{\text{max}} \approx \ell^{1-1/d_w^\ell} \). This value is around \( \hat{\xi}_{\text{max}} \approx 2.15 \) for \( \ell = 40 \) and \( \hat{\xi}_{\text{max}} \approx 2.96 \) for \( \ell = 160 \). This apparently means that the reliable interval for numerical fitting is larger for \( \ell = 160 \) but we must also take into account that the minimum value of \( \xi \) attained in the simulations is \( \xi = t_{\text{max}}^{1/d_w^\ell} \) which is proportional to \( \ell \) (in our simulations \( \xi_{\text{max}} = 1000 \)). For this reason, in order to carry out the numerical fit, we have concatenated the simulation data for which the plots are almost linear. The result \( h(\ell, t) = A \xi^{-\mu \hat{c}} \exp(-\hat{c} \xi^{\hat{c}}) \) with \( A = 1, \mu = -1.8, \hat{c} = 1.65 \) and \( \hat{v} = d_w^\ell/(d_w^{\ell} - 1) = 1.714 \) is also plotted in Fig. 1. The agreement is excellent.

B. Mortality function for the two-dimensional percolation aggregate: Euclidean distance

We also simulated the mortality function \( h(r, t) \) in the percolation aggregate when the circular absorbing boundary has a Euclidean radius \( r \). The analysis of these results parallels those of the previous subsection. In Fig. 2 we have plotted the simulation results for the double logarithm of the mortality function \( \ln[-\ln h(r, t)] \) versus \( \ln \hat{\xi} \) for \( r = 35 \) (triangles), \( r = 55 \) (circles), \( r = 75 \) (squares). The line represents the function \( h(r, t) = A \xi^{-\mu \hat{c}} \exp(-\hat{c} \xi^{\hat{c}}) \) with \( A = 1, \mu = -1.8, \hat{c} = 0.9, \hat{v} = d_w^\ell/(d_w^{\ell} - 1) = 1.714 \) and \( \hat{\xi} = 9 \) is also plotted in Fig. 2. The agreement is excellent.
where trapping boundary, $h$ is clearly wider for the Euclidean case than for the chemical case, although to a quite noticeably lesser extent than for the site mortality function [12] and the propagator [2, 25].

The distribution over the different percolation aggregate realizations is simply the coefficient of $z^j$−1 in the Taylor series expansion up to the term of order $\{1 + \ln \lambda\}$. Proceeding as in Refs. [13, 14, 16], one gets:

$$U_{N,m}(\zeta) = \frac{m}{1 - \zeta} \left[ 1 + \frac{(\mu \ln \lambda - \gamma)}{\ln \lambda} + \frac{\alpha}{2 \ln^2 \lambda} \left\{ (1 + \alpha) \left( \frac{\pi^2}{6} + \gamma^2 \right) + 2 \mu \gamma - 2 \bar{h}_1 t_0^\beta \right. ight.$$

$$- \left. 2 \mu (1 + \alpha) \gamma \ln \lambda + (1 + \alpha) \mu^2 \ln^2 \lambda \right] + O \left( \frac{\ln^3 \ln \lambda}{\ln^3 \lambda} \right)$$

where $\alpha \equiv m/\beta$ and $\gamma \approx 0.577215$ is the Euler constant.

Once the generating function is known, the escape times and their moments are calculated straightforwardly because $(t_{j,N}^m)$ is simply the coefficient of $z^j$−1 in the Taylor series expansion of $U_{N,m}(\zeta)$. Therefore, the $m$th moment of the first passage time of the first out of $N \gg 1$ diffusing particles is equal to $U_{N,m}(0)$, i.e.,
with $\lambda_0 = at_0^{\mu/\beta} = A \lambda^\mu$ and $\alpha = m/\beta = mv/d_w^2$, as before. Notice that the main term for this expression for $m = 1$ agrees with the result given in Eq. 18 if we use $v = d_w^2/(d_w^2 - 1)$.

The calculation of $\langle t_{j,N}^m \rangle$ for $j > 1$ is more involved. We begin with the identity $\ln^n(1-z) = n! \sum_{i=0}^{\infty} (-1)^i S_i(n) z^i/i!$, where $S_i(n)$ are the Stirling numbers of the first kind [27]. Using this relation, the expansions of $1/\ln^2 \lambda$ and similar terms in Eq. (6) in powers of $1/\ln \lambda_0 N$ can be found [14]. After some algebra we have

$$\langle t_{j,N}^m \rangle \sim \frac{t_0^m}{\ln^2 \lambda_0 N} \left\{ 1 + \frac{\alpha (\mu \ln \lambda_0 N - \gamma)}{\ln \lambda_0 N} + \frac{\alpha^2}{2 \ln^2 \lambda_0 N} \left[ (1 + \alpha) \left( \frac{\pi^2}{6} + \gamma^2 \right) + 2\mu \gamma - 2h \lambda_0^\beta \right] \ln \lambda_0 N + O \left( \frac{\ln^3 \lambda_0 N}{\ln \lambda_0 N} \right) \right\}$$

(8)

Let us now compare the theoretical predictions in Eqs. (8), (9) and (13) with simulation results in the two-dimensional percolation aggregate when an absorbing "circular" boundary is placed either at a given chemical distance $z = \ell$ or at a given Euclidean distance $z = r$.

A. Absorbing boundary at a given chemical distance

The first passage time of the first few random walkers out of a set of $N = 2^i$, $i = 2, 3, \ldots, 16$ to a "circular" boundary of chemical radius $\ell = 50$ was simulated on 2000 aggregates. In Fig. 4 we plot the scaled simulation results of $[(t_{1,N})/\ell d_w^\mu]^{-\delta}$, with $\delta = 1/(d_w^\mu - 1)$, versus $\ln N$ for $\ell = 50$ and compare them with the theoretical predictions of Eq. (13), namely,

$$\left[ \frac{(t_{1,N})}{\ell d_w^\mu} \right]^{-\delta} \approx \left[ \frac{\ln N}{\ell} \right] \left\{ 1 - \frac{\delta \ell d_w^\mu}{\beta} \frac{\mu \ln \lambda_0 N - \gamma - \ln \lambda_0^\mu}{\ln N} \right\}$$

(14)

where $\delta = 1/(d_w^\mu - 1)$. Notice that $[(t_{1,N})/\ell d_w^\mu]^{-\delta}$ depends linearly on $\ln N$ if $\beta = d_w^\mu/(d_w^\mu - 1)$. Figure 4 shows that this is indeed the case.

From Eq. (13) one gets

$$R \equiv \frac{\langle t_{j,N} \rangle}{\langle t_{j,N} - t_{1,j} \rangle} \sim \left( \sum_{n=1}^{j-1} \frac{1}{n} \right)^{-1} \frac{\hat{\beta}}{d_w^\mu} \ln N.$$  

(15)

so that $R$ should be linear in $\ln N$ and independent of the radius of the chemical boundary $\ell$. This is confirmed in Fig. 5 where this quotient is plotted versus $\ln N$ for $j = 2, 3$ and $\ell = 50, 80$. A good superposition of the simulation data for $\ell = 50$ and $\ell = 80$ is observed. The simulation results are in good agreement with the prediction of Eq. (15) with $\hat{\beta} = d_w^\mu/(d_w^\mu - 1)$ and $d_w^\mu = 2.4$.

Finally, in Fig. 6 we show the quotient between the average lifetime of the $j$th particle and its variance, $\langle t_{j,N} \rangle/\sigma_{j,N}$, versus $\ln N$ for the first, second and third random walkers to arrive at the chemical boundary with $\ell = 50$. This is a remarkable figure that shows the crucial importance of the corrective terms on the order statistics formulae. Except for the first walker ($j = 1$) we find striking discrepancies between the zeroth-order
(main term) of the asymptotic expression in Eq. (13) and the simulation results. We attribute these discrepancies to the $O(\ln^3 \ln N/\ln N)$ corrective terms not considered in the zeroth-order asymptotic expression. Let us explain why. In Fig. 5 we plot the function $\ln^3 \ln N/\ln^m N$ for several values of $n$ and $m$. One sees that $\ln^3 \ln N/\ln N$ cannot be neglected versus 1 even for values of $N$ larger than the Avogadro’s number. Therefore, it is not strange to find a poor agreement between the zeroth-order asymptotic expression and the simulation results. (In fact, what is truly unexpected is the good agreement of the zeroth-order theoretical prediction with simulation results for $j = 1$.) Notice that if we had calculated another corrective term in the asymptotic expansion (7), then the term neglected in Eq. (13) would be of order $\ln^3 \ln N/\ln^2 N$, which, as shown in Fig. 6, would likely be much smaller than the $\ln^3 \ln N/\ln N$ corrections. Unfortunately, corrective terms of order $\ln^3 \ln N/\ln N$ involve parameters such as $h_1$ [see Eq. (3)] which are very difficult to estimate relying only on simulation results. One also observes in Fig. 7 that the relative errors of the zeroth- and first-order approximations to $\langle t_{j,N} \rangle$ committed by ignoring terms of the form $\ln \ln N/\ln N$ and $\ln^2 \ln N/\ln^2 N$, respectively, will likely be small. This explains the good agreement between theory and simulation for lifetimes shown in Figs. 4 and 5.
B. Absorbing boundary at a given Euclidean distance

We have also simulated the order-statistics of the arrival of $N$ random walkers at a circular boundary of Euclidean radius $r$. The analysis of these results parallels those of the previous subsection. The numerical results for $\langle t_{1,N}/r^{d_w}\rangle^{-\delta}$ and $\langle t_{j,N}/t_{1,N} - t_{1,N}\rangle$ versus $\ln N$ are plotted in Figs. 3 and 4, respectively. The radii of the circular boundary are $r = 50$ and $r = 75$, and we used 2000 aggregates to perform the substrate average. The theoretical predictions of Eqs. (14) and (15) [replacing $\ell$ by $r$, $d_w$ by $d_w$, $\delta$ by $1/(d_w - 1)$ and $\delta$ by $\ln N$] lead to an agreement with numerical results that is not as good as that obtained when the chemical distance was used (see Figs. 3 and 4), although the linear behavior of $T$ and $R$ versus $\ln N$ and the superposition of the simulation data for different values of $r$ is still there. This slightly worse prediction is in accordance with the fact, discussed in Sec. II B, that the collapse of the numerical mortality function for different distances $r$ to a single curve is not so nearly perfect as when chemical distances are used.

IV. CONCLUSIONS AND REMARKS

In this paper we studied the order statistics of the survival times (lifetimes or exit times) of $N \gg 1$ independent random walkers that are put initially on a stochastic lattice at the center of a “spherical” absorbing boundary of radius $z$. We found the moments $\langle t_{j,N}^m \rangle$ of the lifetime of the $j$th random walker of the set of $N$ in terms of an asymptotic series that decays mildly in powers of $1/\ln N$. These theoretical results were compared with numerical simulations for the two-dimensional incipient percolation aggregate with the absorbing boundary placed either at a given chemical distance $z = \ell$ or at a given Euclidean distance $z = r$. The agreement found was reasonable and in accordance with the large size of the asymptotic corrective terms.

The theoretical approach relies on the knowledge of the mortality function $h(z,t)$ for short times. We assumed that the functional form of the short-time mortality function for a stochastic lattice averaged over all realizations coincides with that of the mortality function for Euclidean lattices and for the class of deterministic fractal lattices whose sites are isolated by their nearest neighbors. This is the main assumption of this paper and we showed that it is compatible with our simulation results for the two-dimensional percolation aggregate with Euclidean and chemical absorbing circular boundaries. This allows us to describe the order statistics of the diffusion process on many kinds of substrates (namely, Euclidean media, and deterministic and stochastic fractals) with the same approach and formulae.

A useful feature of the order statistics description is that it allows one to infer properties of the diffusive system (diffusion constant, number of diffusing particles, concentration of traps, effective dimension of the diffusive substrate, . . . ) from the analysis of the behavior of those particles that are trapped first. This could be an advantage when it is impractical or impossible to wait until the entire reaction is over.

Finally, it should be noted that our simulation results for the stochastic fractal we have studied (the incipient percolation aggregate) are insufficient to perform a completely reliable numerical fit to the general form of the mortality function $h(z,t)$ with its dominant exponential and subdominant power-law terms simultaneously. Indeed, even to find the true exponent $v$ of the dominant term for the propagator is not at all easy as past controversies about that true value reveal.
the value of a hypothetical power-law subdominant term (equivalent to the term $\xi^{-\mu}v$ of Eq. (12)) is still a matter of discussion. Hence, one must expect that to find a completely reliable description of the short-time mortality function will be similarly difficult, and this task will therefore require more detailed simulations on larger lattices, with longer simulation times and, of course, averaging over many more realizations of the lattice than those we have used here. This is simply too much for a common workstation, but we think the effort on more powerful systems would be worthwhile: first, because of the interest in the percolation aggregate as a model of disordered media, and, second, because simulation of the mortality function (with circular boundary) is easier than that of the propagator and, since the asymptotic coefficients and exponents of these two functions are related, estimation of these parameters for the mortality function would help to solve the long-standing controversy on the exact asymptotic form of the propagator in fractal media.

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