ARTICLES

Long-time properties of random walks with a single trap

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For both Brownian and enhanced random walks, we can distinguish between systems where the random walker returns to the origin with certainty, and where the random walker may escape from the trapping origin with a probability strictly larger than zero. Examples for the first case are Brownian walks in one and two dimensions, where it is known that the asymptotic probability $f(t)$ to be trapped at time $t\rightarrow\infty$ behaves differently from the asymptotic probability $p(t)$ to pass through the origin in the same system without trap. We also find this result for Lévy flights in one dimension with the exponent of the characteristic function $\geqslant 1$. On the other hand, we compute $p(t)$ and $f(t)$ for various systems with a nonzero escape probability. In particular, we consider an anisotropic walk, which behaves Brownian in one direction and executes Lévy flights along the second direction. For these cases we are able to prove that $f(t)$ and $p(t)$ follow the same inverse power law asymptotically, the ratio given by the squared escape probability. [S1063-651X(98)01510-4]

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I. INTRODUCTION

Many problems in chemical physics, such as recombination kinetics, exciton transfer, or the calculation of rate constants for chemical reactions, are related to the concept that a diffusing particle is trapped. The survival probability of such a particle (random walker) and the time of its first arrival at a trap have been the subject of theoretical investigations carried out for various systems, almost throughout the whole century [1–6]. Part of it deals with models where the trapping sites are uniformly distributed over the system [7–11]. An equivalent formulation is that there is one fixed trap and the initial position of the random walker is uniformly distributed. Due to the average over the initial coordinates, this situation is less complicated than the problem of a single trap and a random walker starting at a particular position. Also, the calculation of the mean first passage time (and higher moments of the first passage time) has a long tradition [12] (see Ref. [13] for early applications). But in many cases these moments do not exist [14].

A multitude of traps is certainly present in bulk experiments. During the last years, however, an increasing number of experiments has been done with individual representatives of the species [15–17]. In this context, more knowledge about the influence of a single trap might be useful, which is the subject of this work. In particular, we shall focus on systems where the random walker has a chance to avoid the trap.

The paper is organized as follows: In Sec. II we recapitulate some concepts of the theory of random walks, and apply them to three different systems, covering both Brownian and enhanced diffusion: First, we model the Brownian case by steps to the nearest neighbors only (one, two, and three dimensions). Second, we deal with Lévy flights in one dimension. Third, we present a combination of the Brownian and Lévy dynamics (two dimensions). For each of the examples, we plot the probability of the first return to the origin as a function of time, computed by an exact enumeration technique. In Sec. III we prove the basic finding of this work, indicated by the results of Sec. II, i.e., the probability for first return follows the same inverse power law as the probability for free passage through the origin (random walk without trapping), if the asymptotic survival probability remains above zero. In Sec. IV we conclude and summarize.

II. FIRST RETURN TO THE ORIGIN

We consider an infinite system with d dimensions. The trap shall be placed at the origin of the system. To keep the formulation of the problem simple, we assume that the particle starts at the origin at time $t=0$, and the trap is activated immediately thereafter. If there were no trap, the probability for the particle to be at the origin at time t shall be denoted by $p(t)$, whereas $f(t)$ means the probability for returning to the origin for the first time. A subscript shall indicate the dimension of the system where necessary. In lattice description, the generating functions defined by

$$P(z) := \sum_{t=0}^{\infty} p(t) z^t, \tag{1}$$

$$F(z) := \sum_{t=0}^{\infty} f(t) z^t \tag{2}$$

provide a convenient way to establish a relation between f and p. The summation is over integer values of t, which means that the random walker moves at equal time steps. With $p(0)=1$ the relation between F and P is [6]

$$F(z) = 1 - 1/P(z), \tag{3}$$

whereas the recursion formula for the coefficients of F reads

$$f(t)=p(t)-\sum_{k=1}^{t-1} p(t-k)f(k), \quad t=1,2,\ldots . \tag{4}$$

$F(1)$ gives the probability of being trapped. If $F(1)$ is strictly smaller than 1, the random walker has a finite (i.e., nonzero) probability to escape. Its survival probability is defined by

$$\Phi(t)=1-\sum_{k=1}^{t} f(k). \tag{5}$$

The generating function for free propagation is related to the characteristic function $\lambda(\mathbf{k})$ by [6]

$$P(z)=\frac{1}{(2\pi)d}\int_{-\pi}^{\pi}\frac{d^d\mathbf{k}}{1-z\lambda(\mathbf{k})}, \tag{6}$$

$\mathbf{k}$ being the Fourier transform variable of the space variable $\mathbf{x}$.

A. Brownian diffusion

For Brownian motion the probability $p(t)$ is proportional to $t^{-d/2}$. Using the formalism described above, we may model the Brownian case by a symmetric random walk with hops to the nearest neighbors only. On a simple cubic lattice such a walk has the characteristic function

$$\lambda(k)=\frac{1}{d}\sum_{j=1}^{d}\cos k_j . \tag{7}$$

In one dimension $f(t)$ can be given explicitly. From Eqs. (3), (6), and (7) one finds

$$F_1(z)=1-\sqrt{1-z^2} \tag{8}$$

and

$$f_1(t)=\frac{1\times 3\times 5\cdots|t-3|}{2\times 4\times 6\cdots(t-2)\times t} \tag{9}$$

for even t, and 0 otherwise. Asymptotically,

$$f_1(t\to\infty)\sim t^{-3/2} \tag{10}$$

for t even. In higher dimensions it is not possible to evaluate $P(z)$ by elementary integrals. That is, for $d=2$, Eqs. (6) and (7) give $P_2(z)=(2/\pi)K(z)$, the complete elliptical integral of the first kind. The usual procedure is an expansion of the denominator in Eq. (6)

$$P(z)=\frac{1}{(2\pi)^d}\int_0^\infty dt\, e^{-t}\prod_{j=1}^{d}\left(\int_{-\pi}^{\pi} e^{(zt/d)\cos k_j}dk_j\right)$$
$$=\int_0^\infty dt\, e^{-t}\left[I_0\left(\frac{zt}{d}\right)\right]^d=\int_0^\infty dt\, e^{-t}\left[\sum_{k=0}^{\infty}\frac{\left(\frac{zt}{2d}\right)^{2k}}{k!^2}\right]^d, \tag{11}$$

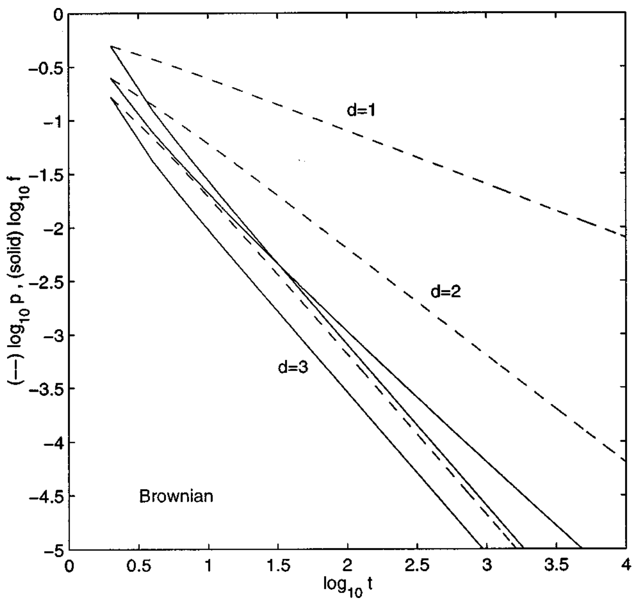

FIG. 1. Probabilities for free passage through the origin, $p(t)$ (upper curves), and first return, $f(t)$ (lower curves), as functions of time (arbitrary units) for simple random walks in one, two, and three dimensions [Eqs. (4) and (12)–(14)].

with $I_0(z)$ the modified Bessel function of the first kind of order 0. Rearranging the sums and integrating over t we obtain the even coefficients of the series in z,

$$p_1(2t)=\frac{(2t)!}{4^t t!^2}, \tag{12}$$

$$p_2(2t)=\sum_{k=0}^{t}\frac{(2t)!}{16^t k!^2(t-k)!^2}=\frac{(2t)!^2}{4^{2t}t!^4}=[p_1(2t)]^2, \tag{13}$$

$$p_3(2t)=\frac{(2t)!}{36^t}\sum_{k=0}^{t}\frac{(2k)!}{(t-k)!^2k!^4} \tag{14}$$

for one, two, and three dimensions [18]. All odd $p(2t-1)$ are zero. Obviously, $p_1(t)$ are just the coefficients of an expansion of $1/\sqrt{1-z^2}$ [19]. Using Eq. (4), $f(t)$ can be evaluated easily on the computer for the first some 1000 time steps, which is enough to show the asymptotic behavior (Fig. 1). The plots for $d=1$ and $d=2$ present the well-known analytical results of Eq. (10) [20] and

$$f_2(t\to\infty)\sim\frac{1}{t(\ln t)^2}. \tag{15}$$

The latter follows from [21]

$$\sum_{k=t}^{\infty} f_2(k)\sim 1/\ln t, \tag{16}$$

the result of a Tauberian theorem [22,6]. For $d=3$ the slope of the double logarithmic plot $(-\frac{3}{2})$ indicates the same asymptotic power law as for $d=1$ or for free propagation in three dimensions, i.e.,

$$f_3(t\to\infty)\sim t^{-3/2}. \tag{17}$$

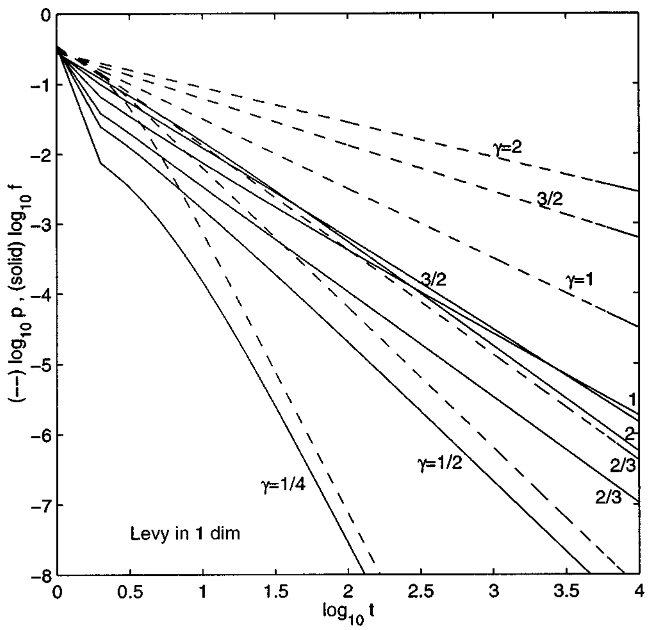

FIG. 2. Probabilities for free passage through the origin, $p(t)$ (dashed curves), and first return, $f(t)$ (solid curves), as functions of time (arbitrary units) for Lévy flights in one dimension with $\gamma = \frac{1}{4}$, $\frac{1}{2}$, $\frac{2}{3}$, 1, $\frac{3}{2}$, and 2 [Eqs. (4) and (A1)–(A5), $a=1$].

We shall prove below that in three dimensions $f(t)$ and $p(t)$ have to follow the same power law, as there is a finite probability for escape.

B. Lévy flights

In the following we are interested in the following question: How will the asymptotic power law for $f(t)$ be changed, if the steps of the random walk have a wide and slowly decaying size distribution, i.e., in the case of a Lévy flight? In continuous space, such Lévy flights are described by the characteristic function [6,5]

$$\lambda(\mathbf{k};t) = e^{-at|k|^{\gamma}}, \tag{18}$$

with $0<\gamma<2$ and a a positive constant. In $\mathbf{x}$ space this corresponds to the following asymptotic behavior of the step size distribution

$$p_{\text{step}}(|x|\to\infty) \simeq \frac{A}{|\mathbf{x}|^{\gamma+1}}, \tag{19}$$

where A is another constant. For one dimension there is some related work by Zumofen and Klafter [23]. They assume an absorbing boundary at the origin, which would be identical to a system with a trap in the case of a simple random walk with steps to the nearest neighbors only. For Lévy flights, however, it makes a difference whether the particle is allowed to jump over the origin or not. Interestingly, the survival probability $\Phi(t)$ does not depend on γ in the latter case. This was shown for $1\leqslant\gamma\leqslant 2$, but is believed to be true for smaller γ as well [23]. For the case of a trap we clearly find a dependence on γ (Fig. 2). The inverse Fourier transform of $\lambda(k;t)=\exp(-at|k|^{\gamma})$ at $x=0$ gives

$$p_1(t) = \frac{\Gamma\left(1+\dfrac{1}{\gamma}\right)}{\pi(at)^{1/\gamma}} \sim t^{-1/\gamma}. \tag{20}$$

In a lattice description, the boundaries of the integral transform read $\pm\pi$ instead of $\pm\infty$ [see Eq. (6)], which means that the prefactor in Eq. (20) will contain an incomplete gamma function converging to the complete one for large at. For some cases shown in Fig. 2, these integrals are given in the Appendix. The plots for $f(t)$ are again obtained from Eq. (4). Contrary to the absorption case, the dependence on γ is expected for a trap, as it is known that there is no escape for the random walker for $\gamma\geqslant 1$ and a finite escape probability for $\gamma<1$ [6], i.e., the survival probability $\Phi(\infty)=1-F(1)$ depends on γ [24]. We again find that $p(t)$ and $f(t)$ have the same asymptotic behavior $\sim t^{-1/\gamma}$, when there is a finite escape probability, i.e., $\gamma<1$. The case $\gamma=1$ with $p\sim 1/t$ is analogous to the simple random walk in two dimensions in Fig. 1. In both cases the survival probability diminishes logarithmically [Eq. (16)]. For $\gamma>1$, Fig. 2 again shows a power law for $f(t\to\infty)$, with the exponent increasing in magnitude for increasing γ.

C. Anisotropic walks

The last example we want to deal with in particular is a random walk in two dimensions, which behaves as Brownian in one direction and executes Lévy flights along the second direction. We use the characteristic function

$$\lambda(k_1,k_2) = \tfrac{1}{2}(\cos k_1 + e^{-a|k_2|^{\gamma}}), \tag{21}$$

and obtain

$$P(z) = \frac{1}{\pi}\sum_{k=0}^{\infty} \frac{z^{2k}}{4^{2k}k!^2} \int_0^{\infty} dt\, e^{-t} t^{2k} \int_0^{\pi} dk_2\, e^{(z/2)t\,\exp(-ak_2^{\gamma})}. \tag{22}$$

By expansion, the last integral yields

$$\int_0^{\pi} dk\, e^{(z/2)t\,\exp(-ak^{\gamma})} = \sum_{l=0}^{\infty} \left(\frac{z}{2}\,t\right)^{l} \frac{\Gamma\left(1+\dfrac{1}{\gamma}\right)}{(al)^{1/\gamma} l!}\, \hat{\gamma}\left(al\pi^{\gamma}, \frac{1}{\gamma}\right), \tag{23}$$

where

$$\hat{\gamma}(x,\alpha) := \frac{1}{\Gamma(\alpha)} \int_0^{x} e^{-t} t^{\alpha-1} dt \tag{24}$$

is the normalized incomplete gamma function, which tends to unity for large x [25]. Next we carry out the first integral to arrive at

$$P(z) = \frac{1}{\pi}\sum_{k=0}^{\infty}\sum_{l=0}^{\infty} \frac{(2k+l)!\,z^{2k+l}}{2^{4k+l} l!\,k!^2} \frac{\Gamma\left(1+\dfrac{1}{\gamma}\right)}{(al)^{1/\gamma}}\, \hat{\gamma}\left(al\pi^{\gamma}, \frac{1}{\gamma}\right). \tag{25}$$

Rearranging the sums, we obtain the coefficients of the series in z,

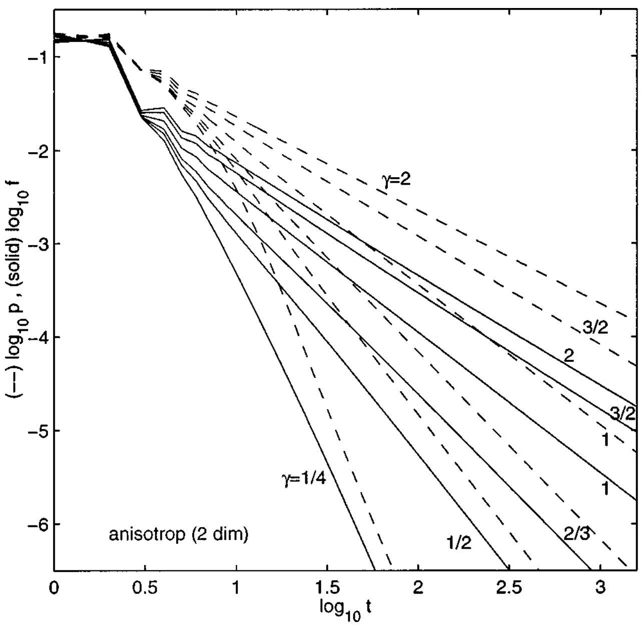

FIG. 3. Probabilities for free passage through the origin, $p(t)$ (dashed curves), and first return, $f(t)$ (solid curves), as functions of time (arbitrary units) for anisotropic walks in two dimensions with $\gamma=\frac{1}{4}$, $\frac{1}{2}$, $\frac{2}{3}$, 1, $\frac{3}{2}$, and 2 [Eqs. (4), (26), and (27); $a=1$].

$$p(2t)=\frac{(2t)!}{16^t}\left\{\frac{1}{t!^2}+\frac{\Gamma\left(1+\frac{1}{\gamma}\right)}{\pi a^{1/\gamma}}\times\sum_{k=1}^{t}\frac{4^k\hat{\gamma}\left(2ak\pi^\gamma,\frac{1}{\gamma}\right)}{(t-k)!^2(2k)!(2k)^{1/\gamma}}\right\}\tag{26}$$

for the even ones, and

$$p(2t-1)=\frac{\Gamma\left(1+\frac{1}{\gamma}\right)}{\pi\alpha^{1/\gamma}}\frac{(2t-1)!}{4^{2t-1}}\times\sum_{k=1}^{2t-1}\frac{2^{2k-1}\hat{\gamma}\left[a(2k-1)\pi^\gamma,\frac{1}{\gamma}\right]}{(t-k)!^2(2k-1)!(2k-1)^{1/\gamma}}\tag{27}$$

for the odd, with $t=1,2,\ldots$ as above. Equation (4) is employed to obtain $f(t)$. Both $p(t)$ and $f(t)$ are shown in Fig. 3 for a set of different values of γ. For γ well below 2, the asymptotics of both probabilities follow the same inverse power law with an exponent $\sim\frac{1}{2}+(1/\gamma)$. For γ close to 2, a more detailed investigation, not displayed in Fig. 2, shows the following behavior within the computed time range: The magnitude of the slope is still a little bit smaller than $\frac{1}{2}+(1/\gamma)$ for $p(t)$, but somewhat larger for $f(t)$. For $\gamma=2$ we expect the same logarithmic behavior as for the one-dimensional Lévy flights with $\gamma=1$ or the two-dimensional Brownian walker.

III. ASYMPTOTICS FOR FINITE ESCAPE

As we saw in Sec. II, the asymptotic probability $p(t\to\infty)$ for return to the origin in a trapless system in general follows an inverse power law. The exponent depends on the dimension of the system and in case of enhanced diffusion also on the exponent γ. There are two possibilities: The random walker will return to the origin either with certainty, or there is a nonzero probability to escape from the trap placed at the origin. Increasing the dimension or decreasing γ may change the system from zero escape to a finite escape probability. For the latter we shall now also prove that $f(t\to\infty)$ follows an inverse power law, the exponents being equal for p and f.

Let us assume that

$$p(t)\simeq ct^{-\alpha}\tag{28}$$

asymptotically, with c a constant. For a finite escape probability, α has to be larger than 1. We regard the case $1<\alpha<2$ first. According to an Abelian theorem [26,6], the singular behavior of the generating function is described by

$$P(z)\simeq P(1)+c\Gamma(1-\alpha)(1-z)^{\alpha-1}\tag{29}$$

in the neighborhood of $z=1$. We define

$$g(z):=\sum_{k=0}^{\infty}g_kz^k:=\frac{F(1)-F(z)}{1-z}.\tag{30}$$

Expanding $1/P(z)$ in Eq. (3) and using Eq. (29), we obtain

$$g(z)=-\frac{c\Gamma(1-\alpha)}{P(1)^2}(1-z)^{\alpha-2}.\tag{31}$$

The singular behavior of this function can be related to its coefficients g_k by means of a Tauberian theorem [22,6]

$$\sum_{k=0}^{t}g_k\simeq\frac{-c\Gamma(1-\alpha)}{P(1)^2\Gamma(3-\alpha)}t^{2-\alpha}\tag{32}$$

and

$$g_t\simeq\frac{ct^{1-\alpha}}{P(1)^2(\alpha-1)},\tag{33}$$

if the g_k are strictly positive and a monotonic function of k, at least from some value of t onwards. This is the case as the coefficients g_k are the partial sums of $f(t)$:

$$g(z)=\sum_{t=1}^{\infty}f(t)(1-z^t)/(1-z)=\sum_{t=0}^{\infty}\sum_{k=0}^{t}f(t+1)z^k=\sum_{k=0}^{\infty}z^k\sum_{t=k+1}^{\infty}f(t),\tag{34}$$

$$g_k=\sum_{t=k+1}^{\infty}f(t).\tag{35}$$

Hence, asymptotically,

$$f(t) \simeq g_t - g_{t+1} \simeq \frac{ct^{-\alpha}}{P(1)^2} = [1-F(1)]^2 ct^{-\alpha} \tag{36}$$

follows the same power law as $p(t)$ and the ratio between both probabilities is given by the squared escape probability. We continue to show that this relation also holds for $\alpha \geqslant 2$.

For $\alpha = 2$ the singular behavior of $P(z)$ can be described by

$$P(z) \simeq P(1) + c(1-z)\ln(1-z) \tag{37}$$

as the coefficients of an expansion in z of this expression are $\simeq c/t^2$ for large t. Using the definition of Eqs. (30) and (3), we obtain

$$g(z) = \frac{c}{P(1)^2} \ln \frac{1}{1-z}, \tag{38}$$

and the Tauberian theorem [22,6] yields

$$\sum_{k=0}^{t} g_k \simeq \frac{c}{P(1)^2} \ln t \tag{39}$$

and

$$g_t \simeq \frac{c}{P(1)^2 t}, \tag{40}$$

respectively. The last equation leads to our result of Eq. (36) like Eq. (33).

For (integer or noninteger) $\alpha > 2$ the proof can be carried out in a way similar to the preceding two paragraphs. We have to define a function $g^{(n)}$ appropriately, so that

$$g^{(n)}(z) = (-1)^n \frac{c\Gamma(1-\alpha)}{P(1)^2} (1-z)^{\alpha-1-n} \tag{41}$$

for $0 < \alpha - n < 1$, and

$$g^{(n)}(z) = -\frac{c}{n!P(1)^2} \ln(1-z) \tag{42}$$

for $\alpha = n+1$, with n a natural number. The lowest order $g^{(1)}(z)$ is identical with $g(z)$ defined above. We shall demonstrate the procedure for $n=2$: Including the powers up to the singularity, $P(z)$ reads

$$P(z) \simeq P(1) - c_1(1-z) + c\Gamma(1-\alpha)(1-z)^{\alpha-1} \tag{43}$$

for noninteger α, and

$$P(z) \simeq P(1) - c_1(1-z) - \frac{c(1-z)^2}{2} \ln(1-z) \tag{44}$$

for $\alpha = 3$, with c_1 a constant equal to

$$c_1 = P(1)^2 \sum_{t=1}^{\infty} t f(t). \tag{45}$$

From the definition

$$g^{(2)}(z) := \frac{c_1/P(1)^2 - g(z)}{1-z}, \tag{46}$$

we obtain

$$g^{(2)}(z) = \sum_{t=1}^{\infty} f(t)\left(t - \sum_{k=0}^{t-1} z^k\right) \Big/ (1-z)$$

$$= \sum_{k=0}^{\infty} z^k \sum_{l=k+1}^{\infty} (l-k) f(l+1), \tag{47}$$

$$g_k^{(2)} = \sum_{t=k+2}^{\infty} (t-k-1) f(t). \tag{48}$$

As $g_k^{(2)}$ is again a strictly positive sum and monotonic in k, the Tauberian theorem [22,6] yields

$$g_t^{(2)} = \frac{c\Gamma(1-\alpha)}{\Gamma(3-\alpha)P(1)^2} t^{2-\alpha} \tag{49}$$

for noninteger α, and

$$g_t^{(2)} = \frac{c}{2P(1)^2 t}, \tag{50}$$

for $\alpha = 3$, respectively. As $g_{t-1}^{(2)} - g_t^{(2)} = g_t$, we obtain the result of Eq. (36) by taking twice the derivative of Eqs. (49) and (50), respectively.

IV. CONCLUDING REMARKS

The most significant finding is the fact that $f(t)$ is proportional to $p(t)$ for large times, if $F(1) < 1$. This is important as the probability $p(t)$ for return to the origin without the need to account for the trap is always easier to be determined than the first return $f(t)$. Moreover, Eq. (36) also gives the ratio between both probabilities. The vertical distance between the straight asymptotes in the double logarithmic plots equals $2 \ln \Phi(\infty)$ in all cases where the escape probability $\Phi(\infty) \neq 0$. This can be checked by calculating the asymptotic survival probability either numerically or, in some cases, using an analytical expression for $\Phi(\infty)$. That is, for the simple cubic lattice $\Phi(\infty) = 0.659...$ [27], and for the anisotropic walk, with $\gamma = 1$ and $a = 1$,

$$P(1) = 2 \ln(3e^{\pi} + \sqrt{3 + 9e^{2\pi} - 12e^{\pi}} - 2)/(\pi\sqrt{3}) \approx 1.80, \tag{51}$$

which gives $\Phi(\infty) \approx 0.55$. With regard to experiments, it would be interesting to look for systems where the trap can be switched off or on. Then both probabilities could be measured directly, yielding a method to determine the asymptotic survival probability regardless of any short-time constants.

We want to point out that the same asymptotic power law may arise from very different systems. That is, we have $f(t) \sim t^{-3/2}$ for three-dimensional Brownian motion, but also in one dimension for Lévy flights with $\gamma = \frac{2}{3}$ or in our two-dimensional model for $\gamma = 1$. In addition, there is another possibility which differs from the systems just mentioned by a vanishing probability for escape: the simple random walk in one dimension. As for the dimensions with regard to Brownian walks, there is also a choice between two different values of γ for one-dimensional Lévy flights, which yield the same asymptotic exponent for f (in the range between 1 and

1.5), but one of which is associated with zero escape, whereas the other permits an escape. The limiting case is $\gamma=1$, near which the slope of f is shallowest.

With regard to our anisotropic model we conclude from the numerical evidence that the properties of such a random walk/flight are simply an addition of both contributions, without any new features. In particular, the exponent for p is the sum of 1/2 (Brownian part) and $1/\gamma$ (Lévy direction). For γ close to 2 this exponent does not show up exactly in our plots, which is attributed to the fact that the asymptotes are reached only very slowly in this region.

If there were a distribution of traps, the survival probability always goes to zero. In general the decay is governed by a (stretched) exponential [8,9]. But if the distribution is very dilute, the assumption of a single trap might also be an appropriate description for an intermediate regime also in this case.

In summary, we have shown how in the asymptotic time regime the probability of first return to the origin is related to the probability of return without trapping in the case that the survival probability remains above zero, which covers a large variety of different systems.

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APPENDIX

The following list contains $p(t)=(1/2\pi)\int_{-\pi}^{\pi}\lambda(k;t)dk$ for some integer and rational values of γ:

$$\gamma=2: \quad \frac{\mathrm{erf}(\pi\sqrt{at})}{2\sqrt{\pi at}}, \tag{A1}$$

$$\gamma=1: \quad \frac{1-e^{-\pi at}}{\pi at}, \tag{A2}$$

$\gamma=\frac{2}{3}$:

$$\frac{3}{4\sqrt{\pi}(at)^{3/2}}\left[\mathrm{erf}(\pi^{1/3}\sqrt{at})-2\left(\frac{at}{\pi}\right)^{1/2}\pi^{1/3}e^{-\pi^{2/3}at}\right], \tag{A3}$$

$$\gamma=\frac{1}{2}: \quad \frac{2}{\pi a^2t^2}\left[1-e^{-\sqrt{\pi}at}(1+\sqrt{\pi}at)\right], \tag{A4}$$

$\gamma=\frac{1}{4}$:

$$\frac{24}{\pi a^4t^4}\left[1-e^{-\sqrt[4]{\pi}at}\left(1+\sqrt[4]{\pi}at+\frac{\sqrt{\pi}}{2}a^2t^2+\frac{\sqrt[4]{\pi^3}}{6}a^3t^3\right)\right]. \tag{A5}$$

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