Subdiffusive target problem: Survival probability

S. B. Yuste\textsuperscript{1} and Katja Lindenberg\textsuperscript{2}
\textsuperscript{1}Departamento de Fisica, Universidad de Extremadura, E-06071 Badajoz, Spain
\textsuperscript{2}Department of Chemistry and Biochemistry 0340 and Institute for Nonlinear Science, University of California–San Diego, 9500 Gilman Drive, La Jolla, California 92093-0340, USA
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The asymptotic survival probability of a spherical target in the presence of a single subdiffusive trap or surrounded by a sea of subdiffusive traps in a continuous Euclidean medium is calculated. In one and two dimensions the survival probability of the target in the presence of a single trap decays to zero as a power law and as a power law with logarithmic correction, respectively. The target is thus reached with certainty, but it takes the trap an infinite time on average to do so. In dimensions higher than two a single trap may never reach the target and so the survival probability is finite and, in fact, does not depend on whether the traps move diffusively or subdiffusively. When the target is surrounded by a sea of traps, on the other hand, its survival probability decays as a stretched exponential in all dimensions (with a logarithmic correction in the exponent for \( d = 2 \)). A trap will therefore reach the target with certainty, and will do so in a finite time. These results may be directly related to enzyme binding kinetics on DNA in the crowded cellular environment.

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I. INTRODUCTION

A number of recent experiments have pointed to the occurrence of subdiffusive motion in biophysical environments \cite{1–9}, that is, motion where the mean square displacement of a “walker” grows sublinearly in time, e.g.,

\[
\langle r^2(t) \rangle \sim t^\gamma, \quad 0 < \gamma < 1.
\] (1)

In particular, recent experiments on transport of large molecules in living cells indicate that the crowded cellular environment leads to such motion \cite{1–5}. The sublinear growth of the mean square displacement comes about because the presence of a large number of macromolecules leads to a medium that may present impediments such as barriers, traps, or otherwise interrupted pathways to the diffusive motions observed in typically much sparser environments designed in vitro. A number of experimental and theoretical papers have begun to address the problem of how to characterize the motion of proteins in living cells and, having characterized it as subdiffusive, of how to estimate the effects of this slowed-down transport on the binding and unbinding of enzyme proteins on DNA target sites \cite{5,8,10,11}. The results are not obvious and involve the balance between reaching a target site more slowly (which might tend to slow down reaction rates) while at the same time retaining the enzyme in the vicinity of a target site for a longer time (which might tend to speed it up). Furthermore, the breaking of ergodicity indicates that one might need to be careful about dealing with time histories of single events vs ensemble averages over many events \cite{11}. These issues arise not only in the biophysical context of cells but in other situations involving crowded environments such as porous media, but it is the cellular context that has awakened interest in the problem most recently. Various theories appear not to be entirely consistent with one another in their predictions. For example, Golding and Cox \cite{5} predict singular features in the binding/unbinding kinetics when the exponent \( \gamma \) crosses the value \( 2/3 \), while others \cite{10,11} do not seem to predict such features.

The theoretical approaches to the problem are fairly complex and range from continuous time random walk formalisms to scaling arguments. The literature that focuses on the biophysical context includes a number of realistic features such as the probability of a protein unbinding from the target site on the DNA before a reaction takes place and having to return to that site. These features add complications surrounding the appropriate boundary conditions. The relation between different boundary conditions used in this problem \cite{10,11} are not entirely clear and seem not to have been discussed in the literature. Furthermore, it may be the case that the final results of interest obey scaling laws that relate the more realistic problem to the simpler one in which binding definitely leads to reaction \cite{10}. In any case, it seems to us that even the “stripped-down” version of the problem wherein a reaction occurs with certainty when the target is reached would benefit from a systematic and clear presentation, and this is our goal in this paper.

Given the scenario of a target surrounded by a sea of subdiffusive particles the first of which to reach the target defines the binding time of interest, what exactly is it that one wishes to calculate? In some instances, the quantity calculated is the probability that a subdiffusive molecule initially at a distance \( r \) from the target ever finds it \cite{5,10,12,13}. In other instances, it is the distribution of the time-averaged probability of the subdiffusive molecule to be in the bound state (vs the unbound state) in a single trajectory \cite{11}. A standard classic measure is the distribution of first reaction times \cite{10,12,14}. We focus on a traditional quantity from which many of these can, in principle, be deduced under appropriate physical conditions, namely, the survival probability \( Q(t) \) of the target as a function of time \cite{13,15,16}. A specific scenario for this calculation arises when a target (perhaps DNA) site is surrounded by many randomly located particles (perhaps site-specific DNA-binding proteins). From the outset, we focus on the problem in a spatial continuum.
and thus rely on the fractional diffusion equation. We note that essentially all the dynamical theories on this topic in the literature either start from or arrive at a continuum formulation, even those that begin with a continuous time random walk, and we furthermore note that there seems to be little argument about the fact that at least asymptotically and on a spatial mesoscale such a formulation is appropriate. The differences in the literature lie not in the use of a fractional diffusion equation but rather in the boundary conditions [10,11,13]. In this context we note that while a portion of the literature deals with partially absorbing targets, that is, targets with a finite probability of letting the attached particle go (to eventually return and to again be trapped or not), or equivalently, with a finite rather than an infinite reaction rate when a particle meets the target, we deal with the fully absorbing target. The connection between the two types of targets may in many cases be straightforward [10].

The paper is organized as follows. In Sec. II we present the formalism for the calculation of the survival probability. The results for all dimensions are presented in Sec. III. Finally, we conclude with a brief summary in Sec. IV.

II. SURVIVAL PROBABILITY: THE FORMALISM

We start by defining \( Q_1(t,R) \) as the probability that a random walker ("the particle") that started at location \( \mathbf{r} \) at time \( t=0 \) has survived until time \( t \) in the presence of an absorbing sphere of radius \( R \) centered at the origin. Here \( R = |\mathbf{r}| \) in recognition of the orientational symmetry of the problem. The probability \( Q_1 \) is the main quantity from which other results are obtained. If instead of one we have \( N \) independent (sub)diffusive particles, their combined survival probability \( Q(t;R) \) is simply the probability that none of the \( N \) particles has entered the absorbing sphere. If we assume that the particles are randomly distributed in a volume \( V \), then

\[
Q(t;R) = \left[ \frac{1}{V} \int_{r>R} d\mathbf{r} Q_1(t,R;R) \right]^N,
\]

and if we further take the limits \( N \to \infty \) and \( V \to \infty \) holding the density \( \rho = N/V \) fixed, then this becomes

\[
Q(t;R) = \exp\left\{ -\rho \int_{r>R} d\mathbf{r} \left[ 1 - Q_1(t,R;R) \right] \right\}.
\]

The calculation of \( Q_1(t,R;R) \) for subdiffusive particles can be directly adapted from the corresponding calculation of this quantity for diffusive particles [17–19]. We introduce the probability density \( w(r',t;R;R) \) that the particle is at location \( \mathbf{r}' \) at time \( t \) if it started at position \( \mathbf{r} \) at \( t=0 \). As before, an absorbing sphere of radius \( R \) is centered at the origin \( \mathbf{r} = 0 \). The survival probability \( Q_1(t,R;R) \) is related to this probability density by

\[
Q_1(t,R;R) = \int d\mathbf{r}' w(r',t;R;R).
\]

The probability density obeys the fractional diffusion equation together with initial and boundary conditions,

\[
\frac{\partial}{\partial t} w(r',t;R;R) = \partial D_t^{1-\gamma}[D\nabla_t^2 w],
\]

where \( \partial D_t^{1-\gamma}[D\nabla_t^2 w] \) is the Riemann-Liouville operator

\[
\partial D_t^{1-\gamma}[f(t)] = \frac{1}{\Gamma(\gamma)} \frac{\partial}{\partial t} \int_0^t dt' f(t,t') (t-t')^{\gamma-1},
\]

and \( D_t^{1-\gamma} \) is a generalized diffusion coefficient.

Here \( \nabla_t^2 \) is the Laplacian operator with respect to the position \( \mathbf{r}' \), \( \partial D_t^{1-\gamma} \) is the Riemann-Liouville operator

\[
\partial D_t^{1-\gamma}[f(x,t)] = \frac{1}{\Gamma(\gamma)} \frac{\partial}{\partial t} \int_0^t dt' \int d\mathbf{x} f(x,t') (t-t')^{\gamma-1},
\]

and \( D_t^{1-\gamma} \) is a generalized diffusion coefficient.

Laplace transforming Eqs. (5), (7), and (8) with respect to time according to

\[
\tilde{g}(u) = \int_0^\infty dt e^{-ut} g(t)
\]

yields for \( \tilde{w}(r',u;R;R) \),

\[
u \tilde{w} - \partial \tilde{w}(r' - r) = u^{1-\gamma} D_t^{1-\gamma} \nabla_t^2 \tilde{w},
\]

and

\[
\tilde{w}(R,u;R;R) = 0,
\]

\[
\lim_{r' \to \infty} \tilde{w}(r',u;R;R) = 0.
\]

Note that \( \tilde{w}(r',u;R;R) \) is the Green function of Eq. (11). The reciprocity of the Green function with respect to the arguments \( r' \) and \( R \) then implies that it also satisfies the adjoint equation

\[
u \tilde{w} - \partial \tilde{w}(r' - r) = u^{1-\gamma} D_t^{1-\gamma} \nabla_t^2 \tilde{w},
\]

along with the boundary conditions [17]

\[
\tilde{w}(r',u;R;R) = 0,
\]

\[
\lim_{r' \to \infty} \tilde{w}(r',u;R;R) = 0.
\]

Integration over \( r' \) then gives for the Laplace transform \( \tilde{Q}_1(r,u;R) \) of the quantity of interest \( Q_1(t,R;R) \) [17],

\[
u \tilde{Q}_1 - 1 = u^{1-\gamma} D_t^{1-\gamma} \nabla_t^2 \tilde{Q}_1,
\]

\[
\tilde{Q}_1(R,u;R) = 0,
\]

\[
\lim_{r' \to \infty} \tilde{Q}_1(r',u;R) = \frac{1}{u}.
\]

Finally, the inverse Laplace transform yields the evolution equation and boundary and initial conditions for the survival probability of the particle that started its walk at \( \mathbf{r} \),

\[
\frac{\partial}{\partial t} Q_1(t,R;R) = \partial D_t^{1-\gamma}[D\nabla_t^2 Q_1],
\]
The last condition is a result of the certain survival at any finite time of a particle initially located at \( r \to \infty \). Due to the spherical symmetry of the problem, the \( d \)-dimensional Laplacian operator is
\[
\nabla^2_r = \frac{\partial^2}{\partial r^2} + \frac{d-1}{r} \frac{\partial}{\partial r},
\]
and the solution in Laplace space, valid for all \( d \) (even for noninteger \( d \)) is
\[
u \tilde{Q}_I(r,u;R) = 1 - \frac{r}{R} \left( \frac{\Gamma(d/2)}{\Gamma(2-d/2)} \right)^{1-d/2} \frac{K_{d/2-1}(\sqrt{ru}/D)}{K_{d/2-1}(\sqrt{R^2 u}/D)},
\]
which will be analyzed in more detail subsequently. Here \( d \) is the dimensionality and the \( K \)'s are modiﬁed spherical Bessel functions of the third kind [20]. Although under some circumstances it may be useful and even illuminating to consider noninteger values of \( d \) (for example, see [19]), our results are only physically relevant for dimensions for which the Laplacian operator (24) is meaningful, that is, for integer dimensions. As an aside, we note that this subdiffusive result is related to the single particle survival probability for a normal diffusive particle by the relation
\[
S(t) \gamma = \int_0^\infty dT S(T) \gamma = 1) T \gamma (t,t)
\]
or, in Laplace space,
\[
\tilde{S}(u) \gamma = u^{-1} \tilde{S}(u^2) \gamma = 1),
\]
where \( S(t) \gamma \) is the survival probability associated with a physical situation involving a subdiffusive particle and \( S(t) \gamma = 1) \) is the survival probability in the same physical situation but involving a normally diffusive particle, and \( T \gamma (t,t) \) is the “time-expanding transformation” [21]
\[
\tilde{T} \gamma (t,u) = u^{-1} \exp(-tu^2).
\]
In our case, Eq. (27) means that \( \tilde{Q}_I(r,u;R) \gamma = u^{-1} \tilde{Q}_I(r,u^2;R) \gamma = 1) \) [see 21,22] for more details on the integral transformation (26) and the scaling relation (27).

Before analyzing these results and using them to obtain explicit survival probabilities, we note that one can provide an alternative expression to Eq. (3) for the survival probability \( Q(t;R) \) in terms of \( Q_I \). For this purpose, we define [19]
\[
f(t;R) = \int_{r>R} dt [1 - Q_I(t;R)].
\]
Note that \( f(0;R) = 0 \) because \( Q_I(t;R) = 1 \). Taking a time derivative of this function,
\[
\frac{df}{dt}(t;R) = - \int_{r>R} dr \frac{d}{dt} Q_I(t;R),
\]
and using Eq. (20), we find that
\[
\frac{df}{dt}(t;R) = - \int_{r>R} dr \frac{t}{D} \tilde{Q}_I^{1-\gamma}[D \tilde{\nabla}^2 \tilde{Q}_I(t;R)]
\]
\[
= -D \tilde{D}^{1-\gamma} \int_A dA \cdot \tilde{\nabla} \tilde{Q}_I(t;R)
\]
\[
= DS_q \tilde{R}^{d-1} \tilde{D}^{1-\gamma} \frac{1}{\tilde{D}^{d-1}} \tilde{Q}_I(t;R)\bigg|_{r=R},
\]
where \( S_q = 2 \pi^{d/2} / \Gamma(d/2) \) is the surface of a sphere of unit radius, and \( dA \) is a surface element in the direction perpendicular to the surface of the sphere. Since \( f(0;R) = 0 \), we can write
\[
f(t;R) = \int_0^t \frac{df}{d\tau}(\tau;R) = DS_q \tilde{R}^{d-1} F(t;R),
\]
with
\[
F(\tau;R) = \int_0^\tau \frac{df}{d\tau}(\tau;R) = DS_q \tilde{R}^{d-1} F(t;R),
\]
and consequently,
\[
Q(t;R) = \exp[-\nu DS_q \tilde{R}^{d-1} F(t;R)].
\]
We will use this route in our calculations. Our task is thus to calculate \( Q_I(t;R) \) and from it \( F(t;R) \), to finally arrive at the survival probability \( Q(t;R) \).

### III. SURVIVAL PROBABILITY: RESULTS

In this section we present asymptotic results for the survival probabilities in all dimensions.

#### A. \( d < 2 \)

For \( d < 2 \) we find that as \( u \to 0 \),
\[
\tilde{Q}_I(r,u;R) \sim \frac{\Gamma(d/2)(r^{2-d} - R^{2-d})}{2^{2-d} \Gamma(2-d/2)} \frac{D^{d+2}}{u^{1-\gamma}}
\]
and using a Tauberian theorem this leads to
\[
Q_I(t;R) \sim \frac{\Gamma(d/2)(r^{2-d} - R^{2-d})}{2^{2-d} \Gamma(2-d/2)} \frac{D^{d+2}}{r^{1-\gamma} \Gamma(1-\gamma+\gamma d/2)},
\]
but
\[
\frac{d}{dt}\tilde{D}^{1-\gamma} \tilde{Q}_I(t;R)\bigg|_{r=R} \sim \frac{\Gamma(d/2)(2-d)R^{1-d}D^{d+2-1(\gamma-\gamma d/2)}}{2^{2-d} \Gamma(2-d/2) \Gamma(\gamma d/2)}.
\]
\[ F(t; R) = \int_0^t dt_1 D_t^{-\gamma} \frac{\partial}{\partial r} Q_t(r; R) \bigg|_{r=R} \sim \frac{2^{d-1} \Gamma(d/2) R^{1-d} D_t^{d/2-1} \gamma d/2}{\Gamma(1-d/2) \Gamma(1+\gamma d/2)}. \]

It then follows that
\[ Q(t; R) \sim \exp \left( -\rho \frac{(4\pi D t)^{d/2}}{\Gamma(1+\gamma/2)} \right), \quad t \to \infty. \]

Note that this is independent of the radius \( R \) of the absorbing sphere. Note also that whereas the survival probability of the target in the presence of a single (sub)diffusive particle decays as a power law with time, the decay becomes a stretched exponential when there are many particles at initially random locations.

With \( d=1 \) this coincides with the result we previously reported in [23],
\[ Q(t; R) \sim \exp \left( -\rho \sqrt{4D t} \right), \quad t \to \infty. \]

When in addition \( \gamma=1 \),
\[ Q(t; R) \sim \exp \left( -\sqrt{\frac{4\pi D t}{\pi}} \right), \quad t \to \infty, \]

which coincides with the result reported in [19].

Actually, for \( d=1 \) the survival probability of the target in the presence of a single (sub)diffusive particle can be given exactly [and the result (36) is then the asymptotic expansion of this exact result] as follows:
\[ \bar{Q}_1(r,u;R) = \frac{1}{u} \exp \left( (R-r) \sqrt{u / D} \right), \]
so that
\[ Q_1(r,t;R) = 1 - H_{11}^{10} \left[ \frac{r-R}{D t^{\gamma/2}} \right] (1, \gamma/2) (0, 1). \]

Here \( H \) is a Fox function. The probability density function \(-dQ_1/dt\) of first passage times corresponding to Eq. (42) agrees with the one previously obtained by Balakrishnan [14] (see also Ref. [12], where \(-dQ_1/dt\) is given in terms of a one-sided Lévy stable density). When \( \gamma=1 \) the Fox function reduces to the complementary error function and Eq. (43) then reduces to the classic diffusive result [18].

### B. \( d=2 \)

In two dimensions the solution of the fractional diffusion equation with the appropriate initial and boundary conditions leads to
\[ \bar{Q}_1(r,u;R) \sim \frac{1}{u} \frac{\ln(R/r)}{\gamma \ln(u/\gamma)}, \quad u \to 0, \]
\[ \bar{Q}_1(r,u;R) \sim \frac{1}{u} \frac{\ln(R/r)}{\gamma \ln[u/\gamma]}, \quad u \to 0, \]
where \( \gamma = 0.577216 \) is the Euler-Mascheroni constant. Applying the Tauberian theorem then leads to the asymptotic result
\[ Q_1(r,t;R) \sim \frac{2 \ln(r/R)}{\ln(4D t^{\gamma}/R^2)} = \frac{2 \ln(r/R)}{\gamma \ln(at)}, \]

To calculate \( F(t; R) \) we need to evaluate the fractional derivative \( G(t) = \partial^\gamma D_t \) with \( g(t) = 1/\ln(at) \). We know that
\[ G(u) = u^{1-\gamma} \bar{G}(u) = \lim_{t \to 0} \frac{1}{\Gamma(\gamma)} \int_0^t ds \frac{g(s)}{(t-s)^{1-\gamma}}. \]

However, the limit term on the right can easily be shown to vanish. Since \( \bar{G}(u) = [u \ln(u/a)]^{-1} \) as \( u \to 0 \), it follows that \( \bar{G}(u) \sim [u^{1-\gamma} \ln(u/a)]^{-1} \) and consequently, \( G(t) \sim t^{\gamma-1}/[\Gamma(\gamma) \ln(at)] \) as \( t \to \infty \). We thus have that
\[ F(t; R) = \frac{2}{R \gamma} \int_0^t \frac{\pi G(\tau)}{\gamma \ln(4D t^{\gamma}/R^2)} dt, \]

and consequently,
\[ Q(t; R) \sim \exp \left( -\rho \sqrt{\frac{4\pi D t}{\pi}} \right), \quad t \to \infty. \]

While the decay of the survival probability of a single particle is thus an inverse logarithm, that of the ensemble of particles is a stretched exponential with a logarithmic correction. Also, when \( d=2 \) [and also when \( d>2 \) (see below)] the survival probabilities do depend on the radius \( R \) of the absorbing sphere.

### C. \( d>2 \)

For \( d>2 \) we have that
\[ \bar{Q}_1(r,u;R) \sim \frac{1}{u} \left[ 1 - \left( \frac{R}{r} \right)^{d-2} \right], \quad u \to 0, \]
so that
\[ Q_1(r,t;R) \sim \left[ 1 - \left( \frac{R}{r} \right)^{d-2} \right], \quad t \to \infty. \]

From this we obtain
\[ F(t) = \int_0^t dt_1 \partial_t D_t^{-\gamma} \frac{\partial}{\partial r} Q_t(r; R) \bigg|_{r=R} = \frac{d-2}{R} \frac{t^{\gamma}}{\Gamma(1+\gamma)}, \quad t \to \infty, \]
from which it follows that
\[ Q(t; R) = \exp \left\{ -\frac{S_d R^{d-2}(d-2)}{\Gamma(1 + \gamma)} Dr^\gamma \right\}, \quad r \to \infty, \quad (53) \]

in agreement with results reported in [13,16]. The survival probability of a single particle approaches a constant at long times, whereas the ensemble survival probability decays as a stretched exponential.

As in the one-dimensional case, for \( d=3 \) it is again possible to provide an exact survival probability of the target in the presence of a single (sub)diffusive particle as follows:

\[ \bar{Q}_1(r,u;R) = \frac{1}{u} - \frac{R}{r} e^{(r-u)/D} \left( 1, \frac{1}{2} \right), \quad (54) \]

so that

\[ Q_1(r,t;R) = 1 - \frac{R}{r} H_{1/2}^1 \left( \frac{r-R}{\sqrt{Dt}}, (0,1) \right). \quad (55) \]

This Fox \( H \) function reduces to a complementary error function when \( \gamma=1 \), and in this limit we recover the classic normal diffusion result (see e.g., [18]). It is interesting to note that for \( d=3 \) this result is the same as Eq. (5.5) in [10] if we take the completely absorbing limit \( k_\gamma \to \infty \) in that formula. Note that the probability density function \( -dQ_1/\partial t \) of first passage times corresponding to Eqs. (54) and (55) agrees with the one previously obtained by Barkai [12].

IV. CONCLUSIONS

We have calculated the asymptotic survival probability of an absorbing target of radius \( R \) at the origin in the presence of one, or of many, (sub)diffusive particles. This calculation, which is based on the fractional diffusion equation, has been carried out for all dimensions, with results that in some cases and limits agree with known results. Equations (36) and (39) give these survival probabilities for \( d=1 \), Eqs. (45) and (48) for \( d=2 \), and Eqs. (50) and (53) for \( d \geq 3 \). Thus, in one dimension the survival probabilities are, respectively, of power law and stretched exponential form, in two dimensions the decay is slower, respectively, logarithmic and stretched exponential with a logarithmic correction. The result in dimensions three [10] and higher are interesting: the survival probability of a single particle goes asymptotically to a constant (i.e., the particle may survive forever with a finite probability), and this probability is independent of the subdiffusive exponent, and thus the same as for a normally diffusive particle. The survival probability of the target surrounded by a sea of subdiffusive traps does decay, again as a stretched exponential that does depend on the subdiffusive exponent. Note that in all cases, while the mean survival time of the target in the presence of a single particle (i.e., the first moment of \( Q_1 \)) is infinite, that of a target in a large (infinite) volume containing a finite density of particles (i.e., the first moment of \( Q \)) is finite. This may be an interesting observation beyond the biophysical examples mentioned in the Introduction, for example, in the search by an enzyme of a DNA target site involving a combination of scanning and relocation events in which the relocation times have a power law distribution with diverging moments. The mean survival time when a single enzyme seeks the target in this three-dimensional search diverges (the survival probability of the target is a power law without moments), but one of many enzymes will reach the target with certainty (the survival probability of the target is a stretched exponential).

The quantities that we have calculated lead immediately to others frequently used in the literature as well as to additional insights. For example, the derivatives \( -\partial Q_1/\partial t \) and \( -\partial Q_1/\partial t \) are the distributions of first passage times to the absorbing target. It is also noteworthy that the probability that a subdiffusive molecule initially at a distance \( r \) from the target ever reaches it is the same for normally diffusive and subdiffusive particles. In particular, since \( \lim_{t \to \infty} Q_1(r,t;R) = Q_1(r,\infty;R) \) vanishes for \( d=1 \) and \( d=2 \), a (sub)diffusive particle will eventually reach the target with certainty. On the other hand, since for \( d \geq 3 \) this limit is finite, \( 1-(R/r)^{d/2} \), a particle escapes the target forever with probability \( (R/r)^{d/2} \). Whether it is diffusive or subdiffusive. When the target is surrounded by a sea of particles, however, since \( \lim_{t \to \infty} Q(r,t;R) = Q(r,\infty;R) \) vanishes for all dimensions, one of the particles, whether diffusive or subdiffusive, eventually reaches the target with certainty. The approach to these asymptotic behaviors of course depends on dimensionality and also on the subdiffusive exponent \( \gamma \).

Our immediate future work on this topic will focus on the effects of a partially absorbing target, that is, a target that does not necessarily “die” upon its first encounter with a (sub)diffusive particle. Some results on this case have been reported in the literature, notably in [10,11], but they do not use the same boundary conditions and they also do not consider this situation in all dimensions. Clearly, there is still work to be done.

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