

Survival probability of an immobile target in a sea of evanescent diffusive or subdiffusive traps: A fractional equation approach

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We calculate the survival probability of an immobile target surrounded by a sea of uncorrelated diffusive or subdiffusive evanescent traps (i.e., traps that disappear in the course of their motion). Our calculation is based on a fractional reaction-subdiffusion equation derived from a continuous time random walk model of the system. Contrary to an earlier method valid only in one dimension ($d = 1$), the equation is applicable in any Euclidean dimension d and elucidates the interplay between anomalous subdiffusive transport, the irreversible evanescence reaction, and the dimension in which both the traps and the target are embedded. Explicit results for the survival probability of the target are obtained for a density $\rho(t)$ of traps which decays (i) exponentially and (ii) as a power law. In the former case, the target has a finite asymptotic survival probability in all integer dimensions, whereas in the latter case there are several regimes where the values of the decay exponent for $\rho(t)$ and the anomalous diffusion exponent of the traps determine whether or not the target has a chance of eternal survival in one, two, and three dimensions.

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I. INTRODUCTION

Geometric and dynamical constraints imposed by complex or crowded environments often result in subdiffusive behavior; that is, in sublinear growth of a particle's mean squared displacement at long times. However, a complete description of the underlying transport process at a mesoscopic level must go beyond the mean squared displacement and involve other properties of experimental interest which may be studied via suitable quantifiers [1,2]. This may help one to discriminate between models when describing realistic experimental situations where subdiffusive (or, more generally, anomalous) transport is observed.

The detailed microscopic subdiffusive transport mechanism is often unknown, and so the literature is populated with a number of different models. One popular choice to mimic situations of experimental interest is the continuous time random walk (CTRW) model [3] with a long-tailed waiting time distribution. The CTRW model has been used successfully as a phenomenological model to describe aging effects in systems as diverse as stock markets [4–7], charge carrier transport in disordered media [8], luminescence quenching in micellar clusters [9], transport in porous media [10,11], escape problems [12], and morphogen gradient formation [13–15].

From a mathematical point of view, the CTRW with a long-tailed waiting time distribution and a jump length distribution of finite variance is known to be equivalent to a fractional diffusion equation in the long-time limit; that is, a diffusion equation with fractional time derivatives rather than ordinary derivatives [16]. Despite the fact that fractional derivatives are nonlocal integrodifferential operators, Laplace transform techniques commonly used for the solution of the ordinary diffusion equation remain applicable and can be used to tackle a wide variety of these problems [17].

One compelling reason to work with CTRW models and the associated fractional equations is that they make it possible to

include reactive processes. We introduce this terminology in the broadest sense of including particle destruction, creation, binding, or transformation processes. While the combination of subdiffusion with its memory effects and reaction processes is complex, at least the CTRW approach offers a way to consider them in combination; something that has proved more elusive with other approaches. In some fortunate situations, the effect of the reactions can be adequately described by suitable boundary conditions imposed upon the corresponding fractional diffusion equation (see, e.g., Ref. [18], Sec. 4.1 in Ref. [17], and references therein); however, such situations are rather exceptional, since in general the combination of reaction with non-Markovian kinetics [19] leads to nonintuitive fractional equations where the parameters describing the chemical kinetics appear in a nonuniversal, model-dependent fashion [20–24]. In particular, heuristic approaches based on fractional equations with separate reaction and transport terms such as we are accustomed to in ordinary reaction-diffusion problems very often lead to unphysical results even in the simplest cases of irreversible first-order reactions.

While fractional reaction-subdiffusion equations have been used to investigate a number of different problems corresponding to different mesoscopic models and different boundary conditions [14,25–30], many subdiffusive versions of classical reaction-diffusion problems [31] remain unexplored. Thus, one can legitimately claim that the field is still in its infancy. One class of problems that has attracted considerable interest in recent years concerns target search processes driven by (sub)diffusion. Such processes are ubiquitous in nature and include binary searches where two objects must meet for a reaction or trapping event to occur. In many instances, Smoluchowski's theory of diffusion-controlled reactions turns out to be a successful tool for the quantitative characterization of diffusional target search. Examples include scavenging reactions [32,33], site location in DNA [34], ligand binding to sites on macromolecules [35], predator-prey models [36],

luminescence quenching [37], intermittent search processes [38], and search processes with resetting to the initial position [39], to name but a few. In this context a key quantity is the so-called *survival probability* of the target, from which the moments of the first-passage-time distribution for target annihilation can also be straightforwardly computed [40,41].

In recent years the classic diffusional target search problem has been generalized to particles that undergo anomalous diffusion [42–45]. In this paper we consider a related problem; namely, the survival probability of an immobile target immersed in a sea of uncorrelated subdiffusive traps *that may die “spontaneously” in the course of their motion*. In other words, there are now two reactions occurring simultaneously: the disappearance of the target and a trap upon encounter with each other, and the disappearance of the traps due to some *other* physical process. We term this latter process “spontaneous” as a way to recall that it is not induced by collision with the target. The spontaneous evanescence process may, for instance, be triggered by particle scavengers in the system, but for practical purposes any process that turns off the interaction between a trap and the target can also be thought of as an evanescence or death process.

A solution to this problem in dimension $d = 1$ was given in Ref. [46] using a functional method first developed by Bray *et al.* [47] for the diffusive case. Here we approach the problem from a different point of view that allows us to also obtain results in higher dimensions. In particular, we make use of a recently derived reaction-subdiffusion equation obtained from a mesoscopic CTRW model with a long-tailed waiting time distribution and a superimposed reactive process. The elimination of the fixed target is incorporated as a boundary condition, while the decay mechanism of the traps as they move subdiffusively is modeled by a phenomenological choice of a monotonically decaying functional form for the trap density $\rho(t)$.

The paper is organized as follows: In Sec. II we briefly recall the results for the survival probability of a target in a sea of nonevanescing traps. Our new general results for the survival probability of the target when the traps are evanescent are presented in Sec. III. In Sec. IV we implement these results for particular forms of evanescence; namely, exponential and power law. We conclude with a short summary in Sec. V.

II. RECAP OF RESULTS FOR NONEVANESCENT TRAPS

We consider a statistical ensemble of systems each of which is composed of a fixed hyperspherical target of radius R located at the center $r = 0$ of a large d -dimensional volume V . Each volume V initially contains N_0 randomly distributed noninteracting point traps. At time $t = 0$ the traps begin to move subdiffusively. If any of them hits the surface of the target, both the target and the trap are instantaneously annihilated (fully absorbing case). Our goal is to compute the survival probability of the target at time t (i.e., the probability that no traps have collided with the target up to this time). In this section, we briefly recall previous results obtained in Ref. [42] when the traps are not subject to spontaneous evanescence. In the next section we use these results to obtain the solution when the subdiffusive traps evanesce.

In the absence of evanescence, the motion of each trap is dictated by the fractional diffusion equation

$$\frac{\partial w(\mathbf{r}, t | \mathbf{r}_0; 0)}{\partial t} = K_\gamma {}_0 D_t^{1-\gamma} \nabla_{\mathbf{r}}^2 w(\mathbf{r}, t | \mathbf{r}_0; 0), \quad 0 < \gamma \leq 1, \quad (1)$$

where $w(\mathbf{r}, t | \mathbf{r}_0; 0)$ is the probability density of finding the trap at location \mathbf{r} at time t if it started at position \mathbf{r}_0 at $t = 0$, K_γ is the anomalous diffusion coefficient, and $\nabla_{\mathbf{r}}^2$ stands for the Laplacian operator with respect to the position \mathbf{r} . The operator ${}_0 D_t^{1-\gamma}$ is the Riemann-Liouville fractional derivative, defined as follows [48]:

$${}_0 D_t^{1-\gamma} f(r, t) = \frac{1}{\Gamma(\gamma)} \frac{\partial}{\partial t} \int_0^t dt' \frac{f(r, t')}{(t-t')^{1-\gamma}}. \quad (2)$$

Strictly speaking, the operator appearing in the derivation of Eq. (1) is the Grünwald-Letnikov fractional derivative rather than the Riemann-Liouville derivative. However, both operators are identical for sufficiently smooth functions [48], as are all the functions we encounter in this problem. The propagator solution of the fractional diffusion equation (1) yields a mean squared displacement with the long-time behavior $\langle r^2 \rangle \sim K_\gamma t^\gamma$ resulting in subdiffusive behavior when γ is less than unity.

Let $Q_T(t; R)$ denote the ensemble averaged survival probability of the target in a sea of randomly distributed uncorrelated traps. This quantity can be obtained from the survival probability of the target in the presence of a *single* trap starting at location \mathbf{r}_0 , $Q_{1,T}(\mathbf{r}_0, t; R)$. We shall focus on the thermodynamic limit; that is, we take $N_0 \rightarrow \infty$ and $V \rightarrow \infty$ while keeping a fixed global initial trap density $\rho_0 = \lim_{N_0, V \rightarrow \infty} N_0/V$. In this limit one has

$$\begin{aligned} Q_T(t; R) &= \lim_{N_0, V \rightarrow \infty} \left[\frac{1}{V} \int_{r_0 > R} Q_{1,T}(\mathbf{r}_0, t; R) d\mathbf{r}_0 \right]^{N_0} \\ &= \exp\{-\rho_0 R^d \sigma(t, R)\}, \end{aligned} \quad (3)$$

where the integration is carried out over the volume that is exterior to the target. We have introduced the auxiliary quantity

$$\begin{aligned} \sigma(t; R) &\equiv \frac{1}{R^d} \int_{r_0 > R} [1 - Q_{1,T}(\mathbf{r}_0, t; R)] d\mathbf{r}_0 \\ &= -\frac{1}{\rho_0 R^d} \ln Q_T(t; R). \end{aligned} \quad (4)$$

Note that the survival probability $Q_{1,T}(\mathbf{r}_0, t; R)$ of the target is identical with the survival probability $Q_1(\mathbf{r}_0, t; R)$ of the trap, because we have assumed that both the target and the trap disappear instantaneously upon encounter (i.e., there is only one decay channel for both particles). Note also that this is no longer the case when the traps undergo spontaneous evanescence. In that case one has $Q_{1,T} > Q_1$ (see next section).

In order to compute $Q_1(\mathbf{r}_0, t; R)$, we must first define the relevant boundary value problem by complementing Eq. (1) with the deterministic initial condition,

$$w(\mathbf{r}, 0 | \mathbf{r}_0; 0) = \delta(\mathbf{r} - \mathbf{r}_0), \quad (5)$$

and the boundary conditions

$$w(R, t | \mathbf{r}_0; 0) = 0, \quad (6)$$

$$\lim_{r \rightarrow \infty} w(\mathbf{r}, t | \mathbf{r}_0; 0) = 0. \quad (7)$$

The boundary condition (6) reflects the fully absorbing nature of the target, which prevents the trap from being found on the target surface or inside the target. The solution $w(\mathbf{r}, t | \mathbf{r}_0; 0)$ is related to $Q_1(\mathbf{r}_0, t; R)$ via the integral relation

$$Q_1(\mathbf{r}_0, t; R) = \int w(\mathbf{r}, t | \mathbf{r}_0; 0) d\mathbf{r}. \quad (8)$$

The spherical symmetry of the target means that $Q_1(\mathbf{r}_0, t; R)$ only depends on the initial distance r_0 of the trap to the target. For this reason, we shall drop the subindex of r_0 and from here on use the simpler notation $Q_1(r, t; R)$. Taking into account Eq. (8), the boundary value problem stated directly in terms of $Q_1(r, t; R)$ then is

$$\frac{\partial Q_1(r, t; R)}{\partial t} = K_\gamma {}_0D_t^{1-\gamma} \nabla_r^2 Q_1(r, t; R), \quad (9a)$$

$$Q_1(r, 0; R) = 1, \quad (9b)$$

$$Q_1(R, t; R) = 0, \quad (9c)$$

$$\lim_{r \rightarrow \infty} Q_1(r, t; R) = 1. \quad (9d)$$

The second equation in this set corresponds to the initial condition and is self-explanatory, while the third one is a boundary condition which reflects yet again the fully absorbing nature of the target. The last equation states that a trap which is “pushed” infinitely far away from the target will survive forever, because its only decay channel is provided by the interaction with the target.

The above problem can be solved exactly in Laplace space [42]:

$$u Q_1(r, u; R) = 1 - \left(\frac{r}{R}\right)^{1-\frac{d}{2}} \frac{K_{d/2-1}(\sqrt{r^2 u^\gamma / K_\gamma})}{K_{d/2-1}(\sqrt{R^2 u^\gamma / K_\gamma})}, \quad (10)$$

where $K_{d/2-1}(\cdot)$ is a modified Bessel function of the second kind and $Q_1(r, u; R) = \int_0^\infty e^{-ut} Q_1(r, t; R) dt$ denotes the Laplace transform (the function and its Laplace transform are clearly distinguished by the argument and so we use the same designation for both). Alternatively, the solution $Q_1(r, u; R | \gamma)$ for $\gamma < 1$ can be found from the corresponding solution for normal diffusion ($\gamma = 1$) by means of the “time-expanding transformation” [49, 50] associated with the so-called subordination principle: $u Q_1(r, u; R | \gamma) = u^\gamma Q_1(r, u^\gamma; R | \gamma = 1)$.

For $d = 1$ and $d = 3$ the Bessel functions can be expressed in terms of exponentials, and explicit exact solutions are available for arbitrary times t . In other dimensions simple expressions are only available at long times. Setting $Q_{1,T} = Q_1$ in Eq. (3), Tauberian theorems can be used to find the long-time behavior in the multiple trap problem [42]:

$$\sigma(t; R) \propto \begin{cases} t^{\gamma/2}, & d = 1 \\ t^\gamma / \ln(\alpha_\gamma t), & d = 2 \\ t^\gamma, & d \geq 3, \end{cases} \quad (11)$$

where $\alpha_\gamma = (4K_\gamma / R^2)^{1/\gamma}$.

Thus, the survival probability of the target goes to zero in all dimensions d . This result is in strong contrast with the single-trap problem, since in that case the probability that the

random walk of the trap never intersects the target becomes nonzero as soon as $d \geq 3$.

III. SURVIVAL PROBABILITY FOR EVANESCENT TRAPS: GENERAL EXPRESSIONS

The behavior of the survival probability of the target changes completely if the traps disappear in the course of their motion. We assume a spontaneous evanescence process; specifically, that the decrease of the global trap density $\rho(t)$ is described by the following differential equation:

$$\dot{\rho}(t) \equiv \frac{d\rho(t)}{dt} = -\lambda(t)\rho(t), \quad (12)$$

where $\lambda(t) > 0$ is a rate coefficient which is in general time dependent. The solution $\rho_0 \exp[-\int_0^t \lambda(t') dt']$ yields a decaying density of surviving traps which describes the time evolution of the trap density in the absence of the fully absorbing target. The case $\lambda = \text{constant}$ leads to an exponentially decaying density.

Our main goal is to compute the survival probability $Q_T^*(t; R)$ of the target at time t (we use survival probabilities with a star to distinguish these quantities from their counterparts in the absence of trap evanescence). We follow the strategy of the previous section; namely, to derive the solution from the single-trap case.

We wish to combine the effects of Eqs. (12) and (1). One might be tempted to proceed as in the case of ordinary diffusion and simply construct some superposition of transport and reaction terms. However, a careful analysis shows that this is *incorrect*. Instead, a rigorous derivation starting at the level of the CTRW shows that the correct equation is [24]

$$\begin{aligned} \frac{\partial w(\mathbf{r}, t | \mathbf{r}_0, 0)}{\partial t} &= \frac{\rho(t)}{\rho_0} K_\gamma {}_0D_t^{1-\gamma} \frac{\rho_0}{\rho(t)} \nabla_r^2 w(\mathbf{r}, t | \mathbf{r}_0, 0) \\ &+ \frac{\dot{\rho}(t)}{\rho(t)} w(\mathbf{r}, t | \mathbf{r}_0, 0). \end{aligned} \quad (13)$$

It is straightforward to show that the survival probability $Q_1^*(\mathbf{r}_0, t; R) = Q_1^*(r_0, t; R) = \int w(\mathbf{r}, t | \mathbf{r}_0; 0) d\mathbf{r}$ of the trap then obeys the equation

$$\begin{aligned} \frac{\partial Q_1^*(r, t; R)}{\partial t} &= \frac{\rho(t)}{\rho_0} K_\gamma {}_0D_t^{1-\gamma} \left(\frac{\rho_0}{\rho(t)} \nabla_r^2 \right) Q_1^*(r, t; R) \\ &+ \frac{\dot{\rho}(t)}{\rho(t)} Q_1^*(r, t; R), \end{aligned} \quad (14a)$$

where we have again dropped the subindex from r_0 for notational convenience. The above equation must now be complemented with the conditions

$$Q_1^*(r, 0; R) = 1, \quad (14b)$$

$$Q_1^*(R, t; R) = 0, \quad (14c)$$

$$\lim_{r \rightarrow \infty} Q_1^*(r, t; R) = \frac{\rho(t)}{\rho_0}. \quad (14d)$$

Note that the last equation is substantially different from the corresponding one in the absence of the evanescence process. Indeed, even the survival probability of a trap which is at an infinite distance from the target decays in time because of the

evanescence reaction, and this probability is equal to the ratio of the global trap density at time t in the absence of the target and the initial density ρ_0 .

At this stage, one can easily check that if one performs the transformation $Q_1^*(r, t; R) = [\rho(t)/\rho_0]Q(r, t; R)$ in Eqs. (14), the resulting set of equations for the transformed function Q is identical with the boundary value problem (9) for Q_1 . Because of the uniqueness of the solution, we thus conclude $Q \equiv Q_1$ and, furthermore,

$$Q_1^*(r, t; R) = \frac{\rho(t)}{\rho_0} Q_1(r, t; R). \quad (15)$$

This expression is intuitively clear: it simply states that the probability that up to time t the trap has neither evanesced (one decay channel) nor hit the target (another decay channel) is equal to the product of the probability $\rho(t)/\rho_0$ that the trap has not evanesced, and the conditional probability that it has not hit the target given that it has not previously evanesced. The latter probability is precisely the survival probability of the trap when no evanescence is at play.

The next step in our route to the solution for the multitraps problem is to derive a relation between $Q_{1,T}^*(r, t; R)$ and $Q_1^*(r, t; R)$. As already anticipated in the previous section, the evanescence of the trap implies that the survival probability of the target and the trap are no longer the same. Let t' be a time in the interval $(0, t)$. In the single-trap problem, the infinitesimal probability $\{d[1 - Q_{1,T}^*(r, t'; R)]/dt'\}dt'$ that the target is annihilated by collision with the trap during the interval $(t', t' + dt')$ is the product of two factors: (i) the probability $\{d[1 - Q_1(r, t'; R)]/dt'\}dt'$ that the trap collides with the target during the time interval $(t', t' + dt')$ given that it has not previously evanesced, and (ii) the probability $\rho(t')/\rho_0$ that up to time t' the trajectory of that trap is not interrupted by an evanescence event. Thus,

$$\frac{d}{dt'}[1 - Q_{1,T}^*(r, t'; R)]dt' = \frac{\rho(t')}{\rho_0} \frac{d}{dt'}[1 - Q_1(r, t'; R)]dt'. \quad (16)$$

We next implement a number of steps [integrate this equation, integrate by parts, and use Eq. (15)] to obtain

$$Q_{1,T}^*(r, t; R) = Q_1^*(r, t; R) - \int_0^t Q_1^*(r, t'; R) \frac{\dot{\rho}(t')}{\rho(t')} dt', \quad (17)$$

which quantifies the difference between $Q_{1,T}^*(r, t; R)$ and $Q_1^*(r, t; R)$.

Having clarified the relation between $Q_{1,T}^*(r, t; R)$ and $Q_1(r, t; R)$ (survival probability of a single nonevanescing trap) and between $Q_{1,T}^*(r, t; R)$ (survival probability of the target in the presence of a single evanescent trap) and $Q_1^*(r, t; R)$ (survival probability of a single evanescent trap), we are now ready to tackle the multiple trap problem by proceeding as in the previous section (i.e., by using the statistical independence of the traps). Equations (3) (for the survival probability of the target in the presence of a collection of traps) and (4) are now respectively replaced with

$$\begin{aligned} Q_T^*(t; R) &= \lim_{N_0, V \rightarrow \infty} \left[\frac{1}{V} \int_{r>R} Q_{1,T}^*(r, t; R) dr \right]^{N_0} \\ &= \exp\{-\rho_0 R^d \sigma^*(t, R)\}, \end{aligned} \quad (18)$$

and

$$\sigma^*(t; R) = \frac{1}{R^d} \int_{r>R} [1 - Q_{1,T}^*(r, t; R)] dr \quad (19)$$

$$= -\frac{1}{\rho_0 R^d} \ln Q_T^*(t; R). \quad (20)$$

Taking the derivative of Eq. (19) with respect to time and using Eq. (17) we get

$$\frac{\partial \sigma^*(t; R)}{\partial t} = -\frac{1}{R^d} \int_{r>R} \left[\frac{\partial Q_1^*}{\partial t} - \frac{\dot{\rho}(t)}{\rho(t)} Q_1^* \right] dr. \quad (21)$$

Next we use Eq. (14a) and the relation (15) in the right-hand side of Eq. (21) and apply Gauss's theorem to change the volume integral to a surface integral. This allows us to write

$$\frac{\partial \sigma^*(t; R)}{\partial t} = S_d \frac{K_\gamma}{R} \frac{\rho(t)}{\rho_0} D_t^{1-\gamma} \left. \frac{\partial Q_1(r, t; R)}{\partial r} \right|_{r=R}, \quad (22)$$

where $S_d = 2\pi^{d/2}/\Gamma(d/2)$ denotes the surface of a d -dimensional hypersphere of unit radius. Finally, integrating from 0 to t and using the condition $\sigma^*(0; R) = 0$ [$Q_{1,T}^*(0, R) = 1$] we obtain the general formula

$$\sigma^*(t; R) = S_d \frac{K_\gamma}{R} \int_0^t \left[D_{t'}^{1-\gamma} \left. \frac{\partial Q_1(r, t'; R)}{\partial r} \right|_{r=R} \right] \frac{\rho(t')}{\rho_0} dt', \quad (23)$$

which extends the result for nonevanescing traps obtained in Ref. [42] to the case of evanescent traps. More specifically, we have again related the logarithm of the survival probability of the target, now in the presence of a collection of *evanescent* traps, to the survival probability of the target in the presence of a single nonevanescing trap. If the trap density $\rho(t)$ decays sufficiently rapidly, then $Q_{1,T}^*(t \rightarrow \infty, R) > 0$ and $\sigma^*(t \rightarrow \infty; R) < \infty$, and it is possible to conclude from Eq. (23) that

$$\begin{aligned} \sigma^*(\infty; R) - \sigma^*(t; R) &= S_d \frac{K_\gamma}{R} \int_t^\infty \left[D_{t'}^{1-\gamma} \left. \frac{\partial Q_1(r, t'; R)}{\partial r} \right|_{r=R} \right] \frac{\rho(t')}{\rho_0} dt'. \end{aligned} \quad (24)$$

To continue from here we need to specify the dimensionality explicitly, and also the explicit form of the trap density as a function of time. First we discuss the dimensionality. The case $d = 1$ has been dealt with in Ref. [46]; suffice it to say that we recover the result obtained therein:

$$\sigma^*(t; R) = \frac{1}{\rho_0 R} \frac{2\sqrt{K_\gamma}}{\Gamma(\gamma/2)} \int_0^t \rho(t') t'^{\gamma/2-1} dt', \quad (25)$$

where we have made use of the explicit one-dimensional form of Q_1 in terms of the Fox H function

$$Q_1(r, t; R) = 1 - H_{11}^{10} \left[\frac{r - R}{\sqrt{K_\gamma t^\gamma}} \middle| \begin{matrix} (1, \gamma/2) \\ (0, 1) \end{matrix} \right] \quad (26)$$

for the survival probability of a nonevanescing trap. For $d = 3$, and exact expression for the survival probability valid for arbitrary t can also be obtained. This is an interesting result so we proceed in more detail. For the single-trap problem without evanescence we have [42]

$$Q_1(r, t; R) = 1 - \frac{R}{r} H_{11}^{10} \left[\frac{r - R}{\sqrt{K_\gamma t^\gamma}} \middle| \begin{matrix} (1, \gamma/2) \\ (0, 1) \end{matrix} \right], \quad (27)$$

leading to

$$\left. \frac{\partial Q_1(r,t;R)}{\partial r} \right|_{r=R} = \frac{1}{R} + \frac{1}{\Gamma(1-\gamma/2)} \frac{1}{\sqrt{K_\gamma t^\gamma}}. \quad (28)$$

With this result we get from Eq. (23)

$$\begin{aligned} \sigma^*(t;R) &= \frac{4\pi\sqrt{K_\gamma}}{\rho_0 R \Gamma(\gamma/2)} \int_0^t \rho(t') t'^{\gamma/2-1} dt' \\ &+ \frac{4\pi K_\gamma}{\rho_0 R^2 \Gamma(\gamma)} \int_0^t \rho(t') t'^{\gamma-1} dt'. \end{aligned} \quad (29)$$

Here we have used the fact that the fractional derivative of C , a constant, is not zero but is instead ${}_0D_t^{1-\gamma} C = Ct^{\gamma-1}/\Gamma(\gamma)$.

In contrast with the $d = 1$ and the $d = 3$ cases, no explicit solution in a simple integral form similar to that of Eqs. (25) and (29) is available for $\sigma^*(t;R)$ when $d = 2$. However, we can write explicit expressions for the approach of the survival probability to its final value. From the result for Q_1 given in Ref. [42], one finds

$$\left. \frac{\partial Q_1(r,t;R)}{\partial r} \right|_{r=R} \sim \frac{2}{R} \frac{1}{\gamma \ln(\alpha_\gamma t)}. \quad (30)$$

The fractional derivative of this expression is best computed in Laplace space [42]. Transforming the resulting expression back into the time domain we find that

$${}_0D_t^{1-\gamma} [1/\ln(\alpha_\gamma t)] \sim t^{\gamma-1}/[\Gamma(\gamma) \ln(\alpha_\gamma t)]. \quad (31)$$

Let us further assume that the trap density $\rho(t)$ decays sufficiently rapidly to ensure that $\sigma^*(\infty;R)$ is finite. From Eq. (24) we find that, for $d = 2$,

$$\sigma^*(\infty;R) - \sigma^*(t;R) \sim \frac{4\pi K_\gamma}{\rho_0 R^2 \Gamma(\gamma+1)} \int_t^\infty \frac{\rho(t') t'^{\gamma-1}}{\ln(\alpha_\gamma t')} dt'. \quad (32)$$

We next implement our general results for particular forms of the decay of the trap density.

IV. IMPLEMENTATION FOR PARTICULAR TRAP DECAY FUNCTIONS

A. Exponentially decaying trap density

We first consider an exponentially decaying trap density, $\rho(t) = \rho_0 \exp(-\lambda t)$, where the characteristic time scale of the decay is given by $\tau \equiv \lambda^{-1}$. This would represent a unimolecular decay if this were the only decay channel, which in the presence of the target it is not. However, for a single particle, or for the first of many, this is still the scenario because we are looking at the decay only up to the time that this second channel first affects the trap density. The focus of our interest is in the final value of the survival probability of the target and the long-time approach to it, but for completeness we also give a general expression for the early time behavior.

For $d = 1$ and $d = 3$ we can directly insert the exponential decay of the density in Eqs. (25) and (29) and perform the integrals. Alternatively, we note that, for the exponentially

decaying trap density, Eq. (17) leads to the relation

$$Q_{1,T}^*(r,t;R) = e^{-\lambda t} Q_1(r,t;R) + \lambda \int_0^t e^{-\lambda t'} Q_1(r,t';R) dt'. \quad (33)$$

Next, from Eq. (10) and the Laplace transform of Eq. (33) it then follows that

$$\begin{aligned} u Q_{1,T}^*(r,u;R) \\ = 1 - \left(\frac{r}{R}\right)^{1-(d/2)} \frac{K_{d/2-1}(\sqrt{r^2(u+\lambda)^\gamma/K_\gamma})}{K_{d/2-1}(\sqrt{R^2(u+\lambda)^\gamma/K_\gamma})}. \end{aligned} \quad (34)$$

The Laplace transform of Eq. (19) then immediately leads to

$$\sigma^*(u;R) = \frac{S_d K_\gamma^{1/2}}{u R (u+\lambda)^{\gamma/2}} \frac{K_{d/2}(\sqrt{R^2(u+\lambda)^\gamma/K_\gamma})}{K_{d/2-1}(\sqrt{R^2(u+\lambda)^\gamma/K_\gamma})}. \quad (35)$$

For odd-valued d (but not for even-valued d) the modified Bessel functions of the second kind can be expressed more simply in standard power series expansions. For $d = 1$ we can follow either path (direct integration or simplification and inversion of the modified Bessel function) to obtain

$$\sigma^*(t;R) = \frac{\ell_\gamma}{R} \left(1 - \frac{\Gamma(\gamma/2, \lambda t)}{\Gamma(\gamma/2)}\right), \quad (36)$$

where $\ell_\gamma \equiv (4K_\gamma \tau^\gamma)^{1/2}$ is a characteristic length scale associated with the distance covered by a trap during its mean survival time when no target is present. Equation (36) is equivalent to Eq. (15) in Ref. [46]. For $d = 3$ we can again follow either route to the solution and find

$$\begin{aligned} \sigma^*(t;R) \\ = 2\pi \frac{\ell_\gamma}{R} \left(1 - \frac{\Gamma(\gamma/2, \lambda t)}{\Gamma(\gamma/2)}\right) + \pi \frac{\ell_\gamma^2}{R^2} \left(1 - \frac{\Gamma(\gamma, \lambda t)}{\Gamma(\gamma)}\right), \end{aligned} \quad (37)$$

resulting in a smaller survival probability than in $d = 1$. As noted already, there are no simple, closed-form solutions valid for arbitrary times for $d = 2$, although we are able to extract some limiting behaviors for this case as well (see below).

The computation of the final value of $\sigma^*(t;R)$ in arbitrary integer dimension is readily obtained from Eq. (35) by means of the final value theorem for the Laplace transform:

$$\sigma^*(\infty;R) = \lim_{u \rightarrow 0} u \sigma^*(u;R) = S_d \frac{\ell_\gamma}{2R} \frac{K_{d/2}(2R/\ell_\gamma)}{K_{d/2-1}(2R/\ell_\gamma)}. \quad (38)$$

Before further evaluation, we note as an aside that the above nonzero survival probability implies an infinite mean survival time of the target in any dimension d . In contrast, if the traps do not evanesce, the mean lifetime of the target is finite [44]. For odd dimensions, Eq. (38) can be conveniently rewritten as rational functions of the argument R/ℓ_γ . For example: $\sigma^*(\infty;R) = \ell_\gamma/R$ for $d = 1$ and $\sigma^*(\infty;R) = (2\pi \ell_\gamma/R)[(1 + \ell_\gamma/(2R))]$ for $d = 3$.

Next we explicitly present the results for the approach to the final value by exhibiting the difference $\sigma^*(\infty;R) - \sigma^*(t;R)$ at long times. For $d = 1$ and $d = 3$ this respectively follows from Eqs. (36) and (37), whereas from $d = 2$ the long-time

behavior can be inferred from Eq. (32). We find

$$\sigma^*(\infty; R) - \sigma^*(t; R) \propto \begin{cases} t^{\gamma/2-1} e^{-\lambda t}, & d = 1 \\ \ln^{-1}(\alpha_\gamma t) t^{\gamma-1} e^{-\lambda t}, & d = 2 \\ t^{\gamma-1} e^{-\lambda t}, & d = 3. \end{cases} \quad (39)$$

This asymptotic behavior also holds in the case of normal diffusion ($\gamma = 1$). One can see that the decay of the survival probability to the final state prescribed by Eq. (39) becomes faster as one goes from one to two dimensions and from two to three dimensions. It is also straightforward to show that the long-time behavior prescribed by Eq. (39) for $d = 3$ remains valid for $d > 3$ (the prefactor, however, depends on d).

The limit $\lambda \rightarrow 0$ (no evanescence) turns out to be singular. Indeed, in the absence of evanescence, $\sigma^*(t; R)$ tends to infinity as given by Eq. (11), which is different from the result obtained when taking the limit $\lambda \rightarrow 0$ in Eq. (39). We conclude that the evanescence reaction completely changes the physics of the problem, affecting both the steady state and the decay form of the survival probability.

Finally, the short time behavior ($t \ll \lambda^{-1}$) is straightforward to obtain via a Tauberian theorem applied to the large- u ($u \gg \lambda$) limit of Eq. (35):

$$\sigma^*(u; R) \sim \frac{S_d K_\gamma^{1/2}}{R u^{1+\gamma/2}} \rightarrow \sigma^*(t; R) \sim \frac{S_d K_\gamma^{1/2}}{R \Gamma(1 + \gamma/2)} t^{\gamma/2}. \quad (40)$$

As one might have guessed, the short-time result is independent of λ (i.e., the effect of the evanescence reaction is still negligible in this regime).

B. Power-law decay of trap density

We next turn to the case of a power-law decaying density; that is,

$$\rho(t) = \frac{\rho_0}{(1 + t/\tau)^\beta}, \quad \beta > 0. \quad (41)$$

This choice corresponds to a time dependent rate constant $\lambda(t)$, which can be used to capture the essential features of complex higher-order kinetics by means of the linear differential equation (12) and a proper choice of the exponent β . Interestingly, the survival probability of the target in this case depends not only on dimensionality but also on the relative values of the power-law decay exponent β and the subdiffusion exponent γ .

The behavior when $d = 1$ follows directly from Eq. (25), and can be summarized as follows [46]:

$$\sigma^*(t; R) \sim \begin{cases} \ell_\gamma \frac{\Gamma(\beta-\gamma/2)}{R \Gamma(\beta)}, & \beta > \gamma/2 \\ \frac{\ell_\gamma}{R \Gamma(\gamma/2)} \ln(t/\tau), & \beta = \gamma/2 \\ \frac{\ell_\gamma}{R(\gamma/2-\beta)\Gamma(\gamma/2)} (t/\tau)^{\gamma/2-\beta}, & \beta < \gamma/2, \end{cases} \quad (42)$$

where ℓ_γ is defined in a way similar to the exponential case [i.e., $\ell_\gamma = (4K_\gamma \tau^\gamma)^{1/2}$].

Next we consider the two-dimensional system. Our starting equation is Eq. (23) with $d = 2$:

$$\sigma^*(t; R) = 2\pi \frac{K_\gamma}{R} \int_0^t \left[{}_0D_{r'}^{1-\gamma} \frac{\partial Q_1(r, t'; R)}{\partial r} \Big|_{r=R} \right] \times (1 - t'/\tau)^{-\beta} dt'. \quad (43)$$

The behavior of the integral on the right-hand side depends on the relative values of β and γ . We consider three different cases:

Case 1 ($\beta > \gamma$). From the asymptotic long-time behavior (30) and the expression for the fractional derivative of the inverse logarithm (31) one finds

$${}_0D_{r'}^{1-\gamma} \frac{\partial Q_1(r, t'; R)}{\partial r} \Big|_{r=R} \sim \frac{2t'^{\gamma-1}}{R \Gamma(\gamma + 1) \ln(\alpha_\gamma t')}. \quad (44)$$

Under the assumption that t is large enough, we now split the interval of integration $[0, t]$ into two subintervals, $I_1 = [0, t_c]$ and $I_2 = [t_c, t]$, where t_c is chosen sufficiently large so as to ensure that the approximation (44) holds over the full extent of I_2 . Hence one has

$$\sigma^*(t; R) \sim \mathcal{C} + \frac{4\pi K_\gamma \tau^\beta}{\Gamma(\gamma + 1) R^2} \int_{t_c}^t \frac{t'^{\gamma-\beta-1}}{\ln(\alpha_\gamma t')} dt', \quad (45)$$

where we have used the long-time approximation $\rho(t')/\rho_0 \approx (t'/\tau)^{-\beta}$ and \mathcal{C} represents the integral from 0 to t_c . In this case one can easily check via partial integration that the integral on the right-hand side of Eq. (45) remains finite as $t \rightarrow \infty$. Hence $\sigma^*(\infty; R)$ is finite and the target has a nonzero chance of eternal survival. Using the explicit form of $\rho(t')$ in Eq. (32) we find

$$\sigma^*(\infty; R) - \sigma^*(t; R) \sim \frac{\pi \ell_\gamma^2}{(\beta - \gamma) \Gamma(\gamma + 1) R^2} \frac{(t/\tau)^{\gamma-\beta}}{\ln(\alpha_\gamma t)}. \quad (46)$$

Unfortunately, it does not seem possible to find an explicit exact expression for $\sigma^*(\infty; R)$ due to the lack of an exact expression for $Q_1(r, t'; R)$ valid for the whole time domain.

Case 2 ($\beta = \gamma$). In this marginal case the target also disappears eventually, but the approach to the empty state has a different analytic dependence, as Eq. (45) now leads to

$$\sigma^*(t; R) \sim \frac{\pi \ell_\gamma^2}{\Gamma(\gamma + 1) R^2} \ln(\ln(\alpha_\gamma t)). \quad (47)$$

Thus, the target is eventually killed with certainty, in agreement with the result given in Ref. [47] for the special case $\beta = \gamma = 1$ (normal diffusive traps).

Case 3 ($\beta < \gamma$). Clearly, in this case the constant \mathcal{C} of Eq. (45) becomes negligible at sufficiently long times and the behavior of $\sigma^*(t; R)$ is dominated by the integral on the right-hand side. Using partial integration one easily sees that the leading long-time behavior is given by

$$\sigma^*(t; R) \sim \frac{\pi \ell_\gamma^2}{(\gamma - \beta) \Gamma(\gamma + 1) R^2} \frac{(t/\tau)^{\gamma-\beta}}{\ln(\alpha_\gamma t)}. \quad (48)$$

Hence when $\beta < \gamma$ the target eventually disappears with certainty.

We next discuss the three-dimensional case. In this case, the integrals in Eq. (29) can be carried out exactly, and one finds

$$\sigma^*(t; R) = \frac{2\pi \ell_\gamma}{R \Gamma(\gamma/2)} B_{t/(\tau+t)}(\gamma/2, \beta - \gamma/2) + \frac{\pi \ell_\gamma^2}{R^2 \Gamma(\gamma)} B_{t/(\tau+t)}(\gamma, \beta - \gamma), \quad (49)$$

where

$$B_\alpha(z, w) = \int_0^\alpha dt t^{z-1} (1-t)^{w-1} \quad \text{with} \quad \text{Re}(z) > 0 \quad (50)$$

is the incomplete Beta function [51]. The long-time behavior of $\sigma^*(t; R)$ again depends on the relative values of β and γ . The analysis is carried out along lines similar to those presented in Ref. [46] for $d = 1$. We shall distinguish three different cases.

Case 1 ($\beta > \gamma$). We can rewrite $\sigma^*(t; R)$ as

$$\sigma^*(t; R) = \frac{2\pi\ell_\gamma}{R\Gamma(\gamma/2)} B(\gamma/2, \beta - \gamma/2) I_{t/(\tau+t)}(\gamma/2, \beta - \gamma/2) + \frac{\pi\ell_\gamma^2}{R^2\Gamma(\gamma)} B(\gamma, \beta - \gamma) I_{t/(\tau+t)}(\gamma, \beta - \gamma). \quad (51)$$

Here $B(z, w)$ is the Beta function [where the requirement $\text{Re}(z) > 0$ and $\text{Re}(w) > 0$ places us in the ‘‘Case 1’’ regime], and $I_x(z, w)$ is the regularized incomplete Beta function as defined in Sec. 6.6.2 (p. 263) of Ref. [51]. Using the property 6.6.3 in Ref. [51] we can set $I_x(a, b) = 1 - I_{1-x}(b, a)$. Applying the relation 26.5.5 in Ref. [51], and making use of the relation between the Beta function and the Gamma function, we arrive at the asymptotic result

$$\sigma^*(t; R) \sim \sigma^*(\infty; R) - \frac{\pi\ell_\gamma^2}{R^2} \frac{(t/\tau)^{\gamma-\beta}}{(\beta - \gamma)\Gamma(\gamma)}, \quad (52)$$

with

$$\sigma^*(\infty; R) = \frac{2\pi\ell_\gamma}{R} \frac{\Gamma(\beta - \gamma/2)}{\Gamma(\beta)} + \frac{\pi\ell_\gamma^2}{R^2} \frac{\Gamma(\beta - \gamma)}{\Gamma(\beta)}, \quad (53)$$

leading to a nonzero survival probability $Q_T^*(\infty; R) = \exp[-\rho_0 R^2 \sigma^*(\infty; R)]$.

Case 2 ($\beta = \gamma$). In this case the incomplete Beta function in the term proportional to ℓ_γ^2 in Eq. (49) can be rewritten as a hypergeometric function and consequently for long times this term can be approximated by

$$\frac{\pi\ell_\gamma^2}{R^2\Gamma(\gamma + 1)} \left(\frac{t}{\tau}\right)^\gamma {}_2F_1(\gamma, \gamma, \gamma + 1, -t/\tau) \sim \frac{\pi\ell_\gamma^2}{R^2\Gamma(\gamma)} \ln(t/\tau). \quad (54)$$

On the other hand, the term proportional to ℓ_γ goes to a constant for long times, as can be seen using the same expansion as the one used for the $\beta > \gamma$ case. Hence, the survival probability $Q_T^*(t; R)$ vanishes as $(t/\tau)^{-\pi\rho_0 R\ell_\gamma^2/\Gamma(\gamma)}$; that is, $\sigma^*(t; R) \propto \ln(t/\tau)$.

Case 3 ($\beta < \gamma$). In this case the term proportional to ℓ_γ^2 can easily be seen to behave as $\{\pi\ell_\gamma^2/[R^2(\gamma - \beta)\Gamma(\gamma)]\}(t/\tau)^{\gamma-\beta}$ by performing a straightforward asymptotic analysis of the corresponding integral. On the other hand the ℓ_γ term is negligible compared to the ℓ_γ^2 term. This results in a stretched exponential decay to zero, [i.e., $Q_T^*(t; R) \propto \exp(-Ct^{\gamma-\beta})$ with $C > 0$; that is, $\sigma^*(t; R) \propto t^{\gamma-\beta}$].

Thus, in two and three dimensions the target has a finite probability of surviving forever only for $\beta > \gamma$. For comparison, in the one-dimensional case it was found that the target has a chance of eternal survival only when $\beta > \gamma/2$ [cf. Eq. (42)]. We thus see that the interplay between subdiffusive transport and the evanescence reaction determines, also in dimensions higher than one, whether the target can ultimately survive.

V. SUMMARY AND OUTLOOK

We have presented a particular application of a recently derived fractional reaction-subdiffusion equation; namely, the study of the behavior of the survival probability of an immobile target surrounded by a sea of noninteracting diffusive or subdiffusive point traps subject to an evanescence reaction. The evanescence reaction is assumed to take place independently of the CTRW jumps performed by the traps, as opposed to a recently introduced model where disappearance takes place at the time of each jump [52].

The problem considered in this paper is only one of a family of many possible boundary value problems which may be dealt with using our equations. However, this particular choice may be of interest in a number of experimental situations (e.g., radical recombination kinetics in the presence of added scavenger molecules [33] also responsible for the disappearance of radicals). As far as we know, the interplay between the scavenging reaction and possible memory effects arising in some environments remains unexplored.

We focused on the case of exponential evanescence and power-law evanescence, extending previous results applicable only to the one-dimensional case. In particular, our results also hold for the normal diffusion case ($\gamma = 1$). The presence of the evanescence reaction was found to completely modify the physics of the problem, both at the level of the steady state and the decay of the survival probability to a finite steady state or to zero. More specifically, with an exponentially decaying trap density $\rho(t) = \rho_0 e^{-\lambda t}$ (with $\lambda > 0$), we find that there is a finite survival probability of the target in all dimensions because the traps die sufficiently quickly in their search for the target. By way of contrast, when the traps do not evanesce the target has a zero survival probability in all dimensions. The long-time approach toward the final value of the survival probability turns out to be more complex than in the case of nonevanescing traps, and in the subdiffusive case $\gamma < 1$ it involves powers of t as well as exponential factors $e^{-\lambda t}$ (with a logarithmic correction in $d = 2$). On the other hand, when the density decays as a power law, $\rho(t) \propto t^{-\beta}$ with $\beta > 0$, the behavior depends on the relative values of β and the anomalous diffusion exponent γ of the traps. In one dimension, the target has a finite asymptotic survival probability if $\beta > \gamma/2$, whereas in two and three dimensions the target only has a finite chance of eternal survival when $\beta > \gamma$.

A natural extension of this work would allow normal diffusive or subdiffusive target motion (the case of normal diffusive target and normal diffusive evanescent traps has been considered in Ref. [53]). Note, however, that in such a case the respective distances between the target and the traps would no longer evolve as independent variables, implying that our asymptotically exact approach would not work in its present form. Nonetheless, approximations based on the fact that at long times the dominant contribution to the survival probability comes from the subset of trajectories where the target remains immobile [54,55] could prove useful to tackle the problem. Ultimately, this behavior finds its roots in what has been termed the ‘‘Pascal principle’’ in the literature [56–58]; namely, a target placed in a symmetric initial distribution of traps survives longer on average if it stays still rather than if it moves.

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