# Reaction-subdiffusion and reaction-superdiffusion equations for evanescent particles performing continuous-time random walks

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Starting from a continuous-time random-walk (CTRW) model of particles that may evanesce as they walk, our goal is to arrive at macroscopic integrodifferential equations for the probability density for a particle to be found at point $\mathbf{r}$ at time $t$ given that it started its walk from $\mathbf{r}_0$ at time $t=0$. The passage from the CTRW to an integrodifferential equation is well understood when the particles are not evanescent. Depending on the distribution of stepping times and distances, one arrives at standard macroscopic equations that may be "normal" (diffusion) or "anomalous" (subdiffusion and/or superdiffusion). The macroscopic description becomes considerably more complicated and not particularly intuitive if the particles can die during their walk. While such equations have been derived for specific cases, e.g., for location-independent exponential evanescence, we present a more general derivation valid under less stringent constraints than those found in the current literature.

## I. INTRODUCTION

Continuous time random walks (CTRWs) offer a sweeping framework to describe the dynamics of particles whose motion may be "anomalous." That is, in addition to providing a way to study the motion of diffusive particles at a more microscopic level than, say, the more macroscopic diffusion equation, CTRWs can also be used to describe particles whose motion is subdiffusive or superdiffusive. The connection between CTRWs and the associated "more macroscopic" description, which is in general an integrodifferential equation rather than a (partial) differential equation as in the case of ordinary diffusion, has been firmly established. The description in terms of integrodifferential equations is often attractive because all the machinery developed to solve the equations subject to a number of different boundary conditions can be brought to bear. It should be noted that the starting point for problems that involve subdiffusive or superdiffusive particles is often simply the integrodifferential equation itself. However, one must exercise caution in simply starting with such a macroscopic description. It should also be noted that these integrodifferential equations can arise from microscopic models other than a CTRW, with the latter not always being the appropriate framework. An example is the motion of particles in random landscapes of various sorts such as, for instance, one in which each site is associated with a potential well whose depth is chosen from a random distribution. The walk in a landscape of potential wells of random depths may be normal in the sense that the escape from each well follows a Kramers law, but the irregularity of the landscape may give rise to trapping events that may slow down the progress of the particles to the point of giving rise to subdiffusion.

The situation becomes much more complicated when the moving particles also undergo reactions [1,2]. This applies to situations such as reversible or irreversible conversion to a different species (e.g., $A \rightarrow B$ [3–5] or $A \rightleftharpoons B$ [5,6]), reactions giving rise to propagating fronts (say, $A+B \rightarrow C$ [7] and $A+B \rightarrow 2A$ [8]), binary reactions (e.g., $A+A \rightarrow 0$), or even spontaneous evanescence ($A \rightarrow 0$). Focusing on the latter case, the description of the evolution of such evanescent particles at the macroscopic level of a subdiffusion or superdiffusion equation, and the investigation of the proper way to include the evanescence or reaction in such equations, is a matter of continuing study and is usually carried out in the context of rather specific models [3,9].

In this paper we pursue this goal somewhat more generically: we consider the deduction of subdiffusion or superdiffusion equations when the moving particles are evanescent. The death rate of the particles may in general depend on location, time, how long a particle has spent at a particular location, or the death sentence may even be imposed by an external agent—one can think of a large variety of death scenarios. Our presentation is based on a CTRW formulation generalized to include particle evanescence. One step of the discussion concerns the way in which evanescence can be built into a CTRW model, a choice which is not unique. The second step is then to go from a CTRW model to a subdiffusion or superdiffusion equation as appropriate.

In Sec. II we start with some basic CTRW quantities and relations, which we then use in Sec. III to construct an integral equation for the probability density of finding a particle at a certain position at time $t$ given that it began its motion at a different position at time $t=0$. This integral equation includes the possibility that the particle evanesces along the way, and is the starting point for the further derivation of subdiffusive and superdiffusive equations. In Sec. IV we explicitly construct our fractional equations for the case of subdiffusion, superdiffusion, and a mixture of the two. We conclude with a summary and some thoughts for the future in Sec. V.

## II. STARTING WITH A CTRW

Our goal is to arrive at integrodifferential equations for the probability density $w(\mathbf{r},t|\mathbf{r}_0,0)$ for a particle that started

its journey at point $\mathbf{r}_0$ at time $t=0$ to be at $\mathbf{r}$ at time $t$. No matter how we arrive at such an equation, the description of the motion of the particles as a CTRW requires the introduction of the probability density $\Psi(\mathbf{r}-\mathbf{r}',t-t')$ that a random walker jumps in a single step from $\mathbf{r}'$ to $\mathbf{r}$ after waiting a time interval $t-t'$ at the position $\mathbf{r}'$. The dependence on only the difference of the two position vectors reflects an assumption of spatial homogeneity for the jumping mechanism. Associated with this probability density is the usual waiting time probability density,

$$\psi(t) = \int d\mathbf{r}' \Psi(\mathbf{r}',t), \tag{1}$$

and also the probability density for a single-step displacement,

$$\chi(\mathbf{r}) = \int_0^\infty dt' \Psi(\mathbf{r},t'). \tag{2}$$

The "normal" or "anomalous" (subdiffusive or superdiffusive) character of the process depends on the forms of the probability densities, which we will specify later.

Since we deal with evanescent particles, we also introduce the probability $\Xi(\mathbf{r},\mathbf{r}';t,t')$ that the particle has not died spontaneously during the stepping process described by $\Psi(\mathbf{r}-\mathbf{r}',t-t')$. Also, finding the particle at $\mathbf{r}$ at time $t$ does not mean that it jumped onto that location exactly at that time. In fact, it might have jumped there at an earlier time and then just waited there without dying, or it might have jumped there at an earlier time, returned any number of times before $t$, and then waited there. As a starting point in our route to an integral equation for $w(\mathbf{r},t|\mathbf{r}_0,0)$, it is thus appropriate to introduce $q_n(\mathbf{r},t|\mathbf{r}_0,0)$, the probability density that a particle arrive at the position $\mathbf{r}$ at time $t$ exactly after making its $n$th jump, given that it started its walk at time $t=0$ at position $\mathbf{r}_0$ [9]. This initial condition is described by the equation

$$q_0(\mathbf{r},t|\mathbf{r}_0,0) = \delta(\mathbf{r}-\mathbf{r}_0)\delta(t). \tag{3}$$

From this follows the definition

$$q(\mathbf{r},t|\mathbf{r}_0,0) \equiv \sum_{n=0}^\infty q_n(\mathbf{r},t|\mathbf{r}_0,0), \tag{4}$$

which is the probability density that the particle steps onto position $\mathbf{r}$ exactly at time $t$ (regardless of how many times the particle has stepped anywhere, including on $\mathbf{r}$, before this time).

It is straightforward to write integral equations for the probability densities $q$ and $q_n$. For $q_n$ no further discussion beyond the definitions is necessary to write the integral recurrence equation [10]

$$q_{n+1}(\mathbf{r},t|\mathbf{r}_0,0) = \int d\mathbf{r}' \int_0^t dt' \Psi(\mathbf{r}-\mathbf{r}',t-t')$$
$$\times q_n(\mathbf{r}',t'|\mathbf{r}_0,0)\Xi(\mathbf{r},\mathbf{r}';t,t'). \tag{5}$$

In words, this equation states that the probability density for the particle to arrive at location $\mathbf{r}$ at time $t$ on the $(n+1)$st

step is equal to the probability that the particle arrived at any point $\mathbf{r}'$ (hence the integral over $\mathbf{r}'$) at any earlier time $t'$ (hence the integral over $t'$) on the $n$th step and then steps from $\mathbf{r}'$ to $\mathbf{r}$ at time $t$, provided that the particle does not die in the interval between these two steps. The usual integral equation without evanescence is immediately recovered upon setting $\Xi$ equal to unity. Summing this equation over step number $n$, the integral equation

$$q(\mathbf{r},t|\mathbf{r}_0,0) = \int d\mathbf{r}' \int_0^t dt' \Psi(\mathbf{r}-\mathbf{r}',t-t')$$
$$\times q(\mathbf{r}',t'|\mathbf{r}_0,0)\Xi(\mathbf{r},\mathbf{r}';t,t') + q_0(\mathbf{r},t|\mathbf{r}_0,0) \tag{6}$$

immediately follows.

The more complex question is now how to go from these integral equations to one for the desired probability density $w(\mathbf{r},t|\mathbf{r}_0,0)$ and thence to a fractional diffusion equation. This is our pursuit in Sec. III.

## III. INTEGRAL EQUATION WITH EVANESCENCE

Before obtaining an integral equation from the CTRW setup of the last section, we note that one could choose to write such an equation directly, for example,

$$w(\mathbf{r},t|\mathbf{r}_0,0) = \int d\mathbf{r}' \int_0^t dt' \Upsilon(\mathbf{r},\mathbf{r}';t,t')w(\mathbf{r}',t'|\mathbf{r}_0,0)$$
$$\times \Xi(\mathbf{r},\mathbf{r}';t,t') + \Phi(t)\Xi(\mathbf{r},\mathbf{r};t,0)\delta(\mathbf{r}-\mathbf{r}_0), \tag{7}$$

where we have introduced,

$$\Phi(t) = 1 - \int_0^t dt' \psi(t') = \int_t^\infty dt' \psi(t'), \tag{8}$$

the probability that the particle does not take a step in the entire time interval up to time $t$. Note that we have made a point of using $\Xi$ rather than $\Xi$ for the function indicating that no death occurs, because in general there is no reason to expect these two to be the same. The function $\Xi$ that appears in the integral equations obtained in the previous section is the probability that the particle does not die in a time interval exactly delineated by two steps (and none in between), one taking it to location $\mathbf{r}'$ and the other to location $\mathbf{r}$. On the other hand, $\Xi$ is the probability that the particle does not die in a time interval $t-t'$ as it moves from $\mathbf{r}'$ to $\mathbf{r}$, with no reference to steps. There is no *a priori* reason for these two probabilities to be equal. If particles die at a constant rate independent of position and of when steps take place, then these two probabilities would be equal. However, for example if particles die only when they take a step, or, conversely, if particles are more likely to die if they remain at one location, then these probabilities would not be equal. Also, we have denoted the kernel of the integral equation by the new symbol $\Upsilon$ because *a priori* we do not know its connection to $\Psi$. One might attempt reasonable guesses, but that is all so far and in fact somewhat risky.

Our goal is to obtain an equation such as Eq. (7) starting from a CTRW (rather than just writing it down). We therefore begin with Eq. (6) together with the exact relation

$$w(\mathbf{r},t|\mathbf{r}_0,0) = \int_0^t dt' \Phi(t-t') q(\mathbf{r},t'|\mathbf{r}_0,0) \Xi(\mathbf{r},\mathbf{r};t,t'). \quad (9)$$

This exact relation simply says that the probability density for the particle to be at $\mathbf{r}$ at time $t$ is the probability that it stepped onto that location at time $t' < t$ and then neither moved nor died until time $t$. Earlier arrivals and returns are implicitly included in this relation.

In order to proceed toward an integral equation of form (7) with known kernel we find ourselves having to make two admittedly debatable assumptions. One is that the functions $\Xi$ and $\bar{\Xi}$ are equal. The other is that this function can be written as a ratio of the form

$$\Xi(\mathbf{r},\mathbf{r}';t,t') = \frac{\varphi(\mathbf{r},t)}{\varphi(\mathbf{r}',t')}. \quad (10)$$

The special case of location-independent exponential decay $\varphi(\mathbf{r},t) = \exp(-kt)$ (that is, the case of evanescence at a constant rate, mentioned earlier), is that of Sokolov *et al.* [3], and in this case $\Xi(\mathbf{r},\mathbf{r}';t,t') = \exp[-k(t-t')]$. This is the only choice for which $\Xi$ depends on time only through the difference of its time arguments. While Eq. (10) is limiting, it is more general than the specific cases that have been treated in the literature. We defer a discussion of some examples that fit this constraint and the associated physical implications to the next section.

To make use of special form (10) we start by making the replacements $t \to t'$ and $t' \to t''$ in Eq. (6). We then multiply by $\Phi(t-t')\Xi(\mathbf{r},\mathbf{r};t,t')$, integrate over $t'$, and use relation (9) to write

$$w(\mathbf{r},t|\mathbf{r}_0,0) = \int_0^t dt' \Phi(t-t') \Xi(\mathbf{r},\mathbf{r};t,t') \left( \int d\mathbf{r}' \int_0^{t'} dt'' \Psi(\mathbf{r}-\mathbf{r}',t'-t'') q(\mathbf{r}',t''|\mathbf{r}_0,0) \Xi(\mathbf{r},\mathbf{r}';t',t'') \right)$$

$$+ \int_0^t dt' \Phi(t-t') \Xi(\mathbf{r},\mathbf{r};t,t') q_0(\mathbf{r},t'|\mathbf{r}_0,0). \quad (11)$$

This equation can be manipulated through a number of simple steps. First, we insert the special form [Eq. (10)] on the right side and divide both sides of the equation by $\varphi(\mathbf{r},t)$. This leaves us with the equation

$$\frac{w(\mathbf{r},t|\mathbf{r}_0,0)}{\varphi(\mathbf{r},t)} = \int d\mathbf{r}' \int_0^t dt' \Phi(t-t') \left( \int_0^{t'} dt'' \Psi(\mathbf{r}-\mathbf{r}',t'-t'') \frac{q(\mathbf{r}',t''|\mathbf{r}_0,0)}{\varphi(\mathbf{r}',t'')} \right) + \int_0^t dt' \Phi(t-t') \frac{q_0(\mathbf{r},t'|\mathbf{r}_0,0)}{\varphi(\mathbf{r},t')}. \quad (12)$$

Next, we recognize that the first term on the right is a double convolution with respect to time. The memory kernels can then be exchanged, as can be verified by taking Laplace transforms. Multiplying the rearranged equation by $\varphi(\mathbf{r},t)$ it is then easy to see that

$$w(\mathbf{r},t|\mathbf{r}_0,0) = \int d\mathbf{r}' \int_0^t dt' \Psi(\mathbf{r}-\mathbf{r}',t-t') \frac{\varphi(\mathbf{r},t)}{\varphi(\mathbf{r}',t')} \left( \int_0^{t'} dt'' \Phi(t'-t'') q(\mathbf{r}',t''|\mathbf{r}_0,0) \frac{\varphi(\mathbf{r}',t')}{\varphi(\mathbf{r}',t'')} \right)$$

$$+ \int_0^t dt' \Phi(t-t') q_0(\mathbf{r},t'|\mathbf{r}_0,0) \frac{\varphi(\mathbf{r},t)}{\varphi(\mathbf{r},t')}. \quad (13)$$

Finally, using Eq. (9) and initial condition (3), and again recalling special form (10), we arrive at the desired integral equation

$$w(\mathbf{r},t|\mathbf{r}_0,0) = \int d\mathbf{r}' \int_0^t dt' \Psi(\mathbf{r}-\mathbf{r}',t-t') w(\mathbf{r}',t'|\mathbf{r}_0,0) \frac{\varphi(\mathbf{r},t)}{\varphi(\mathbf{r}',t')} + \Phi(t) \frac{\varphi(\mathbf{r},t)}{\varphi(\mathbf{r},0)} \delta(\mathbf{r}-\mathbf{r}_0). \quad (14)$$

Equation (14) is the starting point for the derivation of various fractional diffusion equations for different forms of the single-step probability density $\Psi(\mathbf{r}-\mathbf{r}',t-t')$ of the underlying CTRW. It is therefore a centerpiece of this work. Note that special form (10) has led to a kernel in the integral equation that is precisely this single-step probability density even though the times $t$ and $t'$ are not necessarily associated with jumping times. Equation (14) has a clear physical interpretation: it considers all possible

ways for a particle to be at point $\mathbf{r}$ at time $t$ by looking at the positions $\mathbf{r}'$ at prior times $t'$ and then tracking their subsequent arrival at the desired point. If the point under consideration is the initial position, the equation also tracks the possibility that the particle has not moved by time $t$. In the language of Hughes [14] (Sec. 3.2.8), Eq. (14) corresponds to a "partition over the last step." The equation counts only those particles that do not evanesce in the process.

Finally, we shall implement one additional simplifying assumption widely adopted in the literature, namely, that the waiting time and jump displacement distributions are mutually independent, so we can write

$$\Psi(\mathbf{r},t) = \psi(t)\chi(\mathbf{r}). \tag{15}$$

Different fractional diffusion equations then arise depending on the behaviors of the tails of these distributions. We proceed to present various cases in the next section. Our derivations closely follow known results presented in a number of helpful review sources such as the reports of Metzler and Klafter [11,12] and a recent multiauthored compendium on anomalous processes [13]. Our main purpose here is to add evanescence to the mix and to determine how the evanescence "reaction" enters these equations. In particular, when $\psi(t)$ has long tails and $\chi(\mathbf{r})$ does not, we will arrive at a fractional subdiffusion equation. When, on the other hand, $\chi(\mathbf{r})$ has long tails but $\psi(t)$ does not, we arrive at a superdiffusive equation. The most "anomalous" case occurs when both have long tails, which leads to a bifractional equation.

We end this section with a practical consideration. Instead of working with the probability density of interest, $w(\mathbf{r},t|\mathbf{r}_0,0)$, it turns out to be more convenient to work with a ratio introduced earlier,

$$\eta(\mathbf{r},t|\mathbf{r}_0,0) = \frac{w(\mathbf{r},t|\mathbf{r}_0,0)}{\varphi(\mathbf{r},t)}. \tag{16}$$

Dividing Eq. (14) by $\varphi(\mathbf{r},t)$ and taking the Fourier (for space)-Laplace (for time) transform, we find

$$\hat{\tilde{\eta}}(\mathbf{q},u) = \hat{\chi}(\mathbf{q})\tilde{\psi}(u)\hat{\tilde{\eta}}(\mathbf{q},u) + \tilde{\Phi}(u)\frac{e^{i\mathbf{q}\cdot\mathbf{r}_0}}{\varphi(\mathbf{r}_0,0)}. \tag{17}$$

Together with the relation

$$\tilde{\Phi}(u) = \frac{1}{u} - \frac{\tilde{\psi}(u)}{u} \tag{18}$$

and the convolution theorems for both Fourier and Laplace transforms, we arrive at the Fourier-Laplace transformed reaction-diffusion equation with evanescence, equivalent to Eq. (14) when the memory kernel can be factorized as in Eq. (15),

$$u\hat{\tilde{\eta}}(\mathbf{q},u) = u\hat{\chi}(\mathbf{q})\tilde{\psi}(u)\hat{\tilde{\eta}}(\mathbf{q},u) + (1 - \tilde{\psi}(u))\frac{e^{i\mathbf{q}\cdot\mathbf{r}_0}}{\varphi(\mathbf{r}_0,0)}. \tag{19}$$

## IV. FRACTIONAL DIFFUSION EQUATIONS WITH EVANESCENCE

In this section we proceed to deduce the fractional diffusion equations appropriate for long-tailed waiting time distributions, for long-tailed jump distance distributions, and for both simultaneously.

### A. Fractional subdiffusive equation with evanescence

Subdiffusion is characterized by a waiting time distribution with a tail so long that it lacks integer moments, that is,

$$\psi(t) \sim \frac{\kappa}{\tau_D}\left(\frac{t}{\tau_D}\right)^{-\gamma-1}, \tag{20}$$

with $0 < \gamma < 1$. Here $\kappa$ is a dimensionless constant and $\tau_D$ is a characteristic mesoscopic time (but not a first moment). The small-$u$ behavior of the Laplace transform of the waiting time distribution reads as

$$\tilde{\psi}(u) \sim 1 - \mathcal{A}u^\gamma, \tag{21}$$

with $\mathcal{A} = \gamma^{-1}\kappa\Gamma(1-\gamma)\tau_D^\gamma$.

We take the jump distance distribution to be "normal"; that is, it has finite moments. In this case, we are interested in small values of $q = |\mathbf{q}|$, for which one can expand the Fourier transform of the jump displacement distribution and retain only the first two terms,

$$\hat{\chi}(\mathbf{q}) \sim 1 - \frac{\sigma^2\mathbf{q}^2}{2} + \mathcal{O}(\mathbf{q}^4), \tag{22}$$

where the second moment,

$$\sigma^2 = \int d\mathbf{r}\,r^2\chi(\mathbf{r}), \tag{23}$$

is assumed to be finite.

Substitution of expansions (21) and (22) into Eq. (19) and neglect of a term of $\mathcal{O}(u\mathbf{q}^2)$ (which is unimportant in the asymptotic regime of small wave vectors and low frequencies) leaves us with

$$u\hat{\tilde{\eta}}(\mathbf{q},u) - \frac{e^{i\mathbf{q}\cdot\mathbf{r}_0}}{\varphi(\mathbf{r}_0,0)} = -u^{1-\gamma}\frac{\sigma^2\mathbf{q}^2}{2\mathcal{A}}\hat{\tilde{\eta}}(\mathbf{q},u). \tag{24}$$

Laplace and Fourier inversions then yield

$$\frac{\partial\eta(\mathbf{r},t|\mathbf{r}_0,0)}{\partial t} = K_\gamma\,_0\mathcal{D}_t^{1-\gamma}\nabla_\mathbf{r}^2\eta(\mathbf{r},t|\mathbf{r}_0,0), \tag{25}$$

where we have introduced the anomalous diffusion coefficient

$$K_\gamma = \frac{\sigma^2}{2\mathcal{A}}. \tag{26}$$

The integrodifferential operator $_0\mathcal{D}_t^{1-\gamma}$ acting on $y(t)$ is defined as the inverse Laplace transform of $u^{1-\gamma}\tilde{y}(u)$,

$$\mathcal{L}_{u\to t}^{-1}\{u^{1-\gamma}\tilde{y}(u)\} = \,_0\mathcal{D}_t^{1-\gamma}y(t), \tag{27}$$

and is closely related to the Riemann-Liouville operator $_0D_t^{1-\gamma}$ defined by

$$_0D_t^{1-\gamma}f(\mathbf{r},t) = \frac{1}{\Gamma(\gamma)}\frac{\partial}{\partial t}\int_0^t dt'\frac{f(\mathbf{r},t')}{(t-t')^{1-\gamma}}. \tag{28}$$

In fact, both operators $_0\mathcal{D}_t^{1-\gamma}$ and $_0D_t^{1-\gamma}$ are the same when applied to sufficiently regular functions $f(t)$ as determined by the condition $\lim_{t\to0}\int_0^t d\tau(t-\tau)^{\gamma-1}f(\tau)=0$ (see pages 384 and 118 in [13]). This condition is satisfied for all situations of interest here.

In terms of the probability density $w$, Eq. (25) explicitly yields the fractional reaction-subdiffusion equation

$$\frac{\partial w(\mathbf{r},t|\mathbf{r}_0,0)}{\partial t} = \varphi(\mathbf{r},t)K_{\gamma\ 0}\mathcal{D}_t^{1-\gamma}\nabla_{\mathbf{r}}^2\frac{1}{\varphi(\mathbf{r},t)}w(\mathbf{r},t|\mathbf{r}_0,0)$$

$$+ \frac{\dot{\varphi}(\mathbf{r},t)}{\varphi(\mathbf{r},t)}w(\mathbf{r},t|\mathbf{r}_0,0). \tag{29}$$

Equation (29) is the first mesoscopic highlight of our paper in that all further results for subdiffusion are obtained as special cases. It is perhaps the most general fractional subdiffusion equation associated with a CTRW with evanescence for a single-species system obtained to date. It is not as general as one might hope because of the rather stringent condition [Eq. (10)], but it does include a number of cases treated in recent work. Notably, the results of Sokolov *et al.* [3] and the explicit results in Henry *et al.* [9] are recovered if we set $\varphi(\mathbf{r},t) \propto \exp(-kt)$. More complex cases will be discussed in Sec. IV B.

As a final note in this subsection, we mention that under the same conditions that led to our subdiffusive fractional Eq. (29) with a Riemann-Liouville operator, we can arrive at an equivalent fractional subdiffusion equation of Caputo form. It turns out to be

$$\frac{\partial^\gamma}{\partial t^\gamma}\frac{w(\mathbf{r},t|\mathbf{r}_0,0)}{\varphi(\mathbf{r},t)} = K_\gamma\nabla_{\mathbf{r}}^2\frac{w(\mathbf{r},t|\mathbf{r}_0,0)}{\varphi(\mathbf{r},t)}. \tag{30}$$

This is in general not our preferred choice because of the difficulties in carrying out the Caputo fractional derivative of a product.

### B. Beyond exponential evanescence

We next proceed to discuss the connection between Eq. (29) and the recent work of Fedotov [15], which in turn recovers results in [9]. In the language of Fedotov translated to our work, his choice corresponds to the particular selection (in one dimension)

$$\varphi(x,t) = \exp\left\{\int_\tau^t \Re[\rho(x,t')]dt'\right\}. \tag{31}$$

Here in Fedotov's language the chemical reaction responsible for the evanescence is assumed to follow the law of mass action so that the reaction term is of the form $\mathcal{R}(\rho)\rho$, with the rate coefficient thus defined as

$$\mathcal{R}[\rho(x,t)] = \left[\frac{\dot{\rho}}{\rho}\right]_{\text{Reaction}}. \tag{32}$$

In turn, $\rho(x,t)$ is the density of (surviving) particles at point $x$ at time $t$, which in a one-particle system is related to our probability density via an integration over the distribution $p(x_0)$ of all possible initial positions,

$$\rho(x,t) = \int dx_0 p(x_0)w(x,t;x_0,0). \tag{33}$$

The time $\tau$ in Eq. (31) is an arbitrary reference time that can be chosen to be zero. Specifically, the choice $\tau=0$, substitution of Eq. (31) into our general Eq. (29), and subsequent integration over the initial condition leads exactly to Eq. (23)

in Ref. [15], which is thus again a special case of our more general formalism. The result is especially noteworthy because the probability of spontaneous death is not set *a priori* but depends on the changing density itself. This leads to an interesting complex nonlinear problem.

A less demanding version of the above problem arises if the reaction rate depends *only* on the mean global density

$$\bar{\rho}(t) = \frac{1}{V}\int d\mathbf{r}\rho(\mathbf{r},t), \tag{34}$$

where $V$ is the system volume. The corresponding equations for the homogeneous rate coefficient and the associated $\varphi$-function are, respectively,

$$\Re(\bar{\rho}(t)) = \frac{1}{\bar{\rho}}\frac{d\bar{\rho}}{dt}, \tag{35}$$

and (taking $\tau=0$ in one dimension)

$$\varphi(x,t) = \varphi(t) = \exp\left\{\int_0^t \Re(\bar{\rho}(t'))dt'\right\} = \frac{\bar{\rho}(t)}{\bar{\rho}(0)}. \tag{36}$$

Now the physical meaning of $\varphi(t)$ in this case becomes clear: in a many-particle system it is simply the fraction of surviving particles in the system. Then, according to Eq. (10), the probability $\Xi(t,t')$ that a randomly chosen evanescent particle present at time $t'$ in the system survives up to time $t$ is $\bar{\rho}(t)/\bar{\rho}(t')$. This is the correct result that one would expect from the interpretation of $\Xi(t,t')$ as a conditional probability, namely, the survival probability $\bar{\rho}(t)/\bar{\rho}(0)$ of a given particle up to time $t$ must be equal to the probability $\Xi(t,t')$ that it does not die during the interval $(t',t)$ provided that it survived up to time $t'$, multiplied by the probability $\bar{\rho}(t')/\bar{\rho}(0)$ for this condition to be fulfilled. Using Eq. (36) in Eq. (29), the equation for the kinetics of individual particles subject to a density-dependent homogeneous rate coefficient then follows,

$$\frac{\partial w(\mathbf{r},t|\mathbf{r}_0,0)}{\partial t} = \bar{\rho}(t)K_{\gamma\ 0}\mathcal{D}_t^{1-\gamma}\nabla_{\mathbf{r}}^2\frac{1}{\bar{\rho}(t)}w(\mathbf{r},t|\mathbf{r}_0,0)$$

$$+ \frac{\dot{\bar{\rho}}(t)}{\bar{\rho}(t)}w(\mathbf{r},t|\mathbf{r}_0,0). \tag{37}$$

Going even further, note that Eq. (29) is not restricted to the case of a density-dependent rate coefficient. It goes further (and is a key difference from existing work) in allowing for the possibility of an *external* control mechanism of the evanescence rate coefficient not necessarily coupled to the instantaneous particle population. Indeed, one can prescribe a reactivity field $\Re(\mathbf{r},t) < 0$ and subsequently set $\varphi(\mathbf{r},t) = \exp\{\int_0^t \Re(\mathbf{r},t')dt'\}$ which is then the probability that the particle does not vanish up to time $t$ given that it stays at $\mathbf{r}$ all the time (immobile reactant limit). Consequently, our assumption (10) implies that the probability for a particle to vanish during the time interval $(t',t)$ if it is known to be at position $\mathbf{r}'$ at time $t'$ and at position $\mathbf{r}$ at time $t$ depends only on the instantaneous values of the local rate coefficient at the start and end points via the ratio of the associated survival probabilities for particles, respectively, fixed at $\mathbf{r}'$ and $\mathbf{r}$.

However, the probability that the particle evanesces during this time interval does not depend on the particular trajectory followed by the particle between these two points.

### C. Fractional superdiffusive equation with evanescence

In Sec. IV A we dealt with a waiting time distribution with long tails together with a "normal" distribution of displacements, and the end result was a fractional subdiffusion equation. In this subsection we consider a "normal" waiting time distribution along with a distribution of displacements that has long tails. This will lead to a superdiffusive equation.

We thus consider a waiting time distribution with a finite mean waiting time $\tau$ between steps so that its Laplace transform at small argument (corresponding to long times) behaves as

$$\tilde{\psi}(u) \sim 1 - \tau u. \tag{38}$$

For the jump length distribution we assume an inverse power-law behavior,

$$\chi(\mathbf{r}) \sim \frac{\sigma^{\mu}}{r^{1+\mu}} \tag{39}$$

with $1 \leq \mu \leq 2$ and $r = |\mathbf{r}|$. Its variance diverges, and its Fourier transform is

$$\hat{\chi}(\mathbf{q}) \sim 1 - \sigma^{\mu} q^{\mu}, \tag{40}$$

where $q = |\mathbf{q}|$. The steps to follow are now straightforward. Again, it turns out to be convenient to work with Eq. (17). Substitution of the above expansions into this equation, retention of leading terms, and some simple algebra leads to

$$u \hat{\tilde{\eta}}(\mathbf{q}, u) - \frac{e^{i\mathbf{q}\cdot\mathbf{r}_0}}{\varphi(\mathbf{r}_0, 0)} = -\frac{\sigma^{\mu}}{\tau} q^{\mu} \hat{\tilde{\eta}}(\mathbf{q}, u), \tag{41}$$

or, inverting the time Laplace transform,

$$\frac{\partial}{\partial t} \hat{\eta}(\mathbf{q}, t) = -\frac{\sigma^{\mu}}{\tau} q^{\mu} \hat{\eta}(\mathbf{q}, t). \tag{42}$$

Let $\partial^{\mu}/\partial r^{\mu}$ be the operator defined by the following Fourier transform property,

$$\mathfrak{F}\left\{ \frac{\partial^{\mu} f(\mathbf{r})}{\partial r^{\mu}} \right\} = -q^{\mu} f(\mathbf{q}). \tag{43}$$

We can thus write

$$\frac{\partial \eta(\mathbf{r}, t|\mathbf{r}_0, 0)}{\partial t} = K \frac{\partial^{\mu}}{\partial r^{\mu}} \eta(\mathbf{r}, t|\mathbf{r}_0, 0), \tag{44}$$

where we have introduced the anomalous diffusion coefficient

$$K = \frac{\sigma^{\mu}}{\tau} \tag{45}$$

For a one-dimensional system $\partial^{\mu}/\partial r^{\mu} = \partial^{\mu}/\partial x^{\mu}$ is the Riesz operator [11–13].

Undoing variable transformation (16), we finally arrive at the fractional reaction-superdiffusion equation,

$$\frac{\partial w(\mathbf{r}, t|\mathbf{r}_0, 0)}{\partial t} = \varphi(\mathbf{r}, t) K \frac{\partial^{\mu}}{\partial r^{\mu}} \frac{1}{\varphi(\mathbf{r}, t)} w(\mathbf{r}, t|\mathbf{r}_0, 0)$$

$$+ \frac{\dot{\varphi}(\mathbf{r}, t)}{\varphi(\mathbf{r}, t)} w(\mathbf{r}, t|\mathbf{r}_0, 0). \tag{46}$$

This is the second important mesoscopic result of our paper, namely, the derivation of a reaction-superdiffusion fractional equation starting from a CTRW.

### D. Bifractional equation with evanescence

Finally, in this subsection we combine subdiffusion and superdiffusion in that we choose a waiting time distribution that lacks finite moments (and thus leads to subdiffusion by itself) with a jump distance distribution that also lacks moments (and thus leads to superdiffusion by itself). Our methodology directly lends itself to this combination.

We choose the waiting time distribution of Eq. (20) whose Laplace transform is given in Eq. (21), and the jump distribution Eq. (39) whose Fourier transform is given in Eq. (40). The steps to follow are now essentially the same as in the previous sections, with appropriate care given to the retention of the leading contributions. After some algebra we find

$$\frac{\partial \eta(\mathbf{r}, t|\mathbf{r}_0, 0)}{\partial t} = K \, _0\mathcal{D}_t^{1-\gamma} \frac{\partial^{\mu}}{\partial r^{\mu}} \eta(\mathbf{r}, t|\mathbf{r}_0, 0). \tag{47}$$

Undoing variable transformation (16), we finally obtain the fractional reaction-sub/superdiffusion equation,

$$\frac{\partial w(\mathbf{r}, t|\mathbf{r}_0, 0)}{\partial t} = \varphi(\mathbf{r}, t) K \, _0\mathcal{D}_t^{1-\gamma} \frac{\partial^{\mu}}{\partial r^{\mu}} \frac{1}{\varphi(\mathbf{r}, t)} w(\mathbf{r}, t|\mathbf{r}_0, 0)$$

$$+ \frac{\dot{\varphi}(\mathbf{r}, t)}{\varphi(\mathbf{r}, t)} w(\mathbf{r}, t|\mathbf{r}_0, 0). \tag{48}$$

This is our third mesoscopic result and is unique in that it combines both subdiffusion and superdiffusion in a single equation.

### V. SUMMARY AND OUTLOOK

In this paper we have approached the problem of describing the evolution equation of particles that move in a medium in which they can also die as they move. The model is based on a CTRW description of the motion of the particles. The motion may be anomalous (subdiffusive or superdiffusive) and the particles may die at a rate that can depend on position as well as time. We are able to capture the models that have been explicitly considered in the literature, e.g., the space-independent exponential evanescence model of Refs. [3,9], but our model can also capture complicated position dependences of the evanescent behavior such as that of the model of Fedotov [15] that render the problem nonlinear. We confirm in a more general way than had been established previously the known result that the interplay of the (anomalous) motion and the evanescence is quite complex and that in general it cannot be represented as the sum of two processes the way it can in normal reaction-diffusion scenarios. Having said this, we note as an aside that in certain cases

(e.g., that of evanescence at a constant rate $r=k$) it is possible to reduce the reaction-subdiffusion problem to a pure subdiffusion problem by a proper transformation [15], much in the spirit of Danckwerts' solution for the problem of classical diffusion with a linear reaction [16].

We started by constructing an integral equation for the probability density of finding a (surviving) particle at a location $\mathbf{r}$ at time $t$ given that it stepped on location $\mathbf{r}_0$ at time $t=0$. To proceed from this CTRW-based equation to the fractional equations, we found it necessary to make some specific assumptions about the form of the rate of evanescence. The kernel of the integral equation under these conditions is simply related to the single-step jump probability density of the underlying CTRW. In spite of the constraints, our models include as special cases many of the explicit models that have been presented in the literature [3,9,15].

Once we have arrived at an integral equation, the derivation of various fractional diffusion equations relies on fairly standard procedures dictated by the form of the single-step probability properties, except that we have added evanescence to the picture and are thus able to see the complex interplay of motion and evanescence, at least under our assumptions. Eventually we hope to be able to relax some of our more stringent assumptions. We also plan to include other particle loss mechanisms such as bimolecular reactions in our scheme, possibly at the expense of introducing some kind of mean-field assumption to deal with the complexity arising from effects of cooperativity. The existing formalism can be adapted to some situations where instead of evanescence we have particle sources, or perhaps sources and sinks simultaneously. However, in some cases particle sources require special scrutiny. For example, special care is needed when dealing with particles that give rise to offspring because one must specify the rules surrounding the location and time of creation of new particles, especially when the pertinent jump and waiting time distributions have long tails.

In addition to these long-term goals, we are in the process of applying our results to a number of specific systems. We have, for instance, considered the problem of the survival probability $P(t)$ of an immobile target that is surrounded by a $d$-dimensional sea of evanescent traps whose motion may be subdiffusive if the waiting times for motion are too long, or superdiffusive if the jumps are sufficiently long, or a mixture of both. For the one-dimensional subdiffusive case, we have been able to rederive the key result $\ln[P(t)] \propto -\int_0^t dt'\,\bar{\rho}(t')t'^{\gamma/2-1}$ obtained in [17] by a different method [here $\bar{\rho}(t)$ stands for the decaying mean density of traps]. The starting point to recover the above formula is Eq. (37) describing the kinetics of a single trap. In the route to the solution one can apply similar techniques to the ones employed in [18] for the nonevanescent case. The above result for the survival probability as well as higher dimensional extensions thereof are objects of our current research.

Finally, we end with an interesting observation that does not appear obvious. One might, instead of Eq. (14), have been motivated to write the integral equation

$$w(\mathbf{r},t|\mathbf{r}_0,0) = \int d\mathbf{r}' \int_0^t dt'\Psi(\mathbf{r}'-\mathbf{r}_0,t')\frac{\varphi(\mathbf{r}',t')}{\varphi(\mathbf{r}_0,0)}w(\mathbf{r},t|\mathbf{r}',t')$$
$$+ \Phi(t)\frac{\varphi(\mathbf{r}_0,t)}{\varphi(\mathbf{r}_0,0)}\delta(\mathbf{r}-\mathbf{r}_0). \quad (49)$$

Again in the language of Hughes [14], this corresponds to a "partition over the first step." What is interesting is that we are not able to arrive at any reasonable fractional diffusion equation starting from this integral equation, in any case not by the methods followed in this paper for Eq. (14) even though one might have expected a certain symmetry to the situation. This, too, is a question to be explored further.

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