Subdiffusion with particle immobilization process described by a differential equation with Riemann-Liouville-type fractional time derivative

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An equation describing subdiffusion with possible immobilization of particles is derived by means of the continuous time random walk model. The equation contains a fractional time derivative of Riemann-Liouville type which is a differential-integral operator with the kernel defined by the Laplace transform; the kernel controls the immobilization process. We propose a method for calculating the inverse Laplace transform providing the kernel in the time domain. In the long time limit the subdiffusion-immobilization process reaches a stationary state in which the probability density of a particle distribution is an exponential function.

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I. INTRODUCTION

Subdiffusion occurs in a medium in which random walk of molecules is very hindered; see for example Refs. [1–9]. The examples are transport of some molecules in living cells [10], in viscoelastic chromatin networks [11], and in porous media [12]; transport of water in aqueous sucrose glasses [13]; and transport of sugars in agarose gel [14]. In some processes diffusing molecules may be eliminated from further diffusion in different ways. There may be a particle decay due to a reaction when it meets other molecules [15–17]. Since the particle disappears, the probability density $P(x,t)$ that the particle is at a point x in time t is not normalized:

$$\int_{-\infty}^{\infty} P(x,t)dx < 1. \tag{1}$$

Another process that eliminates a particle from further diffusion is a permanent immobilization of the particle. In this case, the probability of finding a molecule in the system is equal to 1 at any time. It is obvious that this process cannot be described by a diffusion-reaction equation. The diffusion-immobilization process has been observed in diffusion of chemically reactive gases in polymer layer systems [18], transport of molecules in zeolites [19], signal transduction in living cells [20], drug release processes [21], immobilization of enzymes affecting catalytic reactions [22], oxygen diffusion trough gels [23], diffusion and immobilization of dyes [24], and lithium ions in some nanocomposite anodes [25]. Both processes mentioned above can occur in diffusion of antibiotics in a bacterial biofilm. One of the bacteria defense mechanisms is to disintegrate the antibiotic molecules, and the process can be described by diffusion-reaction equations. In the other one bacteria can thicken the biofilm immobilizing antibiotic molecules [26,27]; see also Refs. [28,29] and references cited therein. The immobilized molecules have not disappeared; they can further interact with the environment. The process of immobilization of diffusing molecules has been described by a diffusion equation with an additional term describing the immobilization process [18,21,25,30,31].

An effective tool for deriving the normal and anomalous diffusion equations is the continuous time random walk (CTRW) model [1–3,5,7,9,32–36]. In particular, the model has been used to derive subdiffusion-reaction equations [37–41]. We use the CTRW model to derive an equation describing subdiffusion with particle immobilization in a one-dimensional homogeneous system. We assume that after each jump a particle can be immobilized with the same probability which does not change with time and is independent of a particle position. This process is described by the subdiffusion equation with a time derivative of the Riemann-Liouville type, which is a differential-integral operator with a kernel controlling the immobilization process. Based on the Green's functions, we show that the process reaches a steady state in the long time limit.

II. MODEL

Within the CTRW model, when the average length of a single particle jump ϵ is finite the form of the subdiffusion equation is determined by the probability density ψ of the waiting time for a particle to jump. In terms of the Laplace transform, $\mathcal{L}[f(t)](s) = \int_0^\infty e^{-st} f(t)dt \equiv \hat{f}(s)$, the equation generated by the function ψ is as follows:

$$s\hat{P}(x,s) - P(x,0) = \frac{\epsilon^2 s\hat{\psi}(s)}{2[1-\hat{\psi}(s)]} \frac{\partial^2 \hat{P}(x,s)}{\partial x^2}, \tag{2}$$

and the derivation of this equation is described in the Appendix.

We make the following assumptions.

(1) The probability of finding a particle in the system is equal to 1 at any time:

$$\int_{-\infty}^{\infty} P(x,t)dx = 1. \tag{3}$$

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(2) Since the particle can be permanently stopped, the probability that the particle will make a jump is less than 1:

$$\int_0^\infty \psi(t)dt < 1. \tag{4}$$

A. Subdiffusion equation

To obtain the subdiffusion equation we assume

$$\hat{\psi}(s) = \frac{1}{1+\tau s^\alpha}, \tag{5}$$

where $0 < \alpha < 1$, and τ is a parameter with the units of s^α. This function satisfies the normalization condition

$$\int_0^\infty \psi(t)dt \equiv \hat{\psi}(0) = 1. \tag{6}$$

This condition means that the particle cannot be stopped permanently with nonzero probability. From Eqs. (2) and (5) we get

$$s\hat{P}(x,s) - P(x,0) = Ds^{1-\alpha}\frac{\partial^2 \hat{P}(x,s)}{\partial x^2}, \tag{7}$$

where $D = \epsilon^2/2\tau$ is a subdiffusion coefficient given in the units of m^2/s^α. Due to the relations

$$\mathcal{L}^{-1}[s\hat{f}(s) - f(0)](t) = \frac{df(t)}{dt}, \tag{8}$$

$$\mathcal{L}^{-1}[s^\beta \hat{f}(s)](t) = \frac{{}^{\mathrm{RL}}d^\beta f(t)}{dt^\beta}, \tag{9}$$

$0 < \beta < 1$, where

$$\frac{{}^{\mathrm{RL}}d^\beta f(t)}{dt^\beta} = \frac{1}{\Gamma(1-\beta)}\frac{d}{dt}\int_0^t (t-u)^{-\beta} f(u)du \tag{10}$$

is the Riemann-Liouville time fractional derivative of the order $\beta \in (0,1)$. From Eqs. (7)–(9) we get the subdiffusion equation

$$\frac{\partial P(x,t)}{\partial t} = D\frac{{}^{\mathrm{RL}}\partial^{1-\alpha}}{\partial t^{1-\alpha}}\frac{\partial^2 P(x,t)}{\partial x^2}. \tag{11}$$

B. Subdiffusion-immobilization equation

In order to find a function $\psi(t)$ that satisfies Eq. (4), i.e., $\hat{\psi}(0) < 1$, we assume that the Laplace transform of the function is

$$\hat{\psi}(s) = \frac{1}{1+\tau\gamma+\tau s^\alpha}, \tag{12}$$

where $0 < \alpha < 1$, and the parameter γ, which controls molecule immobilization, is given in the units of $1/s^\alpha$. The probability p_s of stopping the molecule permanently is $p_s = 1 - \hat{\psi}(0) = \tau\gamma/(1+\tau\gamma)$, and then $\gamma = p_s/[(1-p_s)\tau]$. The function ψ is interpreted here as a probability density of waiting time for a particle to jump provided that the particle has not been permanently immobilized by this time. From Eqs. (2) and (12) we get

$$s\hat{P}(x,s) - P(x,0) = D\frac{s^{1-\alpha}}{1+\gamma s^{-\alpha}}\frac{\partial^2 \hat{P}(x,s)}{\partial x^2}. \tag{13}$$

The inverse Laplace transform of the right-hand side of Eq. (13) is calculated using the formula

$$\mathcal{L}^{-1}\left[\frac{s^{1-\alpha}}{1+\gamma s^{-\alpha}}\hat{f}(s)\right](t) = \frac{{}^{\mathrm{RL}}_F d^{1-\alpha} f(t)}{dt^{1-\alpha}}, \tag{14}$$

where

$$\frac{{}^{\mathrm{RL}}_F d^{1-\alpha} f(t)}{dt^{1-\alpha}} = \frac{d}{dt}\int_0^t F_\alpha(t-t';\gamma) f(t')dt' \tag{15}$$

is the Riemann-Liouville-type fractional derivative with the kernel F_α which is defined by its Laplace transform

$$\hat{F}_\alpha(s;\gamma) = \frac{1}{\gamma + s^\alpha}. \tag{16}$$

For $\gamma = 0$, this derivative is the Riemann-Liouville derivative Eq. (10) of the order $1-\alpha$. Equations (13)–(16) provide the following subdiffusion-immobilization equation (SIE):

$$\frac{\partial P(x,t)}{\partial t} = D\frac{{}^{\mathrm{RL}}_F\partial^{1-\alpha}}{\partial t^{1-\alpha}}\frac{\partial^2 P(x,t)}{\partial x^2}. \tag{17}$$

Calculation of the inverse transform of Eq. (16) is usually done by power series expansion of the function when $\gamma/s^\alpha < 1$, and then inverting the transform term by term using the formula $\mathcal{L}^{-1}[1/s^\beta](t) = t^{\beta-1}/\Gamma(\beta)$, $\beta > 0$. The result is the Mittag-Leffler (ML) function [42,43]. However, this procedure is valid for relatively large values of the parameter s, which correspond to small values of the time variable. To get the inverse Laplace transform over the whole time domain we propose to use the following method.

(1) Instead of $\hat{F}_\alpha$ in Eq. (16) find the inverse transform of $\hat{F}_\alpha(s,\gamma)e^{-as^\mu}$, where $a, \mu > 0$.

(2) Expand $\hat{F}_\alpha$ in a power series of s considering both cases $s^\alpha > \gamma$ and $s^\alpha < \gamma$ separately.

(3) Use the formula [44]

$$\mathcal{L}^{-1}\left[s^\nu e^{-as^\mu}\right](t) \equiv f_{\nu,\mu}(t;a) = \frac{1}{t^{\nu+1}}\sum_{n=0}^\infty \frac{1}{n!\Gamma(-n\mu-\nu)}\left(-\frac{a}{t^\mu}\right)^n, \tag{18}$$

where $a, \mu > 0$.

(4) Calculate the limit of $a \to 0^+$ in the obtained functions. We note that

$$f_{\nu,\mu}(t;0^+) = \frac{1}{t^{\nu+1}\Gamma(-\nu)}, \tag{19}$$

and the result is independent of the parameter μ.

From the formula

$$\frac{e^{-as^\mu}}{\gamma + s^\alpha} = \begin{cases} e^{-as^\mu}\sum_{n=0}^\infty (-\gamma)^n s^{-(n+1)\alpha}, & s > \gamma^{1/\alpha}, \\ \frac{e^{-as^\mu}}{\gamma}\sum_{n=0}^\infty \left(-\frac{1}{\gamma}\right)^n s^{n\alpha}, & s < \gamma^{1/\alpha}, \end{cases} \tag{20}$$

and Eqs. (18) and (19) we obtain

$$F_\alpha(t;\gamma) = \begin{cases} \frac{1}{t^{1-\alpha}}E_{\alpha,\alpha}(-\gamma t^\alpha), & t < t_b, \\ -\frac{1}{\gamma^2 t^{1+\alpha}}\tilde{E}_{\alpha,\alpha}\left(-\frac{1}{\gamma t^\alpha}\right), & t > t_b, \end{cases} \tag{21}$$

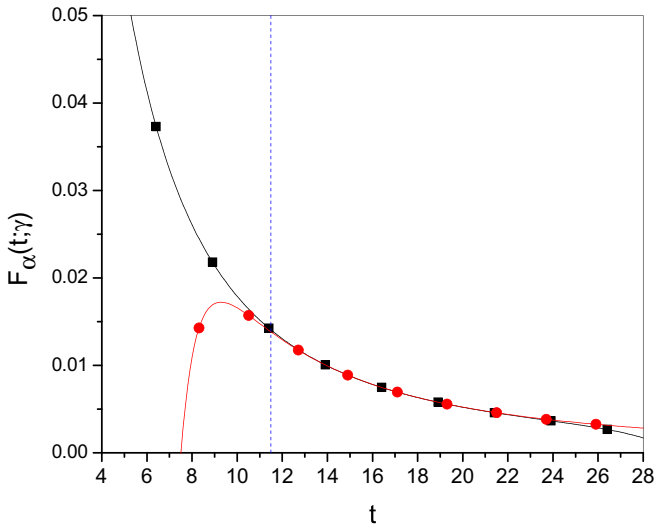

FIG. 1. Plot of the function F_α. The dashed vertical line shows the location of the parameter $t_b = 11.5$. The solid line with squares is the plot of the upper function in Eq. (21) which describes F_α for $t < t_b$; the solid line with circles is the plot of the lower function in Eq. (21) which represents F_α for $t > t_b$. In the numerical calculations, the leading 20 terms in the series appearing in the functions $E_{\alpha,\alpha}$ and $\tilde{E}_{\alpha,\alpha}$ have been included.

where $E_{\alpha,\beta}(u) = \sum_{n=0}^{\infty} \frac{u^n}{\Gamma(\alpha n+\beta)}$, with $\alpha, \beta > 0$, is the two-parameter ML function, and $\tilde{E}_{\alpha,\beta}(u) = \sum_{n=0}^{\infty} \frac{u^n}{\Gamma(-\alpha n-\beta)}$ is a generalization of the ML function for negative parameters. We note that conditions $s > \gamma^{1/\alpha}$ and $s < \gamma^{1/\alpha}$ do not determine the parameter t_b. For example, the condition $s > \gamma^{1/\alpha}$ is equivalent to $1/s^{\beta+1} < 1/(s^\beta \gamma^{1/\alpha})$ for $\beta > 0$ (assuming that s is a real positive parameter). The inverse Laplace transform of the inequality provides $t < \beta/\gamma^{1/\alpha}$ where β is a positive number. Thus, the above inequality does not determine t_b. Here we define the parameter t_b as the shorter time at which the upper and the lower functions in Eq. (21) are matched; see Fig. 1. We mention that the parameter t_b has no physical interpretation; it separates the time domains in which the methods of Laplace transform calculations are different.

In terms of the Laplace transform the solution to Eq. (16) (the Green's function) for the initial condition $P(x, 0) = \delta(x)$, where δ is the Dirac-delta function and boundary conditions $P(\pm\infty, t) = 0$, is

$$\hat{P}(x, s) = \frac{\sqrt{\gamma + s^\alpha}}{2s\sqrt{D}}\, e^{-|x|\frac{\sqrt{\gamma+s^\alpha}}{\sqrt{D}}}. \tag{22}$$

The solution fulfils the condition $\int_{-\infty}^{\infty} \hat{P}(x, s)dx = 1/s$ which provides the normalization of the function P in Eq. (3).

Let $\gamma \neq 0$. We calculate the inverse Laplace transform of the function (22) for small and large values of s separately. In calculation, we use the formulas $\sqrt{1+u} \approx 1 + u/2 - u^2/8$ and $e^{-u} \approx 1 - u + u^2/2$, $u \to 0$, and keep the leading terms in the obtained series. When $s^\alpha > \gamma$ we obtain

$$\hat{P}(x, s) = \frac{1}{2\sqrt{D}s^{1-\alpha/2}}\left(1 - \frac{b_1}{s^{\alpha/2}} + \frac{b_2}{s^\alpha}\right) e^{-\frac{|x|}{\sqrt{D}}s^{\alpha/2}}, \tag{23}$$

where $b_1 = \gamma|x|/2\sqrt{D}$ and $b_2 = (\gamma/2)(1 + |x|^2\gamma/2\sqrt{D})$. If $s^\alpha < \gamma$, we get

$$\hat{P}(x, s) = \frac{\sqrt{\gamma}}{2s\sqrt{D}} e^{-\sqrt{\frac{\gamma}{D}}|x|(1+\frac{s^\alpha}{2\gamma})}\left[1 + \frac{s^\alpha}{2\gamma} - b\frac{s^{2\alpha}}{\gamma^2}\right], \tag{24}$$

where $b = \sqrt{\gamma/D}|x| + 1/8$. Equations (18) and (23) provide the Green's functions in the limit of short time:

$$P(x, t) = \frac{1}{2\sqrt{D}}[f_{-1+\alpha/2,\alpha/2}(t;\eta) - b_1 f_{-1,\alpha/2}(t;\eta) + b_2 f_{-1-\alpha/2,\alpha,2}(t;\eta)], \tag{25}$$

where $\eta = |x|/\sqrt{D}$. From Eqs. (18) and (24) we get the Green's function in the long time limit:

$$P(x, t) = \frac{1}{2}\sqrt{\frac{\gamma}{D}} e^{-\sqrt{\frac{\gamma}{D}}|x|}\left[f_{-1,\alpha}(t;\xi) + \frac{1}{2\gamma} f_{\alpha-1,\alpha}(t;\xi) - \frac{b}{\gamma^2} f_{2\alpha-1,\alpha}(t;\xi)\right], \tag{26}$$

where $\xi = |x|/2\sqrt{D\gamma}$.

Since the mean particle position equals zero, in terms of the Laplace transform the mean square displacement of the particle is

$$\mathcal{L}[\langle(\Delta x)^2(t)\rangle](s) = \int_{-\infty}^{\infty} x^2 \hat{P}(x, s)dx = \frac{2D}{s(\gamma + s^\alpha)}. \tag{27}$$

When $\gamma \neq 0$, for small s we have $\mathcal{L}[\langle(\Delta x)^2(t)\rangle](s) = 2D/[1/s - 1/(\gamma s^{1-\alpha})]$. Thus, in the limit of long time we get

$$\langle(\Delta x)^2(t)\rangle = \frac{2D}{\gamma}\left[1 - \frac{1}{\gamma\Gamma(1-\alpha)t^\alpha}\right]. \tag{28}$$

In the limit $t \to \infty$, the stationary state described by the following function is reached:

$$P(x, t \to \infty) \equiv P_{\rm st}(x) = \frac{1}{2}\sqrt{\frac{\gamma}{D}}\, e^{-\sqrt{\frac{\gamma}{D}}|x|}. \tag{29}$$

For illustration, plots of functions F_α and P are shown in Figs. 1 and 2, respectively. The parameters are $\alpha = 0.7$, $\gamma = 0.6$, and $D = 10$, and all parameters are given in arbitrarily chosen units. In Fig. 3 the Green's functions for the stationary state are presented.

III. FINAL REMARKS

The process of subdiffusion with particle immobilization in a one-dimensional system can be described by an equation with a fractional time derivative of the Riemann-Liouville type, which is a differential-integral operator with the kernel F_α defined by its Laplace transform Eq. (16). The equation can be easily generalized to the three-dimensional case just like the "ordinary" subdiffusion equation. We have proposed a method for determining the inverse Laplace transform of the kernel. In our opinion, this method can be widely used for calculating inverse Laplace transforms $\mathcal{L}^{-1}[\hat{f}(s)](t)$ for a wide class of functions f.

In a homogeneous unbounded system the subdiffusion-immobilization process reaches a stationary state which is described by $P_{\rm st}(x)$ in Eq. (29). This distribution depends only on the quotient γ/D expressed in the units of $1/m^2$ and it does

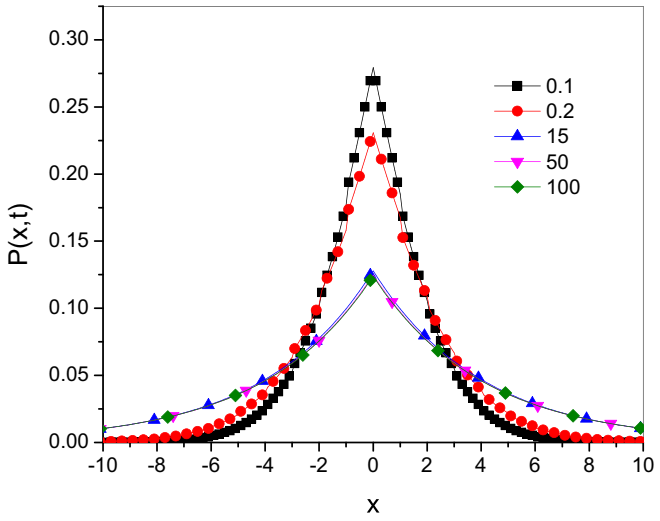

FIG. 2. Plots of Green's functions for times given in the legend. The plots represent the function Eq. (23) for $t = 0.1, 0.5$ and Eq. (24) for $t = 15, 50, 100$.

not explicitly depend on the parameter α. The achievement of the steady state is suggested by Fig. 2, where the Green's functions for relatively long times differ very little from each other. In the stationary state there is $\langle(\Delta x)^2(t \to \infty)\rangle = \frac{2D}{\gamma}$, and the particle is finally immobilized with probability equal to 1. We mention that an interesting issue worth further consideration may be the achievement of a stationary state in a system bounded by partially permeable membranes or impermeable walls, as well as in a heterogeneous system.

The subdiffusion-immobilization process is described by Eq. (17) that can be obtained in practice by replacing the time fractional Riemann-Liouville derivative Eq. (10) with the more general Riemann-Liouville-type derivative with the kernel F_α in Eq. (15) in the ordinary subdiffusion equation Eq. (11). There is a different situation than in the subdiffusion-reaction equation in which the reaction term is involved in the ordinary subdiffusion equation; see for example Refs. [15,17,41]. We mention that the Riemann-Liouville-type fractional derivatives with different kernels have been considered in Refs. [45–48].

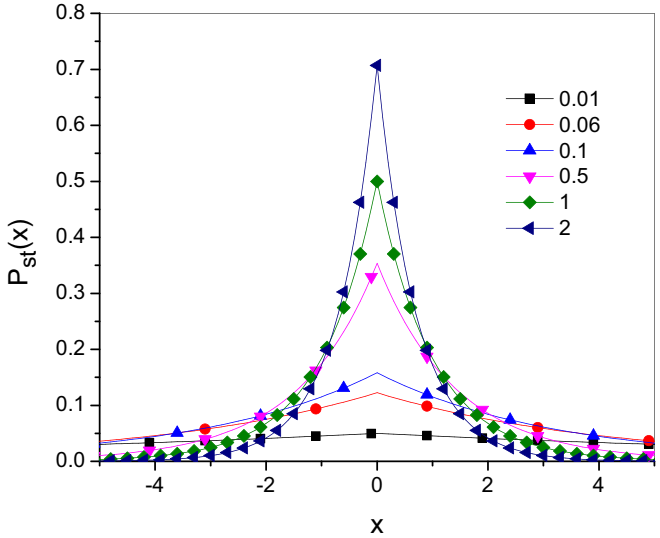

FIG. 3. Plots of the function $P_{\rm st}$ in Eq. (29) for different values of the ratio γ/D given in the legend.

An interesting problem is to consider a subdiffusion-immobilization equation with the fractional Riemann-Liouville-type time derivative with respect to another function g (g-subdiffusion-immobilization equation). Such an equation could significantly extend the range of processes it describes. The g-subdiffusion equation has been recently considered in Ref. [49] (see also references cited therein) in which subdiffusion equation with the fractional Caputo time derivative of the order α, equivalent to Eq. (11), has been considered. The g-subdiffusion equation results in a change in the time scale, $t \to g(t)$, comparing with the ordinary subdiffusion equation. The stochastic interpretation of g subdiffusion is based on a modified CTRW model. The g-subdiffusion equation describes, among others, a process of continuous transition from subdiffusion to ultraslow diffusion (slow subdiffusion), to subdiffusion with changed parameters, and to superdiffusion. Another process in which the time clock has been changed compared to ordinary subdiffusion is the process described by a tempered subdiffusion equation (TSE) with a tempered fractional time derivative [50,51]. This equation can be derived from the CTRW model assuming that ψ is a tempered α-stable distribution. We mention that the equations SIE Eq. (17) and TSE are not equivalent. The qualitative differences between these equations are, among others, in their stationary states. The process described by TSE reaches the stationary state when particle diffuses in a potential field V [51]. When $dV/dx = 0$, the stationary state is described by the uniform distribution for TSE and by the function Eq. (29) for SIE.

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APPENDIX: DERIVATION OF EQ. (2)

To derive the subdiffusion equation we use a simple model of a particle random walk along a one-dimensional homogeneous lattice. Usually, in the CTRW model both a particle jump length and waiting time for a particle to jump are random variables. We assume that the jump length distribution λ has the form $\lambda(x) = \frac{1}{2}[\delta(x-\epsilon) + \delta(x+\epsilon)]$. Random walk with discrete time n is described by the equation $P_{n+1}(m) = \frac{1}{2}P_n(m+1) + \frac{1}{2}P_n(m-1)$, where $P_n(m)$ is a probability that a diffusing particle is at the position m after the nth step. Let the initial particle position be $m = 0$. Moving from discrete m to continuous x spatial variable we assume $x = m\epsilon$ and $P_n(x) = P_n(m)/\epsilon$, where ϵ is a distance between discrete sites. The above equations and the relation $[P_n(x+\epsilon) + P_n(x-\epsilon) - 2P_n(x)]/\epsilon^2 = \partial^2 P_n(x)/\partial x^2$, where $\epsilon \to 0$, provide the following equation in the limit of small ϵ;

$$P_{n+1}(x) - P_n(x) = \frac{\epsilon^2}{2}\frac{\partial^2 P_n(x)}{\partial x^2}. \tag{A1}$$

To move from discrete to continuous time we use the formula $P(x,t)=\sum_{n=0}^{\infty}Q_n(t)P_n(x)$ [32], where $Q_n(t)$ is the probability that a diffusing particle takes n steps in the time interval $(0,t)$. The function Q_n is a convolution of n distributions ψ and a function $U(t)=1-\int_0^t\psi(t')dt'$ which is the probability that a particle does not change its position after the nth step, $\hat{U}(s)=[1-\hat{\psi}(s)]/s$, $Q_n(t)=(\underbrace{\psi*\psi*\ldots*\psi}_{n\text{ times}}*U)(t)$, where $(f*h)(t)=\int_0^t f(u)h(t-u)du$. Due to the following property $\mathcal{L}[(f*h)(t)](s)=\hat{f}(s)\hat{h}(s)$ we obtain

$$\hat{P}(x,s)=\frac{1-\hat{\psi}(s)}{s}\sum_{n=0}^{\infty}\hat{\psi}^n(s)P_n(x). \tag{A2}$$

Combining Eqs. (A1) and (A2) we get Eq. (2).

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