Fractional Fokker-Planck Equations for Subdiffusion with Space-and-Time-Dependent Forces.

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We have derived a fractional Fokker-Planck equation for subdiffusion in a general space-and-time-dependent force field from power law waiting time continuous time random walks biased by Boltzmann weights. The governing equation is derived from a generalized master equation and is shown to be equivalent to a subordinated stochastic Langevin equation.

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Over the past few decades there has been an enormous growth in the numbers of papers devoted to experimental and theoretical aspects of anomalous diffusion [1, 2]. The landmark review by Metzler and Klafter in 2000 [1] has been particularly influential, promoting the description of anomalous diffusion within the framework of continuous time random walks (CTRWs) and fractional calculus. There are now numerous applications utilizing this approach in physics, chemistry, biology and finance [2].

A central theoretical result in this research was the derivation [3, 4] of a fractional Fokker-Planck (Smoluchowksi) equation [5]

$$\frac{\partial P}{\partial t} = {}_0D_t^{1-\alpha}\left[\kappa_\alpha \frac{\partial^2}{\partial x^2} - \frac{1}{\eta_\alpha}\frac{\partial}{\partial x}F(x)\right]P(x,t) \qquad (1)$$

to describe the evolution of the probability density function $P(x,t)$ for subdiffusion in an external space-dependent force field $F(x)$. In this equation,

$$ {}_0D_t^{1-\alpha}Y(t) = \frac{d}{dt}\,{}_0I_t^{\alpha} \qquad (2)$$

where

$$ {}_0I_t^{\alpha} = \frac{1}{\Gamma(\alpha)}\int_0^t \frac{Y(t')}{(t-t')^{1-\alpha}}\,dt' \qquad (3)$$

is the Riemann-Liouville fractional integral of order $\alpha \in (0,1)$, κ_α is a fractional diffusion coefficient, $\eta_\alpha = (\beta\kappa_\alpha)^{-1}$ is a fractional friction coefficient, and β is the inverse temperature $k_B T$. The fractional Fokker-Planck equation (FFPE), Eq.(1), was derived from the continuous time random walk model of Montroll and Weiss [6], with power law waiting times [3, 4].

More recently a modified FFPE,

$$\frac{\partial P}{\partial t} = \left[\kappa_\alpha \frac{\partial^2}{\partial x^2} - \frac{1}{\eta_\alpha}\frac{\partial}{\partial x}F(t)\right]{}_0D_t^{1-\alpha}P(x,t), \qquad (4)$$

was derived from power law waiting time CTRWs, using a generalized master equation [7], for subdiffusion in a time-dependent force field $F(t)$. The modified FFPE, Eq.(4), was also derived for subdiffusion in dichotomously alternating force fields [8, 9], $F(x)\xi(t)$ with $\xi(t) = \pm 1$, but a FFPE to model subdiffusion in general space-and-time-dependent force fields $F(x,t)$ has remained elusive [8–11]. On the other hand, a subordinated stochastic Langevin equation has been proposed for modelling subdiffusion in space-and-time-dependent force fields [10]. More recently [12], in the case of time-dependent forces, the moments of the stochastic process defined by this stochastic Langevin equation were shown to coincide with the moments of the modified FFPE, Eq.(4). There have been numerous papers on FFPEs in recent years relying on *ad hoc* or phenomenological models [8, 11, 13, 14].

In this letter we derive the FFPE,

$$\frac{\partial P}{\partial t} = \left[\kappa_\alpha \frac{\partial^2}{\partial x^2} - \frac{1}{\eta_\alpha}\frac{\partial}{\partial x}F(x,t)\right]{}_0D_t^{1-\alpha}P(x,t), \qquad (5)$$

from power law waiting time CTRWs, using a generalized master equation, for subdiffusion in a space-and-time-dependent force field $F(x,t)$. This FFPE is shown to be formally equivalent to the subordinated stochastic Langevin equation in [10] for space-and-time-dependent forces. We also show that the original FFPE, Eq.(1), generalized by replacing $F(x)$ with $F(x,t)$, can be recovered from power law waiting time CTRWs in an *ad-hoc* generalization of the CTRW particle balance equation if the diffusing particles respond to the force field at the start of the waiting time prior to jumping. These derivations, and further extensions to include reactions, are described in greater detail, for chemotactic forcing, in a related publication [15].

Our starting point is the generalized master equation approach developed in [7, 16]. This approach utilizes two balance conditions. The balance equation for the concentration of particles, $n_i(t)$, at the site i and time t is

$$\frac{dn_i(t)}{dt} = J_i^+(t) - J_i^-(t), \tag{6}$$

where $J_i^{\pm}$ are the gain (+) and loss (-) fluxes at the site i. The second balance equation is a conservation equation for the arriving flux of particles at the point i. In general, to allow for biased CTRWs in a space-and-time-dependent force field, we write

$$J_i^+(t) = p_r(x_{i-1}, t)J_{i-1}^-(t) + p_l(x_{i+1}, t)J_{i+1}^-(t), \tag{7}$$

where $p_r(x,t)$ and $p_l(x,t)$ are the probabilities of jumping from x to the adjacent grid point, to the right and left directions respectively. The two balance equations can be combined to yield

$$\frac{dn_i(t)}{dt} = p_r(x_{i-1}, t)J_{i-1}^-(t) + p_l(x_{i+1}, t)J_{i+1}^-(t) - J_i^-(t). \tag{8}$$

For CTRWs with a waiting time probability density function $\psi(t)$ the loss flux at site i is from those particles that were originally at i at $t = 0$ and wait until time t to leave, and those particles that arrived at an earlier time t' and wait until time t to leave, hence [16]

$$J_i^-(t) = \psi(t)n_i(0) + \int_0^t \psi(t - t')J_i^+(t')\, dt'. \tag{9}$$

We can combine Eq. (6) and Eq. (9) to obtain

$$J_i^-(t) = \psi(t)n_i(0) + \int_0^t \psi(t - t')\left[J_i^-(t') + \frac{dn_i(t')}{dt}\right] dt', \tag{10}$$

and then

$$\widehat{J_i^-}(s) = \widehat{\psi}(s)n_i(0) + \widehat{\psi}(s)\left[\widehat{J_i^-}(s) + s\widehat{n}_i(s) - n_i(0)\right], \tag{11}$$

where the hat denotes a Laplace transform with respect to time and s is the Laplace transform variable. This simplifies further to

$$\widehat{J_i^-}(s) = \frac{\widehat{\psi}(s)}{\widehat{\Phi}(s)}\widehat{n}_i(s), \tag{12}$$

where $\widehat{\Phi}(s)$ is the Laplace transform of the survival probability

$$\Phi(t) = \int_t^\infty \psi(t')\, dt'. \tag{13}$$

In the CTRW model, subdiffusion originates from a heavy-tailed waiting-time density with long-time behaviour [1]

$$\psi(t) \sim \frac{\kappa}{\tau}\left(\frac{t}{\tau}\right)^{-1-\alpha} \tag{14}$$

where α is the anomalous exponent, τ is a characteristic waiting-time scale, and κ is a dimensionless constant. Using a Tauberian (Abelian) theorem for small s [17]

$$\frac{\widehat{\psi}(s)}{\widehat{\Phi}(s)} \sim A_\alpha \frac{s^{1-\alpha}}{\tau^\alpha} \tag{15}$$

where $A_\alpha = \frac{\alpha}{\kappa\alpha(1-\alpha)}$. In the special case of a Mittag-Leffler waiting time density [18] the ratio, Eq.(15), is exact and $A_\alpha = 1$. The loss flux can now be obtained by using the ratio, Eq(15), in Eq.(12) and inverting the Laplace transform. Noting that the Laplace Transform of a Riemann-Liouville fractional derivative of order α, where $0 < \alpha \le 1$, is given by [19]

$$\mathcal{L}\left\{{}_0D_t^\alpha f(t)\right\}(s) = s^\alpha \widehat{f}(s) - \left[{}_0I_t^{1-\alpha} f(t)\right|_{t=0} \tag{16}$$

this yields

$$J_i^-(t) = \frac{A_\alpha}{\tau^\alpha}\,{}_0D_t^{1-\alpha} n_i(t), \tag{17}$$

where we have assumed that the last term in Eq. (16) is zero. Using this result in Eq.(8) yields,

$$\frac{dn_i(t)}{dt} = \frac{A_\alpha}{\tau^\alpha}\left\{p_r(x_{i-1}, t)\,{}_0D_t^{1-\alpha} n_{i-1}(t) + p_l(x_{i+1}, t)\,{}_0D_t^{1-\alpha} n_{i+1}(t) - {}_0D_t^{1-\alpha} n_i(t)\right\}. \tag{18}$$

The jump probabilities are biased by the external space-and-time-dependent force. Here we consider (near thermodynamic equilibrium) Boltzmann weights with

$$p_r(x_i, t) = C\exp\left(-\beta\left(V(x_{i+1}, t) - V(x_i, t)\right)\right), \tag{19}$$

and

$$p_l(x_i, t) = C\exp\left(-\beta\left(V(x_{i-1}, t) - V(x_i, t)\right)\right). \tag{20}$$

The jump probabilities are determined at the end of the waiting time, when the particle must jump, so that

$$p_r(x_i, t) + p_l(x_i, t) = 1, \tag{21}$$

which defines C, and then

$$p_l(x_i, t) - p_r(x_i, t) = \frac{e^{-\beta V(x_{i-1}, t)} - e^{-\beta V(x_{i+1}, t)}}{e^{-\beta V(x_{i-1}, t)} + e^{-\beta V(x_{i+1}, t)}}. \tag{22}$$

The spatial continuum limit of Eq.(18) can now be obtained by setting $x_i = x$ and $x_{i\pm1} = x \pm \Delta x$ and carrying out Taylor series expansions in x. Retaining terms to order $(\Delta x)^2$ and using Eq.(21) yields

$$\frac{\partial n(x,t)}{\partial t} = \frac{A_\alpha}{\tau^\alpha}\left\{\Delta x\frac{\partial}{\partial x}\left[\left(p_l(x,t) - p_r(x,t)\right){}_0D_t^{1-\alpha} n(x,t)\right] + \frac{\Delta x^2}{2}\frac{\partial^2}{\partial x^2}\,{}_0D_t^{1-\alpha} n(x,t)\right\}. \tag{23}$$

The Taylor series expansion of Eq.(22) yields

$$p_l(x,t) - p_r(x,t) \approx \beta\Delta x \frac{\partial V(x,t)}{\partial x} + O(\Delta x^3), \qquad (24)$$

and then

$$\frac{\partial n(x,t)}{\partial t} = \frac{A_\alpha \beta \Delta x^2}{\tau^\alpha} \frac{\partial}{\partial x}\left[\frac{\partial V(x,t)}{\partial x} \, {}_0D_t^{1-\alpha} n(x,t)\right] + \frac{A_\alpha \Delta x^2}{2\tau^\alpha}\frac{\partial^2}{\partial x^2} \, {}_0D_t^{1-\alpha} n(x,t) + O(\Delta x^4). \qquad (25)$$

In the limit $\Delta x \to 0$ and $\tau \to 0$, with $\kappa_\alpha = \frac{A_\alpha \Delta x^2}{2\tau^\alpha}$, and $\eta_\alpha = (2\beta\kappa_\alpha)^{-1}$ we recover the FFPE, Eq.(5), for subdiffusion in an external space-and-time-dependent force field

$$F(x,t) = -\frac{\partial V(x,t)}{\partial x}. \qquad (26)$$

In carrying out the limit $\Delta x \to 0$ we restrict ourselves to external forces that are spatially smooth but the temporal part of the diffusion limit, $\tau \to 0$, does not place any restrictions on the temporal behaviour of the external force. Implicitly there may be restrictions on the external force in the sense that we have assumed that the external force does not affect the waiting time probability density. This is not a restriction on the temporal or spatial behaviour of the force *per se* but similar to [4, 7] it restricts modelling applications to situations where the waiting time densities can be affected by spatial heterogeneities of the medium but not affected by the external force. Our assumption that the probability to jump left or right is weighted by near thermodynamic equilibrium Boltzmann weights is equivalent to the assumption that the difference between the probabilities to jump left and right is proportional to the external force at that instant in time. This assumption was also made in the derivations of the FFPE in [4, 8, 9].

The FFPE, Eq.(5), can also be derived from the subordinated stochastic Langevin equation motivated by physical arguments [10] to model subdiffusion in a space-and-time-dependent force field. This representation can be formulated as a system of stochastic equations,

$$\begin{pmatrix} dY_t \\ dZ_t \end{pmatrix} = \begin{pmatrix} F(Y_t, Z_t)\eta^{-1} \\ 0 \end{pmatrix} dt + \begin{pmatrix} (2\kappa)^{1/2} dB_t \\ dU_t \end{pmatrix}, \qquad (27)$$

where B_t is a one-dimensional Brownian motion and U_t is a α-stable Lévy subordinator in $[0,\infty), 0 < \alpha < 1$. It is asumed that B_t and U_t are independent stochastic processes and the initial condition is $Y_0 = Z_0 = 0$. The stochastic process representing subdiffusion in a space-and-time-dependent force field is postulated to be given by [10] $X_t = Y(S_t)$ where for $t \geq 0$, S_t is the random time the process U_t exceeds t.

The stochastic differential equation, Eq.(27), belongs to the general class of stochastic processes driven by Lévy noise [20, eq.(6.12)]. The infinitesimal generator for the process Eq.(27) is then given by [20, eq.(6.42)]

$$Af(y,z) = \frac{F(y,z)}{\eta}\frac{\partial}{\partial y} f(y,z) + \kappa \frac{\partial^2}{\partial y^2} f(y,z) + \int_0^\infty \left[f(y, z+z') - f(y,z)\right] \frac{\alpha}{\Gamma(1-\alpha)} z'^{-1-\alpha} dz'. \qquad (28)$$

The Fokker-Planck evolution equation for the probability density $q_t(y,z)$ of the process (Y_t, Z_t) is given by [20, eq.(3.24)]

$$\frac{\partial}{\partial t} q_t(y,z) = A^\dagger q_t(y,z) \qquad (29)$$

where $A^\dagger$ is the operator adjoint to A,

$$A^\dagger f(y,z) = \kappa \frac{\partial^2}{\partial y^2} f(y,z) - \frac{\partial}{\partial y}\left(\frac{F(y,z)}{\eta} f(y,z)\right) - {}_0D_z^\alpha f(y,z). \qquad (30)$$

Now we relate the densities $p_t(x)$ and $q_t(y,z)$ of the stochastic processes X_t and (Y_t, Z_t) respectively. We write ω for a particular (random) path of the latter process, and note that the coordinates at time t, $(Y_t(\omega), Z_t(\omega))$ and $X_t(\omega)$, are functions of ω. For a fixed interval I we can write

$$\int_I p_t(x)\, dx = \langle \delta_I(X_t(\omega)) \rangle \qquad (31)$$

where the angle brackets represent an ensemble average over all paths ω and $\delta_I(x)$ is the indicator function defined as one if $x \in I$ and zero otherwise. Given that X_t can be interpreted as the Y coordinate of the last position of the Markov process (Y, Z) before it exits the set $\mathbb{R} \times [0, t]$, we have

$$\delta_I(X_t(\omega)) = \sum_{t'>0} H\left(t', \omega, \Delta Z_{t'}(\omega)\right), \qquad (32)$$

where $H(t', \omega, \Delta z)$ is given by $\delta_I(Y(t')(\omega))$ if $Z_{t'-}(\omega) \leq t \leq Z_{t'-}(\omega) + \Delta z$ and zero otherwise. This follows since all summands in Eq.(32) equal zero except for $t' = S_t$, in which case $Y_{t'}(\omega) = X_t(\omega)$. The jumps $\Delta z = \Delta Z_{t'}(\omega)$ are a Poisson point process on $(0,\infty)$ whose characteristic measure has the density [20, p.50] $\frac{\alpha}{\Gamma(1-\alpha)}\Delta z^{-1-\alpha}$. We can now combine Eqs.(31), (32) and use the compensation formula in [21, XII (1.10)], to write

$$\int_I p_t(x) dx = \left\langle \int_0^\infty \int_0^\infty H(t', \omega, \Delta z) \frac{\alpha \Delta z^{-1-\alpha}}{\Gamma(1-\alpha)} d\Delta z\, dt' \right\rangle. \qquad (33)$$

After integrating over Δz the right hand side simplifies further to

$$\left\langle \int_0^\infty dt' \delta_I(Y_{t'}(\omega)) \delta_{[0,t]}(Z_{t'}(\omega)) \frac{(t - Z_{t'}(\omega))^{-\alpha}}{\Gamma(1-\alpha)} \right\rangle. \qquad (34)$$

The ensemble average is evaluated using the probability density $q_t(y,z)$ so that

$$\int_I p_t(x)dx = \int_0^\infty dt' \int_I dy \int_0^t dz\; q_{t'}(y,z)\frac{(t-z)^{-\alpha}}{\Gamma(1-\alpha)},$$

and thus

$$p_t(x) = \int_0^\infty dt'\; {}_0I_t^{1-\alpha} q_{t'}(x,t). \tag{35}$$

It also follows that

$$ {}_0D_t^{1-\alpha}p_t(x) = \int_0^\infty q_{t'}(x,t)dt', \tag{36}$$

$$\frac{\partial}{\partial t}p_t(x) = \int_0^\infty {}_0D_t^\alpha q_{t'}(x,t)dt'. \tag{37}$$

We now solve Eqs.(29), (30) for ${}_0D_t^\alpha q_{t'}(x,t)$ and subsitute this into Eq.(37) to obtain

$$\begin{aligned}\frac{\partial}{\partial t}p_t(x) &= \int_0^\infty \left(\kappa\frac{\partial^2}{\partial x^2}q_{t'}(x,t) - \frac{1}{\eta}\frac{\partial}{\partial x}(F(x,t)q_{t'}(x,t))\right.\\ &\quad \left. - \frac{\partial}{\partial t'}q_{t'}(x,t)\right)dt' \end{aligned}\tag{38}$$

and finally, using Eq.(36),

$$\frac{\partial}{\partial t}p_t(x) = \kappa\frac{\partial^2}{\partial x^2}{}_0D_t^{1-\alpha}p_t(x) - \frac{\partial}{\partial x}\left(\frac{F(x,t)}{\eta}{}_0D_t^{1-\alpha}p_t(x)\right) \tag{39}$$

where we have used $q_\infty(x,t) = 0$ and $q_0(x,t) = \delta_{(0,0)}(x,t)$ and assumed $t > 0$. Equation (39) recovers the FFPE for space-and-time-dependent forces, Eq.(5).

A different FFPE can be obtained from the following generalization of the CTRW particle balance equation

$$\begin{aligned} n_i(t) &= n_i(0)\Phi(t) + \int_0^t \left\{p_r(x_{i-1},t')n_{i-1}(t')\right.\\ &\quad \left.+p_l(x_{i+1},t')n_{i+1}(t')\right\}\psi(t-t')\,dt', \end{aligned}\tag{40}$$

where the jump probabilities are evaluated at the start of the waiting time prior to jumping. Note that this equation can be derived from the Montroll-Weiss CTRW formalism (see e.g., [2]) if and only if the jumping probabilities are independent of time. Our inclusion of time dependence is an *ad-hoc* generalization for time dependent jumps. Using Laplace transform methods as above then leads to the discrete space evolution equation

$$\begin{aligned}\frac{dn_i}{dt} &= \frac{A_\alpha}{\tau^\alpha}{}_0D_t^{1-\alpha}\left\{-n_i(t) + p_r(x_{i-1},t)n_{i-1}(t)\right.\\ &\quad \left.+p_l(x_{i+1},t)n_{i+1}(t)\right\}. \end{aligned}\tag{41}$$

It follows from Eqs. (16) and (17) that this is the evolution equation for the loss flux in the generalized master equation approach. After taking the spatial continuum limit of Eq.(41) with Boltzmann weighted jumping probabilities we have

$$\frac{\partial n}{\partial t} = {}_0D_t^{1-\alpha}\left[\kappa_\alpha\frac{\partial^2 n(x,t)}{\partial x^2} + \frac{1}{\eta_\alpha}\frac{\partial}{\partial x}\left(\frac{\partial V(x,t)}{\partial x}n(x,t)\right)\right]. \tag{42}$$

This provides an interpretation of the FFPE

$$\frac{\partial P}{\partial t} = {}_0D_t^{1-\alpha}\left[\kappa_\alpha\frac{\partial^2}{\partial x^2} - \frac{1}{\eta_\alpha}\frac{\partial}{\partial x}F(x,t)\right]P(x,t) \tag{43}$$

as an equation for the loss flux in subdiffusion in a space-and-time-dependent force field.

An experimental test of the FFPEs could be carried out by comparing first moment calculations with the measured response of a subdiffusive system to an external field. The experiment of Allegrini *et al* [22] measured the change in transmitted light intensity as the system response for a nematic liquid crystal in the weakly turbulent regime when the crystal is subject to an alternating electric field. As a first comparison we have computed the first moment $\langle x\rangle$ (which compares with $\Lambda^-(t)$ in the experiment) for an external force $F(x,t) = \cos t$ for each of Eq.(5) and Eq.(43) using $\alpha = 1/2$. The long time response from Eq.(5), $\sim \frac{1}{\sqrt{2}} + \frac{1}{\sqrt{\pi t}}\sin t$, exhibits a so called death of linear response (decay of oscillations) and a Freud effect (oscillations not around zero) [7]. The corresponding results from Eq.(43), $\sim \frac{1}{\sqrt{2}}(\cos t + \sin t) - \frac{1}{\sqrt{\pi t}}\frac{1}{2t}$, exhibit no decay in oscillations and no Freud effect. The experimental results compare reasonably well with Eq.(5) apart from the absence of the Freud effect in the experimental measurement of $\Lambda^-(t)$. This cancellation of the Freud effect could be obtained from Eq.(5) if there was an initial transient external force $F_0 > \epsilon$ before the onset of modulations in the external perturbation $\epsilon\cos t$ and $-\epsilon\cos t$. The results from Eq.(5) showing decaying oscillations and the Freud effect are also consistent with a phenomenological linear response theory, and decaying oscillations but no Freud effect are consistent with a dynamical linear response theory [22].

It is straightforward to obtain numerical solutions of the discrete space evolution equations Eqs.(18), (41) and to simulate the CTRWs using Monte Carlo methods [15].

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