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Use and Abuse of a Fractional Fokker-Planck Dynamics for Time-Dependent Driving

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We investigate a subdiffusive, fractional Fokker-Planck dynamics occurring in time-varying potential landscapes and thereby disclose the failure of the fractional Fokker-Planck equation (FFPE) in its commonly used form when generalized in an *ad hoc* manner to time-dependent forces. A modified FFPE (MFFPE) is rigorously derived, being valid for a family of dichotomously alternating force fields. This MFFPE is numerically validated for a rectangular time-dependent force with zero average bias. For this case, subdiffusion is shown to become enhanced as compared to the force free case. We question, however, the existence of any physically valid FFPE for arbitrary varying time-dependent fields that differ from this dichotomous varying family.

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Normal Brownian motion occurring on potential landscapes that vary in time is known to exhibit a multifaceted collection of interesting phenomena, such as Brownian motors, anomalous nonlinear response behaviors, and stochastic resonance [1], to name a few. Therefore, it is tempting to ask whether an explicit time-dependent force entails a similarly versatile scenario also in the case of anomalously slow relaxation processes, relevant in many systems, such as polymer chains, networks, proteins, glasses, and charge-carriers in semiconductors [2]. This issue is in fact contained already in the first works on the motion of charge-carriers in semiconductors [3,4] and has been the subject of some further investigations ever since, see, e.g., the works [5-8], but never really has attracted proper attention on its fundamental level. Ultraslow relaxation in time-dependent external potential-fields thus still constitutes a challenge that is far from trivial.

A widely used approach to study subdiffusive processes is based on the fractional Fokker-Planck equation (FFPE) [9,10],

$$\frac{\partial}{\partial t}P(x,t) = {}_{0}\hat{D}_{t}^{1-\alpha} \left[-\frac{\partial}{\partial x} \frac{F(x)}{\eta_{\alpha}} + \kappa_{\alpha} \frac{\partial^{2}}{\partial x^{2}} \right] P(x,t). \quad (1)$$

Here, F(x) is the force, η_{α} is the fractional friction coefficient, κ_{α} is the fractional free diffusion coefficient, and ${}_{0}\hat{D}_{t}^{1-\alpha}$ denotes the Riemann-Liouville fractional derivative,

$${}_{0}\hat{D}_{t}^{1-\alpha}\chi(t) = \frac{1}{\Gamma(\alpha)}\frac{\partial}{\partial t}\int_{0}^{t}dt'\frac{\chi(t')}{(t-t')^{1-\alpha}}.$$
 (2)

For time-independent forces, the FFPE (1) can be rigorously derived from continuous time random walk (CTRW) theory [9,10], which corresponds to the random walk, including the non-Markovian character of the process via a Mittag-Leffler residence time distribution (RTD) $\psi(\tau) \propto \tau^{-1-\alpha}$, $\tau \to \infty$ (0 < α < 1) [2,3,11].

With this Letter, we show that the FFPE (1) fails in the case of a time-dependent force F(x, t). Furthermore, we

argue that the FFPE (1), when generalized *ad hoc* to a time-dependent force, does not correspond to a physical sto-chastic process. This affects a still steadily growing body of current research [12] and implies that these so obtained results therein are physically defeasible.

In a different context, the study of a subdiffusive dynamics in the case of a purely time-dependent force F(t) has given rise to a fractional Fokker-Planck equation which differs from Eq. (1) [8]. In this Letter, we derive an equation of similar form for the class of dichotomously alternating force-fields $F(x, t) = F(x)\xi(t)$ with $\xi(t) = \pm 1$, varying in space and time. In the case of a Mittag-Leffler RTD, it reads,

$$\frac{\partial}{\partial t}P(x,t) = \left[-\frac{\partial}{\partial x} \frac{F(x,t)}{\eta_{\alpha}} + \kappa_{\alpha} \frac{\partial^2}{\partial x^2} \right]_0 \hat{D}_t^{1-\alpha} P(x,t). \quad (3)$$

Notably, for a time-independent force, i.e., F(x, t) = F(x), Eqs. (1) and (3) are equivalent because the fractional derivative acts on time t only. Below, we prove the modified FFPE (MFFPE) in Eq. (3) in terms of CTRW theory and additionally validate it via the comparison of analytical solutions for a rectangular time-varying periodic force $F(x,t) \equiv F(t) = \xi(t)F_0$ with the numerical simulations of the underlying CTRW. Different force-fields, such as $F(x,t) = \pm \sin(x)$, have also been successfully tested (the details will be presented in a longer follow up work). Our main point is, however, that the reasoning provided in proving (3) forces us to scrutinize the *physical validity* of MFFPE in (3) already beyond a dichotomous driving F(t), e.g., for a sinusoidal driving $\xi(t)$ used in Ref. [8].

As an interesting result, we also show that a symmetric dichotomous force with average zero bias enhances the diffusion in respect to the free case. Furthermore, it is found that for sufficiently slow driving, the effective fractional diffusion coefficient $\kappa_{\alpha}^{(\text{eff})}$ exhibits a maximum vs the fractional exponent α .

It is well known that neither a non-Markovian Fokker-Planck equation nor its solution with the initial condition $P(x, t) = \delta(x - x_0)$ can fully define the non-Markovian stochastic process [13]. This is due to the fact that all non-Markovian processes, such as a CTRW with a nonexponential waiting time distribution, lack the factorization property, which would allow to express all the higherorder (multievent) probability density functions in terms of the first two. In order to generalize the physics of the FFPE (1) to time-dependent forces, we again start out from the underlying CTRW theory [9,10]. However, the usual scheme of merely replacing a time-independent force F in Eq. (1) in an ad hoc manner with a time-dependent F(t)is doomed to failure. The reason is that the underlying CTRW possesses a RTD with an infinite mean. Thus, any regular driving with a large but finite period is nonadiabatic. This very circumstance lies at the heart of the overall failure of Eq. (1) for time-dependent force fields.

In terms of a renewal description, a CTRW is a semi-Markovian process, meaning that the sojourn times spent on the localization sites are independently distributed. Let us consider a one-dimensional CTRW on a lattice $x_i = i\Delta x$ $(i = 0, \pm 1, \pm 2, ...)$. After a time τ drawn from the RTD $\psi_i(\tau)$, the particle at site i jumps with the probability q_i^{\pm} to one of the nearest neighbor sites. The external force field F(x) specifies both $\psi_i(\tau)$ and q_i^{\pm} , see Ref. [10]. Modulating the force F(x) in time, q_i^{\pm} obviously assumes a time dependence, and $\psi_i^{\pm}(\tau|t) = q_i^{\pm}(t+\tau)\psi_i(t+\tau,t)$ becomes conditioned on the entrance time t for the site i [14]. For a Markovian CTRW with time-dependent rates $w_i^{\pm}(t)$, it is known that

$$\psi_i(t+\tau,t) = w_i(t+\tau) \exp\left[-\int_t^{t+\tau} w_i(t')dt'\right], \quad (4)$$

with $w_i(t) = w_i^+(t) + w_i^-(t)$ and $q_i^{\pm}(t) = w_i^{\pm}(t)/w_i(t)$. For a driven non-Markovian CTRW, however, a relation similar to Eq. (4) is lacking. As a result, the use of a FFPE when generalized to the time-dependent case of a time-varying force-field remains moot. The usual scheme of the derivation of the generalized FFPE from the underlying CTRW can be used only if $\psi_i(\tau)$ remains unmodified by the timedependent fields, i.e., if only the jump probabilities $q_i^{\pm}(t)$

change. This yields $\psi_i^{\pm}(\tau|t) = q_i^{\pm}(t+\tau)\psi_i(\tau)$. As a matter of fact, the RTD $\psi_i(\tau)$ remains unaffected only in the case of a dichotomous flashing force $F(x, t) = F(x)\xi(t)$, where $\xi(t) = \pm 1$ is a general dichotomic function of time t which can change periodically or also stochastically. $q_i^{\pm}(t) = \exp[F(x_i)\xi(t)\Delta x/2]/\{\exp[F(x_i)\Delta x/2] +$ $\exp[-F(x_i)\Delta x/2]$. We assume that F(x) is continuous. Then, the MFFPE (3) can be derived rigorously in the continuous space limit. The derivation precisely follows the same reasoning as detailed in Ref. [10], while taking $\psi_i(\tau)$ as being the Mittag-Leffler distribution. It must be emphasized that for other driving forms $\xi(t)$, e.g., for a sinusoidal driving $F_0 \sin(\omega t)$, this outlined derivation becomes flawed because $\psi_i(\tau)$ is affected by such timevarying fields, as being unveiled already with Eq. (4). This in turn singles out the dichotomous force variation. Moreover, due to the weak ergodicity breaking [15,16], this MFFPE (3) describes the dynamics of an ensemble of particles rather than the dynamics of an individual particle.

Notably, Eq. (1) applied to the case of a time-dependent force may well define an interesting mathematical object in its own right; its connection to a known physical process, however, remains open to question. One may attempt to justify Eq. (1) for a time-inhomogeneous situation by appealing to the concept of "subordination," noting that this equation corresponds to a random process described by a usual Langevin equation but with the operational time being a random stochastic process [17,18]. A timedependent physical force, however, physically varies in deterministic, real time, which cannot be transformed to random time.

In the following, we study the particular case of a periodic rectangular driving force $F(t) = F_0(-1)^{\lfloor 2t/\tau_0 \rfloor}$, where τ_0 denotes the time period and [a] is the integer part of a. Put differently, we consider a dichotomous modulation of a biased free subdiffusion where the absolute value of the bias is fixed, but the direction of the force flips periodically in time. The average bias is zero. Let us begin by finding the recurrence relation for the moments $\langle x^n(t)\rangle$. Multiplying both sides of Eq. (3) by x^n and integrating over the x-coordinate, one obtains,

$$\frac{d\langle x^n(t)\rangle}{dt} = nF(t)D_t^{1-\alpha}\langle x^{n-1}(t)\rangle/\eta_\alpha + n(n-1)\kappa_\alpha D_t^{1-\alpha}\langle x^{n-2}(t)\rangle,\tag{5}$$

for $n \ge 2$. When n = 1, the last term on the right-hand side of the latter equation is absent. Then,

$$\frac{d\langle x(t)\rangle}{dt} = \frac{F(t)}{\eta_{\alpha}} D_t^{1-\alpha} 1 = \frac{F(t)}{\eta_{\alpha} \Gamma(\alpha)} t^{\alpha-1}.$$
 (6)

Integrating Eq. (6) in time, the solution for $\langle x(t) \rangle$ reads

$$\langle x(t) \rangle = \begin{cases} x_N + \frac{v_\alpha t^\alpha}{\Gamma(\alpha+1)}, & N\tau_0 \le t < (N + \frac{1}{2})\tau_0, \\ x_N' - \frac{v_\alpha t^\alpha}{\Gamma(\alpha+1)}, & (N + \frac{1}{2})\tau_0 \le t < (N+1)\tau_0, \end{cases}$$

$$<(N+1) au_0,$$

where

$$x_N = \langle x(0) \rangle - \frac{\nu_{\alpha} (N\tau_0)^{\alpha}}{\Gamma(\alpha+1)} + \frac{\nu_{\alpha} \tau_0^{\alpha}}{\Gamma(\alpha+1)}$$
$$\times \sum_{n=0}^{N-1} [2(n+1/2)^{\alpha} - n^{\alpha} - (n+1)^{\alpha}], \quad (8)$$

$$x_N' = x_N + \frac{2\nu_\alpha \tau_0^\alpha}{\Gamma(\alpha + 1)} (N + 1/2)^\alpha. \tag{9}$$

Here, $v_{\alpha} = F_0/\eta_{\alpha}$ and N counts the number of time periods passed. The analytical solution (7) for the mean particle position $\langle x(t) \rangle$ from the MFFPE (3) is compared with the numerical solution of the CTRW in Fig. 1 for different values of the fractional exponent α . The good agreement between our analytical and numerical results confirms that Eq. (3) is a correct method to describe the CTRW driven by a rectangular time-dependent force. Furthermore, the results depicted in Fig. 1 exhibit the phenomenon of the "death of linear response" of the fractional kinetics to time-dependent fields in the limit $t \rightarrow \infty$, reported also in Refs. [6,8]; i.e., in the long-time limit, the mean particle position approaches a constant value, rather than being oscillatory, i.e.,

$$\langle x(\infty) \rangle = v_{\alpha} \tau_0^{\alpha} b(\alpha) / \Gamma(\alpha + 1),$$
 (10)

where $b(\alpha) = \sum_{n=0}^{\infty} [2(n+1/2)^{\alpha} - n^{\alpha} - (n+1)^{\alpha}]$, with the amplitude of the oscillations decaying to zero as $1/t^{1-\alpha}$, see Eq. (6). The function $b(\alpha)$ describes the initial field phase effect which the system remembers forever when $\alpha < 1$. It changes monotonously from b(0) = 1 to b(1) = 0. The averaged traveled distance $\langle x(\infty) \rangle$ scales as $\tau_0^{\alpha} = (2\pi/\Omega)^{\alpha}$, where Ω is the corresponding angular frequency. This "death of linear response" to timeperiodic fields is also in agreement with the results for a driven non-Markovian two state system [19] in the formal limit of infinite mean residence times. Notably, this feature is overcome when one introduces a cutoff of the RTD's at long times (yielding a finite first moment), as used already in the pioneering work [4]. In this case, the subdiffusive behavior emerges as a transient, crossover behavior to asymptotically normal diffusion. Then, linear response theory based on the fluctuation-dissipation theorem becomes applicable [4].

We next study the mean square displacement and the effective fractional diffusion coefficient $\kappa_{\alpha}^{(\mathrm{eff})}$. We recall that the free fractional diffusion is described by $\langle \delta x^2(t) \rangle = 2\kappa_{\alpha}t^{\alpha}/\Gamma(1+\alpha) \propto t^{\alpha}$, while in the presence of a constant bias, surprisingly, $\langle \delta x^2(t) \rangle \propto t^{2\alpha}$ [3,11]. Strikingly enough, the same difference in the behavior remains true for a CTRW proceeding in a periodic potential with zero bias for which $\langle \delta x^2(t) \rangle \propto t^{\alpha}$ [20] and in a washboard potential with finite bias for which $\langle \delta x^2(t) \rangle \propto t^{2\alpha}$ [10,16], described by the FFPE. The question thus arises, whether a time-modulated subdiffusion follows the biased fractional sub-

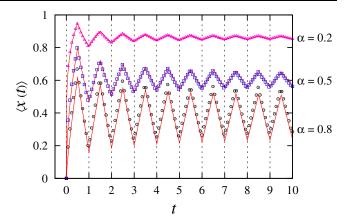


FIG. 1 (color online). Average particle position $\langle x(t) \rangle$ for various values of the fractional exponent α : Symbols represent the numerical results for the CTRW obtained by averaging over 10^6 trajectories, while continuous lines represent the analytical solution (7) of the MFFPE (3). The time period of the force is $\tau_0 = 1$, and $F_0/(\eta_\alpha \sqrt{\kappa_\alpha}) = 1$ is used in numerical simulations. The simulation algorithm is described in Ref. [16].

diffusion behavior $\propto t^{2\alpha}$, or rather assumes the unbiased behavior $\propto t^{\alpha}$, as the average bias is zero. To obtain the answer, we use the Laplace-transform method and the Fourier series expansion for the driving force $F(t+\tau_0)=F(t)$ with frequency $\Omega=2\pi/\tau_0$,

$$F(t) = \sum_{n = -\infty}^{\infty} f_n e^{in\Omega t}, \qquad f_{-n} = f_n^*$$
 (11)

with $f_{2n} = 0$ and $f_{2n+1} = -(2i/\pi)F_0/(2n+1)$ for the rectangular driving under consideration. We assume that $\langle x(0) \rangle = 0$ and $\langle x^2(0) \rangle = 0$ and denote the Laplace-transforms of the first and second moment by $\tilde{x}(s)$ and $\tilde{y}(s)$, yielding

$$s\tilde{x}(s) = \sum_{n=-\infty}^{\infty} \nu_n (s - in\Omega)^{-\alpha}, \tag{12}$$

where $v_n = f_n/\eta_\alpha$. Because $\langle x(\infty) \rangle$ is finite, we evaluate the asymptotical behavior of $\langle x^2(t) \rangle$ rather than $\langle \delta x^2(t) \rangle$. Because the Laplace transform of $D_t^{1-\alpha} \langle x(t) \rangle$ is $s^{-\alpha} s \tilde{x}(s)$, the Laplace transform of the second moment reads

$$s\tilde{y}(s) = 2\kappa_{\alpha}/s^{\alpha} + 2\sum_{m=-\infty}^{\infty} \nu_{m}(s - im\Omega)^{-\alpha} \sum_{n=-\infty}^{\infty} \nu_{n}[s - i(m+n)\Omega]^{-\alpha}.$$
(13)

Note that in the double sum, only the terms with m = -n contribute to the effective subdiffusion coefficient, being averaged over the driving period. Therefore, we find that

$$\kappa_{\alpha}^{(\text{eff})} = \kappa_{\alpha} + 2 \frac{\cos(\pi \alpha/2)}{\Omega^{\alpha}} \sum_{n=1}^{\infty} \frac{|\nu_{n}|^{2}}{n^{\alpha}}$$

$$= \kappa_{\alpha} + g(\alpha)F_{0}^{2}/(\eta_{\alpha}^{2}\Omega^{\alpha}), \tag{14}$$

where

$$g(\alpha) = (2/\pi^2)\zeta(2+\alpha)[4-2^{-\alpha}]\cos(\pi\alpha/2)$$
 (15)

is a function decaying from g(0)=1 towards g(1)=0 and $\zeta(x)$ is the Riemann's zeta-function. From Eq. (13), one finds that the asymptotic behavior of the mean square displacement is proportional to t^{α} as in the force free case. It is characterized by an effective fractional diffusion coefficient $\kappa_{\alpha}^{(\mathrm{eff})}$ instead of the free value κ_{α} , i.e., $\langle \delta x^2(t) \rangle = 2\kappa_{\alpha}^{(\mathrm{eff})} t^{\alpha}/\Gamma(1+\alpha)$ for $t \to \infty$. The driving-induced part of the effective subdiffusion coefficient is

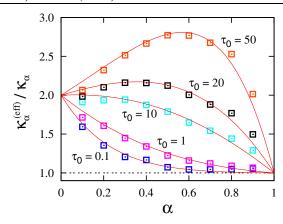


FIG. 2 (color online). Scaled effective fractional diffusion coefficient $\kappa_{\alpha}^{(\mathrm{eff})}$ versus fractional exponent α for different driving periods τ_0 . The analytical prediction (14) (continuous lines) is compared with the numerical results (symbols) obtained from the CTRW by averaging over 10^5 trajectories. For $\tau_0 > 2\pi \exp[-\frac{d}{d\alpha} \ln g(\alpha)|_{\alpha=0}] \approx 8.818$, the effective fractional diffusion coefficient $\kappa_{\alpha}^{(\mathrm{eff})}(\alpha)$ exhibits a maximum.

proportional to the square of driving amplitude and inversely proportional to Ω^{α} . For slowly oscillating force fields, this leads to a profound acceleration of subdiffusion as compared with the force free case: An optimal value of the fractional exponent α exists, at which the driving-induced part of the effective fractional diffusion coefficient possesses a maximum, cf. Figure 2.

In this Letter, we discussed the dynamics of anomalously slow processes in time-varying potential landscapes within the CTRW and FFPE descriptions. We demonstrated that the common form of the FFPE given by Eq. (1) is not valid for time-dependent forces; it fails to correspond to the underlying CTRW modulated by an external time-dependent force field. A modified form of the FFPE, Eq. (3), is derived for dichotomously alternating force fields. As an exactly solvable example, we studied a periodic rectangular force with zero average and successfully tested the analytical results via numerical simulations of the underlying time-modulated CTRW.

Our study, however, is not able to validate the correctness of the MFFPE (3) when extended ad hoc to an arbitrary time-dependent potential landscape different from the dichotomous case. A description of timedependent fields via subordination in conjunction with a CTRW approach is also doomed to failure because of the distinct difference between the deterministic physical time and the merely mathematical random subordination time. As a matter of fact, any slowly nonzero frequency timevarying force varies infinitely fast within the realm of fictitious, operational subordination time. This causes CTRW subdiffusion to fail in responding to time-periodic fields in an ordinary manner. In addition, all those theories modeling dielectric response which are based on such an approach are thus also physically defeasible. A way out of this dilemma consists in relying on models of driven subdiffusion which either are based on the generalized Langevin dynamics [21] or on fractal Brownian motion. The challenge of modeling subdiffusion in a time-varying potential landscape thus necessitates plenty of further enlightening research.

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- [1] P. Hänggi and F. Marchesoni, Chaos 15, 026101 (2005); P. Reimann and P. Hänggi, Appl. Phys. A: Mater. Sci. Process. 75, 169 (2002); R. D. Astumian and P. Hänggi, Phys. Today 55, No. 11, 33 (2002); M. Borromeo and F. Marchesoni, Chaos 15, 026110 (2005); L. Gammaitoni, P. Hänggi, P. Jung, and F. Marchesoni, Rev. Mod. Phys. 70, 223 (1998).
- [2] H. Scher, M. F. Shlesinger, and J. T. Bendler, Phys. Today 44, No. 1, 26 (1991); M. F. Shlesinger, G. M. Zaslavsky, and J. Klafter, Nature (London) 363, 31 (1993); J. Klafter, M. F. Shlesinger, and G. Zumofen, Phys. Today 49, No. 2, 33 (1996).
- [3] H. Scher and E. W. Montroll, Phys. Rev. B **12**, 2455 (1975).
- [4] H. Scher and M. Lax, Phys. Rev. B 7, 4491 (1973).
- [5] I. M. Sokolov, A. Blumen, and J. Klafter, Europhys. Lett. 56, 175 (2001); I. M. Sokolov, A. Blumen, and J. Klafter, Physica A (Amsterdam) 302, 268 (2001).
- [6] F. Barbi, M. Bologna, and P. Grigolini, Phys. Rev. Lett. 95, 220601 (2005).
- [7] I. M. Sokolov, Phys. Rev. E 73, 067102 (2006).
- [8] I. M. Sokolov and J. Klafter, Phys. Rev. Lett. 97, 140602 (2006).
- [9] R. Metzler and J. Klafter, Phys. Rep. 339, 1 (2000); R. Metzler, E. Barkai, and J. Klafter, Phys. Rev. Lett. 82, 3563 (1999); E. Barkai, Phys. Rev. E 63, 046118 (2001).
- [10] I. Goychuk, E. Heinsalu, M. Patriarca, G. Schmid, and P. Hänggi, Phys. Rev. E 73, 020101(R) (2006).
- [11] M. F. Shlesinger, J. Stat. Phys. **10**, 421 (1974).
- [12] See, e.g., in: J.-L. Dejardin and J. Jadzyn, J. Chem. Phys. 123, 174502 (2005); M. Y. Yim and K. L. Liu, Physica A (Amsterdam) 369, 329 (2006).
- [13] P. Hänggi and H. Thomas, Phys. Rep. 88, 207 (1982).
- [14] I. Goychuk and P. Hänggi, Phys. Rev. E 69, 021104 (2004).
- [15] G. Bel and E. Barkai, Phys. Rev. Lett. **94**, 240602 (2005).
- [16] E. Heinsalu, M. Patriarca, I. Goychuk, G. Schmid, and P. Hänggi, Phys. Rev. E 73, 046133 (2006).
- [17] A. A. Stanislavsky, Phys. Rev. E 67, 021111 (2003).
- [18] M. Magdziarz, A. Weron, and K. Weron, Phys. Rev. E **75**, 016708 (2007).
- [19] I. Goychuk and P. Hänggi, Phys. Rev. Lett. 91, 070601 (2003).
- [20] E. Heinsalu, M. Patriarca, I. Goychuk, and P. Hänggi, J. Phys. Condens. Matter 19, 065114 (2007).
- [21] H. Mori, Prog. Theor. Phys. 33, 423 (1965); R. Kubo, Rep. Prog. Phys. 29, 255 (1966).