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Fluctuation relations for anomalous dynamics generated by time-fractional Fokker–Planck equations

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Abstract

Anomalous dynamics characterized by non-Gaussian probability distributions (PDFs) and/or temporal long-range correlations can cause subtle modifications of conventional fluctuation relations (FRs). As prototypes we study three variants of a generic time-fractional Fokker–Planck equation with constant force. Type A generates superdiffusion, type B subdiffusion and type C both super- and subdiffusion depending on parameter variation. Furthermore type C obeys a fluctuation–dissipation relation whereas A and B do not. We calculate analytically the position PDFs for all three cases and explore numerically their strongly non-Gaussian shapes. While for type C we obtain the conventional transient work FR, type A and type B both yield deviations by featuring a coefficient that depends on time and by a nonlinear dependence on the work. We discuss possible applications of these types of dynamics and FRs to experiments.

1. Introduction

Understanding fluctuations far from equilibrium defines a key topic of non-equilibrium statistical physics. A new line of activities started about three decades ago by discovering different forms of fluctuation relations (FRs) which generalize fundamental laws of thermodynamics to small systems in non-equilibrium; see [1–8] for reviews and further references therein. More recently these laws got unified by over-arching schemes, most notably the deterministic dissipation function approach by Evans and coworkers [1], and by stochastic thermodynamics [7, 9–11]. The latter theory starts from defining entropy production on the level of individual trajectories in stochastic models such as Langevin and master equations. Given that stochastic thermodynamics is based on rather simple Markov models one may ask to which extent FRs derived from it are reproduced if the dynamics is more complicated. In our paper we address this problem by testing FRs for stochastic dynamics that is anomalous due to non-Markovian dynamical correlations and/or strongly non-Gaussian probability distributions (PDFs).

Anomalous dynamics has been observed in many experiments and is widely studied by the theory of anomalous stochastic processes [12–17]. A characteristic property of anomalous dynamics is that the mean square displacement (MSD) grows nonlinearly in time yielding anomalous diffusion in the long time limit [15]. In contrast, Markovian dynamics like Brownian motion generates a MSD that increases linearly for long times. If the MSD grows faster than linear one speaks of superdiffusion, if it grows slower than linear one obtains subdiffusion. There are many different ways to model anomalous stochastic dynamics such as continuous time random walks (CTRW) [12, 18–20], generalized Langevin equations [13, 21–23], Lévy flights and walks [17, 24], fractional diffusion equations [16], scaled Brownian motion [25, 26] and heterogeneous diffusion processes [27], to name a few.

The study of FRs for anomalous stochastic processes appears to be rather at the beginning: Crooks and Jarzynski work relations as well as transient and steady state fluctuation theorems have been confirmed for non-Markovian Gaussian dynamics modelled by generalized Langevin equations with memory kernels, given specific conditions are fulfilled [28–31]. These results have been reproduced and generalized by a stochastic thermodynamics approach [32]. For non-Gaussian PDFs generated by Langevin equations with non-Gaussian noise, such as Lévy noise or Poissonian shot noise, violations of conventional steady state and transient fluctuation relations (TFRs) have been reported [33–38]. For a CTRW model with a power law waiting time distribution it was found that the steady state FRs may or may not hold depending on the exponent of the waiting time distribution [39]. Computer simulations of glassy dynamics exhibiting anomalous diffusion also showed violations of TFRs [40, 41]. In [42–44] several of the above types of stochastic dynamics including fractional Fokker–Planck equations (FFPEs) were considered. It was found that the validity of fluctuation–dissipation relations [45] for a given anomalous stochastic process plays a crucial role for the validity or violation of conventional FRs.

In this article we test TFRs for a class of anomalous stochastic processes that so far has not been in the focus of investigations, which are time-FFPEs. Such equations model the emergence of non-Gaussian PDFs by using power law memory kernels via time-fractional derivatives [46]. They need to be distinguished from equations modeling correlations in space via space-fractional derivatives as they naturally arise, e.g., for generating Lévy flights [12, 42]. FFPE can be derived from stochastic equations of motion either by CTRWs [12, 16] or by subordinated Langevin dynamics [47]. Quite a variety of them have been studied in the literature, both from a purely theoretical point of view and with respect to applications to experiments: Prominent examples are fractional Klein–Kramers equations that were used to analyse biological cell migration data [48–50]. Another type was designed to model the dynamics of tracer particles in random environments [51]. Closely related time-fractional diffusion equations [12, 21, 52] have been used to model a variety of different processes, from diffusion in crowded cellular environments [15, 53] to geophysical and environmental systems [14]. They have also been derived for weakly chaotic dynamical systems [54, 55]. A bifractional diffusion equation famously reproduced the spreading of dollar bills in the United States [56].

Our paper is structured as follows: In section 2 we discuss three types of FFPEs which differ from each other in terms of their anomalous diffusive properties, and by whether or not they fulfill fluctuation–dissipation relations. We solve these models for their position PDFs and study their properties both analytically and numerically. In section 3 we test the (work) TFR for our three models by analytical asymptotic expansions and by numerically plotting the results. We conclude with a summary and an outlook towards physical applications in section 4.

2. Time-FFPEs

This section introduces to three different types of FFPEs: we first outline how starting from stochastic dynamics a FFPE generating superdiffusion can be constructed in the form of an overdamped Langevin equation with correlated noise. Our argument illustrates how a time-fractional derivative naturally emerges from modelling power law time correlation decay. The other two types of FFPEs that we consider have already been derived in the literature from CTRW theory and are either subdiffusive or exhibit a transition from sub- to superdiffusion under parameter variation. We analytically calculate the first and second moments for all three models, which enables us to check for the validity of the fluctuation–dissipation relation of the first kind (FDR1). We also comment on the Galilean invariance of our models. We then analytically calculate the position PDFs of all FFPEs and study the solutions numerically by plotting the results.

2.1. Constructing a superdiffusive FFPE
The study of an overdamped Langevin equation for the position $x(t)$ of a particle on the line driven by a correlated stochastic process and an external force allows to gain insight into the origin of a superdiffusive FFPE. Our Langevin equation of interest is given by

$$\frac{\mathrm{d}x}{\mathrm{d}t} = \frac{F_0}{m\gamma_\alpha} + v(t), \tag{1}$$

where F_0 denotes a constant external force, γ_α a friction coefficient and m the mass of the particle. We assume that $v(t)$ is a stationary correlated stochastic process with zero mean $\langle v(t) \rangle = 0$ and a power–law correlation function

$$\left\langle v(t) \, v\left(t_1\right)\right\rangle = \frac{K_\alpha}{\Gamma(\alpha-1)} \frac{1}{|t-t_1|^{2-\alpha}} \tag{2}$$

with $1 < \alpha < 2$, gamma function Γ and generalized diffusion coefficient K_α. Note that we do not further specify the noise. Following the pseudo-Liouville hybrid approach of Balescu [57, 58] (see appendix A) one obtains the following exact result in equation (A.8) for the PDF $f(x, t)$:

$$\left(\frac{\partial}{\partial t} + v_0 \frac{\partial}{\partial x}\right) f(x, t) = \frac{\partial^2}{\partial x^2} \int_0^t dt_1 \left\langle v(t) v\left(t_1\right) f\left(x - \Delta\left(t, t_1\right), t_1\right)\right\rangle \tag{3}$$

with $v_0 = F_0/(m\gamma_\alpha)$ and $\Delta(t, t_1) = v_0(t - t_1) + \int_{t_1}^t dt_2 \, v(t_2)$. This exact equation is non-local in time (i.e. non-Markovian) and non-local in space. We now make a local-in-space approximation by neglecting the term of the fluctuating displacement $\Delta(t, t_1)$ on the right hand side of the probability density f. Such an approximation seems to be reasonable in the long time and large space asymptotic limit if the drift and velocity fluctuations are weak enough. This assumption results in the following non-Markovian Fokker–Planck equation:

$$\left(\frac{\partial}{\partial t} + v_0 \frac{\partial}{\partial x}\right) f(x, t) = \frac{\partial^2}{\partial x^2} \int_0^t dt_1 \left\langle v(t) v\left(t_1\right)\right\rangle f\left(x, t_1\right). \tag{4}$$

Insertion of the correlation function of velocities equation (2) into equation (4) leads to

$$\left(\frac{\partial}{\partial t} + v_0 \frac{\partial}{\partial x}\right) f(x, t) = \frac{\partial^2}{\partial x^2} \frac{K_\alpha}{\Gamma(\alpha-1)} \int_0^t dt_1 \left(t - t_1\right)^{\alpha-2} f\left(x, t_1\right). \tag{5}$$

The integral on the right hand side matches to the Riemann–Liouville (RL) fractional integral of order μ given by [59]

$$J_t^\mu \, g(t) \equiv D_t^{-\mu} g(t) = \frac{1}{\Gamma(\mu)} \int_0^t d\tau \, (t - \tau)^{\mu-1} g(\tau) \tag{6}$$

with $\mu > 0$ and $\mu = \alpha - 1$ for equation (5). We also introduce the definition of the RL fractional derivative of positive order

$$D_t^\mu \, g(t) = \frac{d^n}{dt^n} J_t^{n-\mu} \, g(t) \tag{7}$$

with $\mu > 0$, $n = [\mu] + 1$, where $[\ldots]$ refers to the integer part of the given number. Applying equations (6) to (5) gives us our first type of FFPE that we denote as

$$\text{type A:} \quad \frac{\partial f_A(x, t)}{\partial t} = -\frac{\partial}{\partial x}\left[v_0 - K_\alpha D_t^{1-\alpha} \frac{\partial}{\partial x}\right] f_A(x, t), \, 1 < \alpha < 2. \tag{8}$$

To show the relation of this equation with previous works we put $v_0 = 0$. Then it can be written as

$$\frac{\partial^2}{\partial t^2} f(x, t) = K_\alpha \frac{\partial^2}{\partial x^2} D_t^{2-\alpha} f(x, t), \, 1 < \alpha < 2. \tag{9}$$

This equation was called a fractional wave equation in the seminal paper of Schneider and Wyss [52] and has also been derived for a long-range correlated dichotomous stochastic process [60] from a fractional Klein–Kramers equation [48] and from a generalized Chapman–Kolmogorov equation [61]. The solution of this equation has been studied in detail in [62] where it was called a fractional kinetic equation for sub-ballistic superdiffusion. The equivalent form of this equation using the Caputo fractional derivative was investigated in [63].

Our presentation above illustrates how a FFPE can be derived from a Langevin equation with power–law decay in the velocity correlation function. It furthermore demonstrates that a fractional derivative provides the natural mathematical formulation to model equations containing power law memory kernels.

2.2. Definition and properties of FFPEs

In addition to type A FFPE equation (8) we consider two further types of FFPEs. Both have been derived from CTRW theory [12, 18–20]. Note that the underlying stochastic dynamics and the derivation of these two FFPEs are very different from what we presented for type A above. Indeed, both type B and type C are essentially (almost) Markovian models, in contrast to type A. Our two new FFPEs describe subdiffusion under the influence of a constant external force and naturally appear in physical systems where diffusion is slowed down by deep traps [12, 20, 64]. The difference between these two types arises from the position of the fractional RL derivative with respect to the diffusive and drift part of the equations and the range of the anomaly parameter α. Our second FFPE is defined as

$$\text{type B:} \quad \frac{\partial f_B(x, t)}{\partial t} = -\frac{\partial}{\partial x}\left[v_0 - K_\alpha D_t^{1-\alpha}\frac{\partial}{\partial x}\right]f_B(x, t). \tag{10}$$

For type C FFPE the RL fractional derivative is also included in the drift term:

$$\text{type C:} \quad \frac{\partial f_C(x, t)}{\partial t} = -\frac{\partial}{\partial x}\left[A_\alpha v_0 D_t^{1-\alpha} - K_\alpha D_t^{1-\alpha}\frac{\partial}{\partial x}\right]f_C(x, t), \tag{11}$$

where A_α has a dimension of time to the power of $1 - \alpha$. Note that type B and type C FFPEs are defined for $0 < \alpha < 1$ whereas for type A FFPE α is in the range $1 < \alpha < 2$. For all three FFPEs we use the initial condition $f_{A,B,C}(x, t = 0) = \delta(x)$. By means of Fourier and Laplace transforms

$$\hat{f}(k) = \int_{-\infty}^\infty \mathrm{d}x e^{ikx}f(x), \qquad \tilde{f}(s) = \int_0^\infty \mathrm{d}t e^{-st}f(t) \tag{12}$$

a solution of equations (8), (10) and (11) can be obtained in Fourier-Laplace space as

$$\hat{\tilde{f}}_{A,B}(k, s) = \frac{1}{s + v_0 ik + K_\alpha k^2 s^{1-\alpha}}, \tag{13}$$

$$\hat{\tilde{f}}_C(k, s) = \frac{1}{s + A_\alpha v_0 iks^{1-\alpha} + K_\alpha k^2 s^{1-\alpha}}, \tag{14}$$

where the fractional derivative $D_t^{1-\alpha}f(t)$ transforms to $s^{1-\alpha}\tilde{f}(s)$. The solutions of type A and type B FFPE only differ in the range of α as defined above. The representation in Fourier-Laplace space allows the calculation of moments by differentiation with respect to k:

$$\langle x^n(t) \rangle = \mathcal{L}^{-1}\left\{ (i)^n \left.\frac{\partial^n \hat{\tilde{f}}(k, s)}{\partial k^n}\right|_{k=0} \right\}. \tag{15}$$

After Laplace inversion one obtains the first two moments and the central second moment for $\delta x = x - \langle x \rangle$ of type C FFPE defined in equation (11) [20]

$$\langle x \rangle_C = \frac{A_\alpha v_0 t^\alpha}{\Gamma(\alpha + 1)}, \tag{16}$$

$$\langle x^2 \rangle_C = \frac{2K_\alpha t^\alpha}{\Gamma(\alpha + 1)} + \frac{2A_\alpha^2 v_0^2 t^{2\alpha}}{\Gamma(2\alpha + 1)}, \tag{17}$$

$$\langle (\delta x)^2 \rangle_C = \frac{2K_\alpha t^\alpha}{\Gamma(\alpha + 1)} + A_\alpha^2 v_0^2 t^{2\alpha}\left[\frac{2}{\Gamma(2\alpha + 1)} - \frac{1}{\Gamma(\alpha + 1)^2}\right]. \tag{18}$$

These results show that the FDR1 [43, 45] $\langle x(t) \rangle_C \sim \langle x^2(t) \rangle_C^{v_0=0}$ is valid for type C. Interestingly the external force influences the second central moment $\sim v_0^2 t^{2\alpha}$. Technically this is due to the coupling term $v_0 iks^{1-\alpha}$ in the Laplace-Fourier representation of equation (14). The first moment increases sublinearly despite the constant external force. This can be interpreted as a partial sticking effect of particles [65]. By contrast, the second central moment shows a crossover from $\sim t^\alpha$ to $\sim t^{2\alpha}$. Thus, for $v_0 \neq 0$ type C switches from a subdiffusive behavior of the second central moment for $0 < \alpha < 1/2$ to a superdiffusive behavior for $1/2 < \alpha < 1$ [12].

Analogously, the moments of type A and type B FFPEs of equations (8) and (10) are obtained as [20]

$$\langle x \rangle_{A,B} = v_0 t, \tag{19}$$

$$\langle x^2 \rangle_{A,B} = \frac{2K_\alpha t^\alpha}{\Gamma(\alpha + 1)} + v_0^2 t^2, \tag{20}$$

$$\langle (\delta x)^2 \rangle_{A,B} = \frac{2K_\alpha t^\alpha}{\Gamma(\alpha + 1)}. \tag{21}$$

In both cases the first moment only depends on v_0 and increases linearly in time. The second central moment shows a superdiffusive and subdiffusive increase $\sim t^\alpha$ for type A and type B FFPE, respectively. In contrast to type C FFPE, the second moment of type A and type B FFPEs is without any coupling to v_0. In addition, type A and type B FFPEs break FDR1 between the first $\langle x(t) \rangle_{A,B}$ and the second moment $\langle x^2(t) \rangle_{A,B}^{v_0=0}$. In both cases this is what one should expect according to the definition of both models: Type A is based on the Langevin equation (1) where the fluctuation–dissipation relation of the second kind (FDR2) is broken by construction. Note that FDR2 establishes a relation between the noise and the friction [45]. The breaking of FDR2 suggests a breaking of FDR1 as was shown for Gaussian stochastic processes in [43]. For type B the fractional derivative acts only on the

diffusion term in equation (10) thus breaking FDR1 while for type C it acts simultaneously on both the drift and the diffusion terms in equation (11) hence preserving FDR1.

A second difference between these FFPEs consists in their behavior under Galilean transformation. With $X = x - v_0 t$ and $T = t$ the PDF $f(x, t)$ is transformed to $\Omega(X, T)$. The coupling of the fractional RL derivative to the v_0 drift term of type C FFPE in equation (11) breaks Galilean invariance. However, type A and B FFPE of equations (8) and (10) fulfill Galilean invariance in the long time and large space limit [12, 20, 66], where they can be written as

$$\frac{\partial \Omega_{A,B}(X, T)}{\partial T} = K_\alpha D_T^{1-\alpha} \frac{\partial^2 \Omega_{A,B}(X, T)}{\partial X^2}. \tag{22}$$

This means that in this limit breaking or preserving FDR1 corresponds to preserving respectively breaking Galilean invariance in the case of these FFPEs. This property will be exploited in the next subsection where we discuss analytical and numerical solutions of our three types of FFPEs.

2.3. Analytical solution of time-FFPEs

Type C FFPE: Fourier inversion [42] leads to the solution of type C FFPE in (x, s) space:

$$\tilde{f}_C(x, s) = \frac{s^{\alpha-1}}{\sqrt{A_\alpha^2 v_0^2 + 4K_\alpha s^\alpha}} \exp\left(\frac{A_\alpha v_0 x}{2K_\alpha} - |x| \frac{\sqrt{A_\alpha^2 v_0^2 + 4K_\alpha s^\alpha}}{2K_\alpha}\right). \tag{23}$$

In this case, a solution in (x, t) space can be given as a superposition of the $\alpha = 1$ Gaussian solution with a Lévy kernel [12, 67]. However, for numerical analysis we apply a direct numerical Laplace inversion of equation (23).

Type A and B FFPE: Analogously to equation (23) the solutions of type A and type B FFPEs can be calculated in (x, s) space with $A_\alpha v_0 \to v_0 s^{\alpha-1}$ to

$$\tilde{f}_{A,B}(x, s) = \frac{s^{\alpha-1}}{\sqrt{v_0^2 \, s^{2\alpha-2} + 4K_\alpha s^\alpha}} \exp\left(\frac{v_0 s^{\alpha-1} x}{2K_\alpha} - |x| \frac{\sqrt{v_0^2 \, s^{2\alpha-2} + 4K_\alpha s^\alpha}}{2K_\alpha}\right). \tag{24}$$

As the FFPEs of type A and type B are Galilean invariant in the long time and large space limit, the solution for $v_0 = 0$ allows the exact calculation of the PDFs with drift v_0 in this limit [12, 20], which becomes approximate otherwise [66]. The solution to equation (22) is well-known [12] and is given using a Fox H-function (see Appendix B for definitions). Thus, applying Galilean transformation and replacing x with $x - v_0 t$ gives solutions of type A and type B FFPEs in (x, t) space as

$$f_{A,B}(x, t) = \frac{1}{\sqrt{4K_\alpha t^\alpha}} H_{11}^{10}\left[\frac{|x - v_0 t|}{\sqrt{K_\alpha t^\alpha}} \middle| \begin{array}{c} (1 - \alpha/2, \alpha/2) \\ (0, 1) \end{array}\right]. \tag{25}$$

These approximate solutions in terms of shifted Fox functions are the basis for our further analysis of type A and B FFPEs.

2.4. Numerical analysis of time-FFPEs

Numerical methods are required to study the analytical results given in form of Fox H-functions of type A and type B FFPE and in Laplace space for type C FFPE.
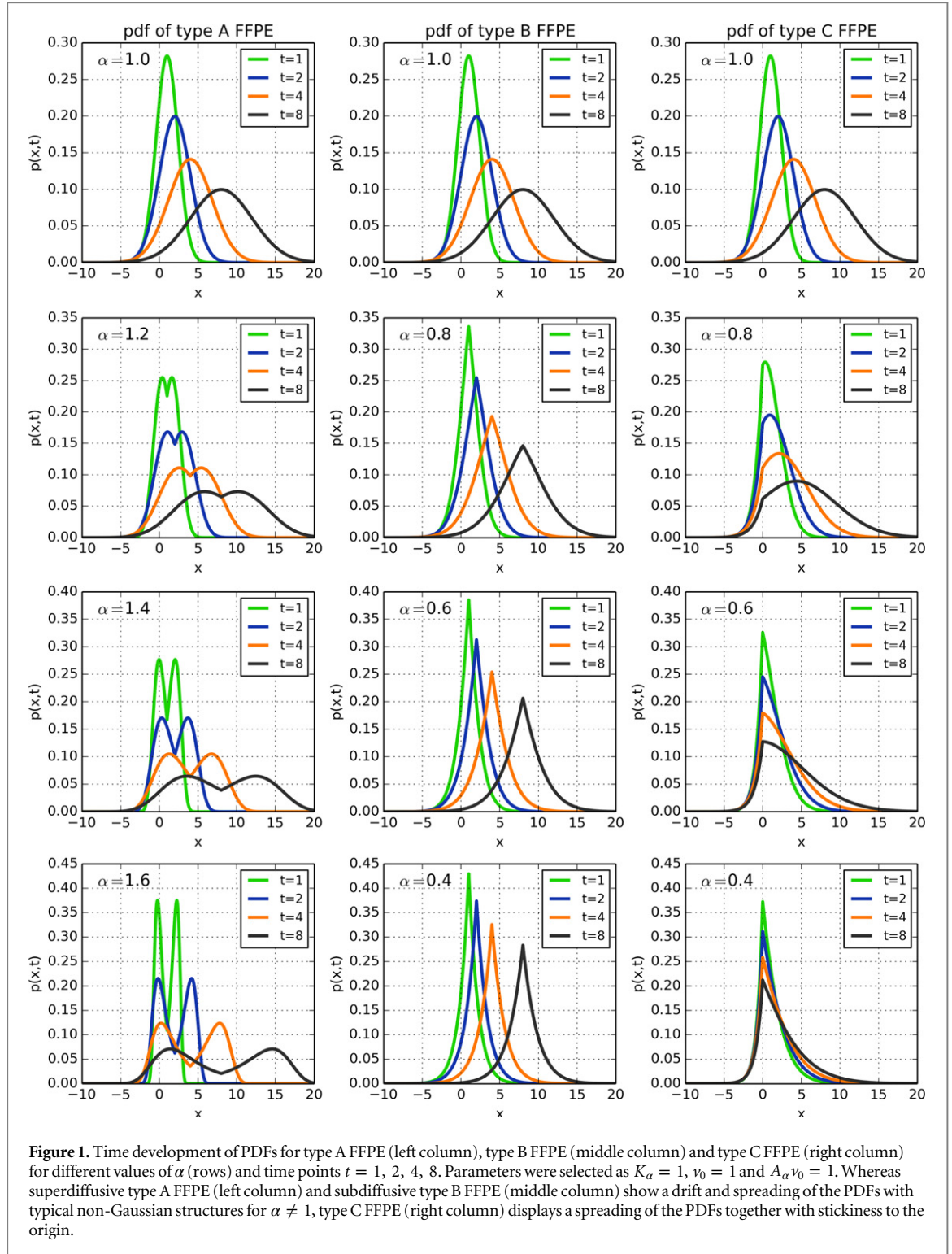
Type A and type B FFPE: the series expansion of the solution $f_{A,B}(x, t)$ of equation (25) as given by equation (B.3) is used for numerical evaluations,

$$f_{A,B}(x, t) = \frac{1}{\sqrt{4K_\alpha t^\alpha}} \sum_{j=0}^{\infty} \frac{(-1)^j}{j!\,\Gamma(1 - \alpha(j + 1)/2)} \left(\frac{(x - v_0 t)^2}{K_\alpha t^\alpha}\right)^{j/2} \tag{26}$$

with $1 < \alpha < 2$ for type A FFPE and $0 < \alpha < 1$ for type B FFPE. The series is evaluated with multiple-precision arithmetic.

Type C FFPE: direct numerical Laplace inversion is applied to equation (23) to obtain the probability density function $f_C(x, t)$. Here we use a multiple-precision algorithm for the Laplace inversion based on Talbot's method [68, 69].

Typical behavior in space and time: figure 1 shows the time development of the solutions $f(x, t)$ of the three FFPE types for different times t = 1, 2, 4, 8. Parameters were selected as $A_\alpha v_0 = 1$ and $K_\alpha = 1$, the anomaly index α was chosen from $\alpha \in (0.4, 0.6 \ldots 1.6)$. The first row shows the Gaussian limit $\alpha \to 1$ for all three types. In this normal diffusive case the PDF is spreading with $\sqrt{2K_1 t}$ and its center is moving according to $v_0 t$. The PDFs of type A (left column) and type B FFPE (middle column) preserve this constant drift for $\alpha \neq 1$. However, the shapes of the PDFs of both models immediately change profoundly showing characteristically different types of

Figure 1. Time development of PDFs for type A FFPE (left column), type B FFPE (middle column) and type C FFPE (right column) for different values of α (rows) and time points $t = 1, 2, 4, 8$. Parameters were selected as $K_\alpha = 1$, $v_0 = 1$ and $A_\alpha v_0 = 1$. Whereas superdiffusive type A FFPE (left column) and subdiffusive type B FFPE (middle column) show a drift and spreading of the PDFs with typical non-Gaussian structures for $\alpha \neq 1$, type C FFPE (right column) displays a spreading of the PDFs together with stickiness to the origin.

non-Gaussian behavior: For type A the PDFs spread superdiffusively with the variance of equation (21) by exhibiting a double-peaked structure with a dip in the middle. Qualitatively, the highly characteristic double-peak structure is explained in [58]: The propagator of type A decays asymptotically *faster* than the Gaussian, see equation (B.5). However, since two maxima move away from the origin in the opposite directions, superdiffusion is possible in spite of the thin tail of the propagator; see also equation (9) [62]. Note that there are cusp singularities in all three models for $\alpha \neq 1$, in contrast to the smooth behavior of the Gaussian PDF shown in the top row. In the Galilean invariant cases A and B the propagators are symmetric with respect to their cusps, which are translated with velocity $v_0 = 1$, as it should be. For the Galilean non-invariant model C the propagator is asymmetric with respect to its cusp, which stays fixed at the origin [12].

3. Work FRs for FFPEs

3.1. Definition of FRs

Using the results of the previous section, we now study the PDF $p(W, t)$ of the mechanical work $W = -F_0 x$ generated by the constant external field F_0. For a constant field the PDF of work $p(W, t)$ is related to the PDF $f(x, t)$ of positions by the simple scaling transformation

$$p(W, t) = \frac{1}{F_0} f\left(\frac{W}{F_0}, t\right). \tag{27}$$

It is the main aim of this work to study the TFR of the work PDFs defined by the logarithmic fluctuation ratio

$$\mathsf{R}(W, t) := \log \frac{p(W, t)}{p(-W, t)} \tag{28}$$

for the three types of FFPEs. All three FFPE types reduce to a normal Gaussian process with drift for $\alpha \to 1$. For a Gaussian PDF the ratio R is trivially given by the ratio of the first and second central moment, i.e. $2\langle W\rangle/\langle\delta W^2\rangle$ [43]. Thus one obtains a normal or conventional FR for $\alpha = 1$,

$$\mathsf{R}(W, t)\big|_{\alpha=1} = \frac{v_0}{F_0 K_1} W = W/\left(k_B T\right). \tag{29}$$

with a linear increase in W that is independent of time as it has been found for a large class of systems [1–3, 6–8]. The last expression has been obtained by using the Einstein relation $K_\alpha = k_B T/(m\gamma_\alpha)$ with temperature T, Boltzmann constant k_B and the definition $v_0 = F_0/(m\gamma_\alpha)$. The general case for $\alpha \neq 1$ is studied in the next section.

3.2. FRs for FFPEs

Type C FFPE: for this type the fluctuation ratio can be studied analytically [42]. With equation (23) $\mathsf{R}(W, t)$ is given in Laplace space by

$$\frac{\tilde{p}_C(W, s)}{\tilde{p}_C(-W, s)} = \frac{\tilde{f}_C\left(W/F_0, s\right)}{\tilde{f}_C\left(-W/F_0, s\right)} = \exp\left(\frac{A_\alpha v_0}{F_0 K_\alpha} W\right). \tag{30}$$

As the right side is independent of the Laplace variable s, the Laplace inverse of the PDFs can be calculated directly after multiplication with $\tilde{p}_C(-W, s)$. Thus, despite the complicated form of the PDFs a linear normal TFR is obtained for type C FFPE:

$$\log \frac{p_C(W, t)}{p_C(-W, t)} = \frac{A_\alpha v_0}{F_0 K_\alpha} W. \tag{31}$$

This result based on the Laplace transformed ratio of $\tilde{p}_C(W, s)$ seems to be surprising with respect to the complex form of the PDF in Laplace space and the asymmetric sticking behavior at the origin of the PDFs as illustrated in the right column of figure 1. The right side of figure 2 shows the numerical calculation of the fluctuation ratio which is linear and constant for all times in agreement with the given analytical result.
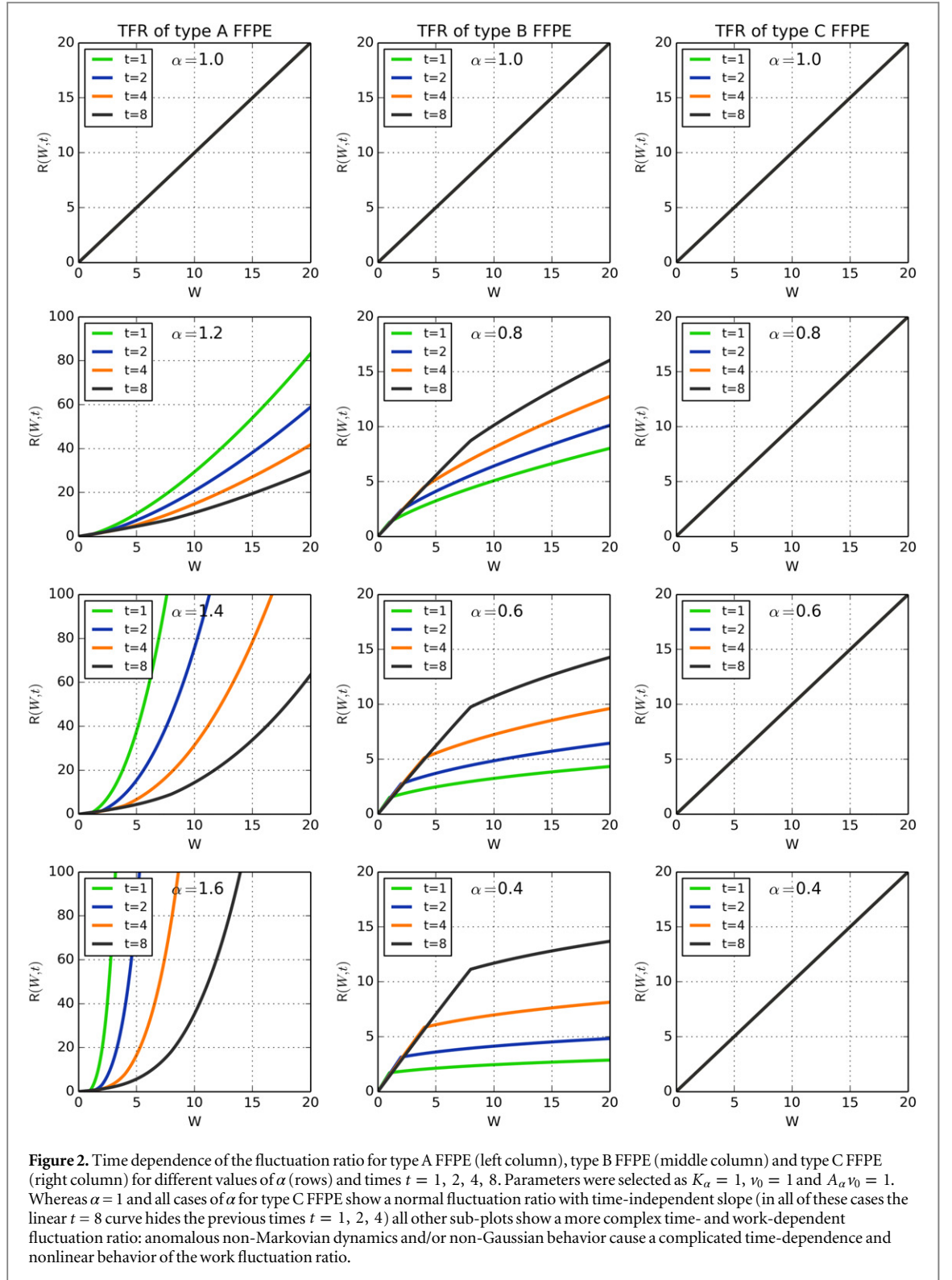
We remark that a normal TFR for type C can also be obtained with the use of the subordination principle: Indeed, it is known that the fractional kinetic equation C can be derived from the coupled Langevin equations for the motion of a particle [42, 47, 70]

$$\frac{\mathrm{d}x(u)}{\mathrm{d}u} = \frac{F_0}{m\gamma} + \xi(u), \quad \frac{\mathrm{d}t(u)}{\mathrm{d}u} = \tau(u), \tag{32}$$

where the random walk $x(t)$ is parameterized by the random variable u. The random process $\xi(u)$ is a white Gaussian noise, $\langle\xi(u)\rangle = 0$, $\langle\xi(u)\xi(u')\rangle = 2k_b T\delta(u - u')/(m\gamma)$, and $\tau(u)$ is a white stable Lévy noise, which takes positive values only and obeys a totally skewed α-stable Lévy distribution with $0 < \alpha < 1$. The PDF $f(x, t)$ of the process $x(t)$ is then given by

$$f(x, t) = \int_0^\infty \mathrm{d}u f_1(x, u) h(u, t), \tag{33}$$

where $f_1(x, u)$ is a shifted Gaussian PDF with drift, and $h(u, t)$ is the inverse one-sided Lévy stable density [67]. It is then easy to show that the linear normal TFR equation (31) holds due to Gaussianity of f_1. Moreover, it becomes clear that the normal TFR also holds for a more general form of the PDFs $h(u, t)$, that is, for a more general class of the positively valued stochastic processes $\tau(u)$.

Figure 2. Time dependence of the fluctuation ratio for type A FFPE (left column), type B FFPE (middle column) and type C FFPE (right column) for different values of α (rows) and times $t = 1, 2, 4, 8$. Parameters were selected as $K_\alpha = 1$, $v_0 = 1$ and $A_\alpha v_0 = 1$. Whereas $\alpha = 1$ and all cases of α for type C FFPE show a normal fluctuation ratio with time-independent slope (in all of these cases the linear $t = 8$ curve hides the previous times $t = 1, 2, 4$) all other sub-plots show a more complex time- and work-dependent fluctuation ratio: anomalous non-Markovian dynamics and/or non-Gaussian behavior cause a complicated time-dependence and nonlinear behavior of the work fluctuation ratio.

Type A and B FFPEs: for these two types the fluctuation ratio in Laplace space is more complicated than for type C FFPE in equation (30). It is obtained with equation (24) as

$$\frac{\tilde{p}_{A,B}(W, s)}{\tilde{p}_{A,B}(-W, s)} = \frac{\tilde{f}_{A,B}\left(W/F_0, s\right)}{\tilde{f}_{A,B}\left(-W/F_0, s\right)} = \exp\left(\frac{v_0}{F_0 K_\alpha} s^{\alpha-1} W\right). \tag{34}$$

In contrast to equation (30), here the right hand side depends on the Laplace variable *s*. Consequently, one may expect an anomalous ratio R which is confirmed numerically in the overview of figure 2. The fluctuation ratios of type A (left column) and type B FFPEs (middle column) show a nonlinear increase as functions of *W*. For type

B FFPEs there is a clear transition at the current maximum of the PDFs at $W_{max} = v_0 F_0 t$ which is equal to t with $v_0 = 1$ and $F_0 = 1$ in figure 2. For $W > W_{max}$ the fluctuation ratio increases with time. In contrast, the TFR of type A FFPE increases faster in W than for type B. At the scale of this overview plot there is no transition point visible as for type B FFPE. However, the qualitative time-dependence of the fluctuation ratio for type A FFPE is the opposite to type B FFPE: the ratio increases faster for smaller times. To gain further insight into this behavior, some asymptotic expansions of the TFR for type A and type B FFPEs are performed in the next section.

3.3. Asymptotic expansions of the fluctuation ratio for type A and B FFPE

In this section we analyze the asymptotic behavior of the work fluctuation ratio for type A and B FFPE. Differences between type A and type B simply correspond to the value of α which is $1 < \alpha < 2$ for the superdiffusive FFPE of type A and $0 < \alpha < 1$ for the subdiffusive type B FFPE. Type C is not considered anymore, as the analytical calculation of equation (31) and the numerical analysis in figure 2 have delivered a normal FR with a time-independent linear increase in the work W.

Small W expansion: first, the behavior of the TFR for the work PDFs of the FFPEs is studied for small W as a function of time. The logarithmic ratio of a continuously differentiable function $p(z)$ can be expanded as Taylor series for positive z as

$$\log \frac{p(z)}{p(-z)} = \frac{2}{p(z=0)} \left. \frac{dp(z)}{dz} \right|_{z=0} z + \mathcal{O}\left(z^2\right). \tag{35}$$

Inserting the approximate work PDF $p(W, t)$ from equation (25) together with the transformation of equation (27) into equation (35) requires the calculation of the derivative of the Fox H-function. Using equation (B.4) with $r = 1$, $h = 1$, $c = -1/\sqrt{K_\alpha t^\alpha}/F_0$, and $d = v_0 t/\sqrt{K_\alpha t^\alpha}$ allows us to calculate the linear term in the Taylor expansion of equation (35). With the assumption $W/F_0 < v_0 t$ and after some simplifications using the definition of the Fox H-function by the Mellin–Barnes integral in equation (B.1) one obtains the fluctuation ratio for small W as a quotient of two Fox H-functions:

$$\mathsf{R}(W, t)|_{W\to 0} = \left(\frac{2}{v_0 t}\right) \frac{H_{11}^{10}\left[\frac{v_0 t}{\sqrt{K_\alpha t^\alpha}} \left| \begin{matrix} (1-\alpha/2, \alpha/2) \\ (1, 1) \end{matrix}\right.\right]}{H_{11}^{10}\left[\frac{v_0 t}{\sqrt{K_\alpha t^\alpha}} \left| \begin{matrix} (1-\alpha/2, \alpha/2) \\ (0, 1) \end{matrix}\right.\right]} \frac{W}{F_0} = \Lambda(t)\, W. \tag{36}$$

The prefactor $\Lambda(t)$ summarizes the time-dependence of the fluctuation ratio. Its numerical evaluation based on the Taylor series of equation (B.3) is shown in figure 3(A). In the superdiffusive case $1 < \alpha < 2$ (type A FFPE) the prefactor $\Lambda(t)$ increases as a function of time, whereas in the subdiffusive case it decreases with time. The argument of the Fox H-functions $z = v_0 t/\sqrt{K_\alpha t^\alpha}$ in equation (36) scales $\sim t^{1-\alpha/2}$ with $1 - \alpha/2 > 0$ for $0 < \alpha < 2$. Thus the asymptotic expansion of these Fox H-functions can be used for $t \to \infty$. In the long time limit the scaling function $\Lambda(t)$ converges towards the following non-zero constant value:

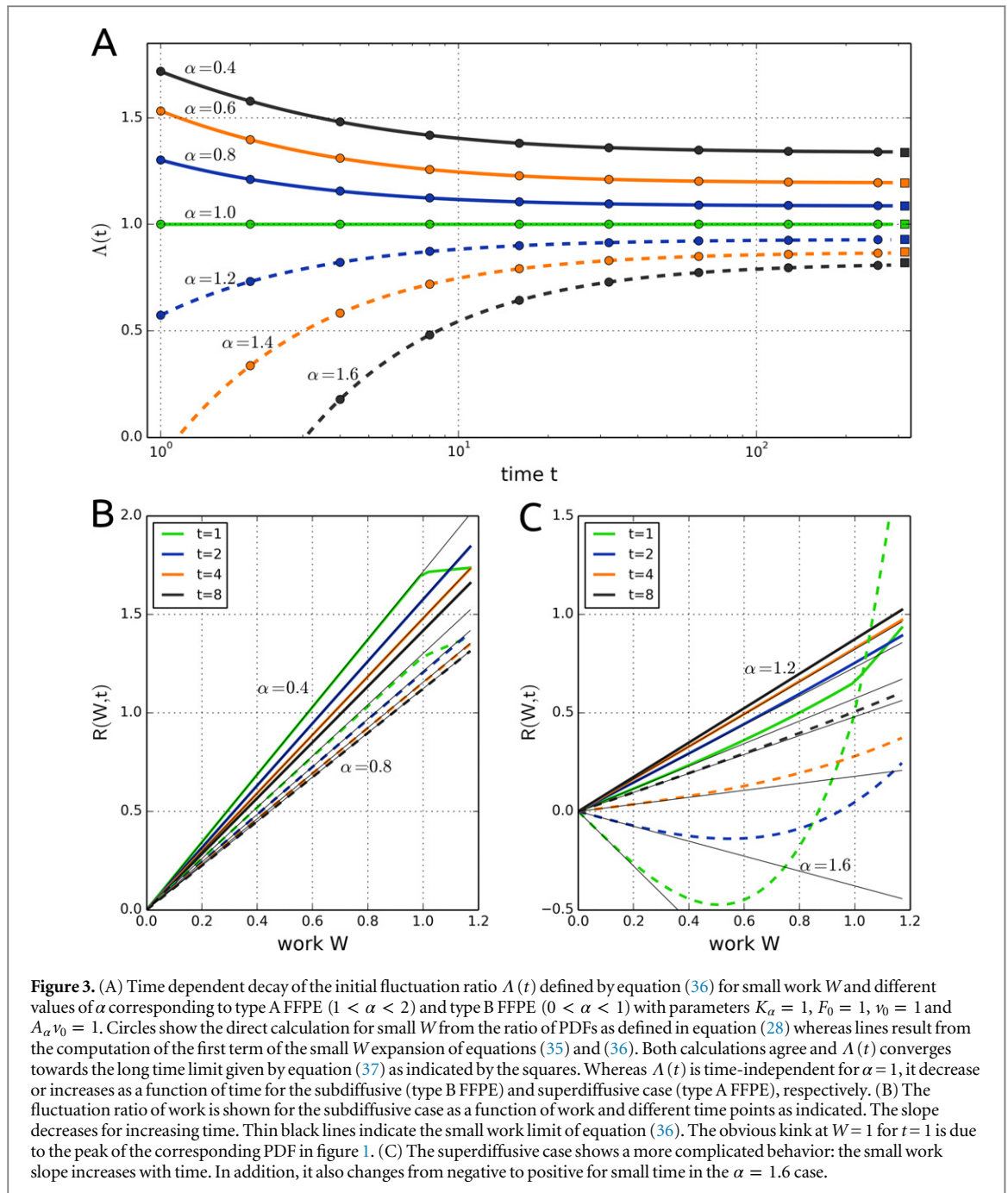$$\lim_{t\to\infty} \Lambda(t) = 2\left(\frac{2}{\alpha}\right)^{\alpha/(\alpha-2)} \left(\frac{v_0}{K_\alpha^{1/\alpha}}\right)^{\alpha/(2-\alpha)} \frac{W}{F_0} \to \frac{v_0}{K_1 F_0} W \quad \text{for} \quad \alpha = 1. \tag{37}$$

The corresponding values are shown as squares in figure 3(A) indicating the predicted asymptotic behavior. Figure 3(B) shows the spatial behavior of the work fluctuation ratio for two subdiffusive examples $\alpha = 0.4$ and 0.8 at different instants of time t (compare to small W values in the overview given in figure 2). The slope of the ratio decreases with increasing time and agrees well with the small W expansion given in equation (36). The superdiffusive case in figure 3(C) shows a reverse behavior as the small W ratio increases with time. As indicated in figure 3(A) it can also be negative as show in figure 3(C) for $\alpha = 1.6$ and $t = 1, 2$. In the superdiffusive case, the small W expansion has a smaller region of agreement with the exact ratio. The more complex behavior is technically due to the two separating peaks of the PDF as illustrated in figure 1.

Large W expansion: finally, the behavior of the work fluctuation ratio is studied for large values of the work W. The overview given in figure 2 shows a different nonlinear behavior for the subdiffusive and superdiffusive case. Assuming $W/F_0 > v_0 t$ and large arguments of the Fox H-function for type A and type B FFPE in equation (25) allows us to use the asymptotic expansion of the corresponding Fox H-function in equation (B.5). For large W one obtains the following relation:

$$\mathsf{R}(W, t)|_{W\to\infty} = \frac{2v_0 t}{F_0} \left(\frac{\alpha}{2}\right)^{\alpha/(2-\alpha)} \left(\frac{1}{\sqrt{K_\alpha t^\alpha}}\right)^{2/(2-\alpha)} W^{\alpha/(2-\alpha)}. \tag{38}$$
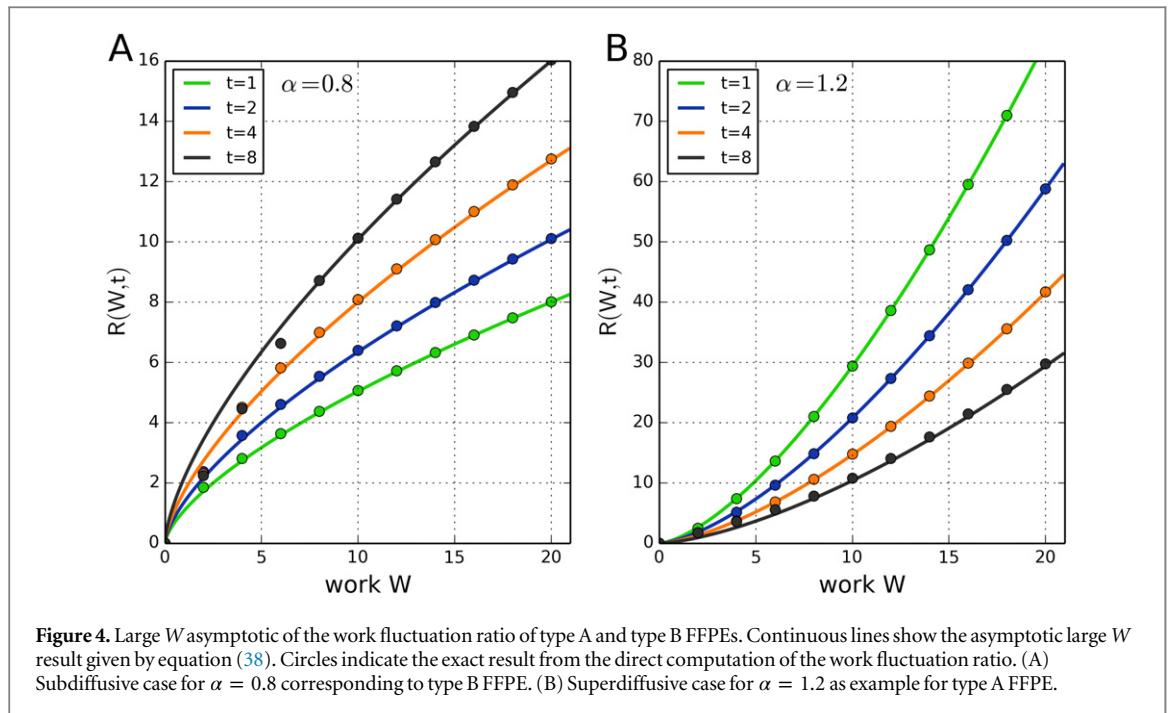
Thus the work fluctuation ratio scales as a power law with an exponent $\alpha/(2 - \alpha)$. This exponent is between 0 and 1 for the subdiffusive type B FFPE. For superdiffusive type A FFPE it is larger than 1. This asymptotic power

Figure 3. (A) Time dependent decay of the initial fluctuation ratio $\Lambda(t)$ defined by equation (36) for small work W and different values of α corresponding to type A FFPE ($1 < \alpha < 2$) and type B FFPE ($0 < \alpha < 1$) with parameters $K_\alpha = 1$, $F_0 = 1$, $v_0 = 1$ and $A_\alpha v_0 = 1$. Circles show the direct calculation for small W from the ratio of PDFs as defined in equation (28) whereas lines result from the computation of the first term of the small W expansion of equations (35) and (36). Both calculations agree and $\Lambda(t)$ converges towards the long time limit given by equation (37) as indicated by the squares. Whereas $\Lambda(t)$ is time-independent for $\alpha = 1$, it decrease or increases as a function of time for the subdiffusive (type B FFPE) and superdiffusive case (type A FFPE), respectively. (B) The fluctuation ratio of work is shown for the subdiffusive case as a function of work and different time points as indicated. The slope decreases for increasing time. Thin black lines indicate the small work limit of equation (36). The obvious kink at $W = 1$ for $t = 1$ is due to the peak of the corresponding PDF in figure 1. (C) The superdiffusive case shows a more complicated behavior: the small work slope increases with time. In addition, it also changes from negative to positive for small time in the $\alpha = 1.6$ case.

law behavior is shown in figure 4 for two examples. Continuous lines represent the result of equation (38) and agree for larger W values with the exact results denoted by circles. Equation (38) additionally contains a time-dependent scaling factor that is proportional $t^{(2\alpha-2)/(\alpha-2)}$. This factor is positive for the subdiffusive type B FFPE and negative for type A FFPE.

4. Summary and outlook

In this work we studied three different types of FFPEs generating anomalous diffusion: a superdiffusive one (type A), a subdiffusive one (type B), and another one that exhibits a transition from sub- to superdiffusion under parameter variation (type C). Type A and type B break FDR1 while type C preserves it. Type A can be derived, under certain assumptions, from an overdamped Langevin equation with power law correlations of the velocity fluctuations, types B and C have been derived before in the literature from CTRW theory. Type C can also be obtained via subordination. We then calculated position PDFs for all models analytically and studied the shapes of all PDFs numerically under variation of the anomaly index as they evolve in time. Finally we checked the work

Figure 4. Large *W* asymptotic of the work fluctuation ratio of type A and type B FFPEs. Continuous lines show the asymptotic large *W* result given by equation (38). Circles indicate the exact result from the direct computation of the work fluctuation ratio. (A) Subdiffusive case for $\alpha = 0.8$ corresponding to type B FFPE. (B) Superdiffusive case for $\alpha = 1.2$ as example for type A FFPE.

TFR for all three models. Especially, we studied the time dependence of the ratio of the work fluctuations both for small and for large work by analytical asymptotic expansions in comparison to numerical evaluations.

We find that our type C model with FDR1 exhibits a conventional work TFR for all times, meaning the fluctuation ratio is constant in time and linear in the work. For a correlated Gaussian stochastic process it was shown that FDR1 implies the existence of a conventional TFR [43]. Our work generalizes this result to an example of non-Gaussian PDFs generated by FFPE dynamics. It is interesting that the conventional TFR is still obeyed, despite the highly non-trivial dynamics exhibited by both the position PDFs and the corresponding moments. The existence of the conventional TFR for this case is connected to the fact that only the equation for Type C describes a subordinated process, namely the one subordinated to Brownian motion with drift under random time transformation. An important open question is to which extent figure 1 in [43] summarizing the interplay between FDR1, FDR2 and TFRs for correlated Gaussian stochastic processes in terms of necessary and sufficient conditions can be generalized to non-Gaussian processes. For our other two models type A and type B the position PDFs show also very subtle and non-trivial non-Gaussian shapes. However, in contrast to type C they are characterized by a highly non-trivial fluctuation ratio: For type A the latter decreases with time, for type B it increases. Similar results have been obtained for the work TFR of strongly correlated Gaussian stochastic processes without FDR1 [42, 43]. On top of this, for both types of FFPEs the fluctuation ratio yields different long time limits depending on whether the work is small or large: for small work the fluctuation ratio converges to linearity in the work with constant prefactors, which reminds of the conventional TFR; however, here the slopes depend on the anomaly index of the dynamics. For large work the fluctuation ratio remains nonlinear in the work, with convex and concave shapes for type A and type B, respectively.

Our work was motivated by experiments on cell migration [50], where data were successfully fitted by solutions of a fractional Klein–Kramers equation [48]. Several generalizations of such a Klein–Kramers equation have been proposed to describe processes under external fields [48, 49, 51], which in turn yield FFPEs for the position only, similar to the ones studied in our paper, as special cases [12, 21, 52]. We thus believe that our present work might have important applications to understand cell migration in non-equilibrium situations such as under chemical gradients; see [44] for first results. More generally, our theory might have applications to understand glassy non-equilibrium dynamics: In computer simulations of a number of glassy systems violations of conventional TFRs have been observed featuring fluctuation ratios that are nonlinear in the work with time-dependent prefactors [40, 41].

Apart from such experimental applications, our first approach for deriving a FFPE pioneered by Balescu [57, 58] deserves to be studied in more detail. For example, it would be interesting to derive a superdiffusive FFPE from it that preserves FDR1, and to check again the TFR. On a broader scale it would be important to generalize our approach by considering more general observables, ideally dissipation functions [1] or related functionals defined within stochastic thermodynamics [7]. More general force fields than simply constant forces

[42] and other types of FRs could be tested as well. Such theoretical studies may pave the way to identify different classes of anomalous FRs characterized by specific functional forms, generalized FDRs associated with them, and to explore the physical significance of these results. Last not least the quality of the Galilean invariant approximate solution equation (25) [12, 20] of the FFPEs (8), (10) needs to be investigated in detail.

Acknowledgments

Appendix A. Pseudo-Liouville approach

Following the so-called pseudo-Liouville hybrid approach of Balescu [57, 58] allows us to relate the dynamics of a particle defined by a Langevin equation to the corresponding PDF of the stochastic process. We start from the Langevin equation for the position $x(t)$ of a particle

$$\frac{\mathrm{d}x(t)}{\mathrm{d}t} = v_0 + v(t),\tag{A.1}$$

where $v(t)$ is a correlated stochastic process with zero mean $\langle v(t)\rangle = 0$ and a given correlation function $\langle v(t)v(t')\rangle = \mathsf{T}(t-t')$, where the average is performed over the stochastic process $v(t)$. v_0 denotes a constant external force. The stochastic function $F(x, t)$

$$F(x, t) = \delta(x - x(t))\tag{A.2}$$

represents the exact density of the process. Derivation of equation (A.2) with respect to time and the usage of the Langevin equation (A.1) delivers the continuity equation for the exact density $F(x, t)$:

$$\frac{\partial F(x, t)}{\partial t} = -\frac{\partial}{\partial x}\delta(x - x(t))\,\frac{\mathrm{d}x(t)}{\mathrm{d}t} \longrightarrow \frac{\partial F(x, t)}{\partial t} + \left[v_0 + v(t)\right]\frac{\partial F(x, t)}{\partial x} = 0.\tag{A.3}$$

Now, the exact density $F(x, t)$ is decomposed into an averaged part $f(x, t)$ and fluctuations $\delta f(x, t)$

$$F(x, t) = f(x, t) + \delta f(x, t)\quad \text{with}\quad f(x, t) = \langle F(x, t)\rangle.\tag{A.4}$$

It is the further aim of this appendix to calculate the PDF $f(x, t)$ for the stochastic process defined by the Langevin equation (A.1) for given correlations of $v(t)$. Averaging of the exact density in equation (A.3) leads to

$$\left(\frac{\partial}{\partial t} + v_0\frac{\partial}{\partial x}\right)f(x, t) = -\frac{\partial}{\partial x}\langle v(t)\delta f(x, t)\rangle.\tag{A.5}$$

Subtraction of equation (A.5) from equation (A.3) results in

$$\left(\frac{\partial}{\partial t} + \left[v_0 + v(t)\right]\frac{\partial}{\partial x}\right)\delta f(x, t) = -\frac{\partial}{\partial x}(v(t)\,f(x, t) - \langle v(t)\,\delta f(x, t)\rangle).\tag{A.6}$$

Equation (A.6) can be solved with the method of characteristics

$$\delta f(x, t) = -\frac{\partial}{\partial x}\int_0^t \mathrm{d}t'(v(t')\,f(x - \Delta(t, t'), t') - \langle v(t')\,\delta f(x - \Delta(t, t'), t')\rangle)\tag{A.7}$$

with the definition $\Delta(t, t') = v_0(t - t') - \int_{t'}^t \mathrm{d}t_1\,v(t_1)$. Inserting equation (A.7) into equation (A.5) delivers the final equation for the PDF $f(x, t)$:

$$\left(\frac{\partial}{\partial t} + v_0\frac{\partial}{\partial x}\right)f(x, t) = \frac{\partial^2}{\partial x^2}\int_0^t \mathrm{d}t'\langle v(t)v(t')\,f(x - \Delta(t, t'), t')\rangle.\tag{A.8}$$

This is an exact relation for $f(x, t)$ that is generally non-local in space and non-local in time, i.e. non-Markovian. Applications and approximations of this relation are studied in section 2.1.

Appendix B. Definiton and properties of Fox *H*-functions

The Fox *H*-function is defined as inverse Mellin transform of the function $\chi(s)$ [12, 71]

$$H_{p,q}^{m,n}(z) = H_{p,q}^{m,n}\left[z\,\left|\,\begin{matrix}\left(a_j, A_j\right)_{j=1,p}\\\left(b_j, B_j\right)_{j=1,q}\end{matrix}\right.\right] = \frac{1}{2\pi\mathrm{i}}\int_{\mathcal{L}}\chi(s)z^s\mathrm{d}s\tag{B.1}$$

over a suitable path $\mathcal{L}$, with

$$\chi(s) = \frac{\prod_{j=1}^{m} \Gamma\left(b_j - B_j s\right) \prod_{j=1}^{n} \Gamma\left(1 - a_j + A_j s\right)}{\prod_{j=m+1}^{q} \Gamma\left(1 - b_j + B_j s\right) \prod_{j=n+1}^{p} \Gamma\left(a_j - A_j s\right)},$$ (B.2)

$0 \leqslant n \leqslant p, 1 \leqslant m \leqslant q, (a_j, b_j) \in \mathbb{C}.$, and $(A_j, B_j) \in \mathbb{R}^+$. Empty products in equation (B.2) are taken as one.

A series expansion allows the numerical calculation of Fox H-functions. The following form for a special Fox H-function is used:

$$H_{11}^{10}\left[z \left|\begin{array}{c} \left(a_1, A_1\right) \\ \left(b_1, B_1\right) \end{array}\right.\right] = \sum_{k=0}^{\infty} \frac{(-1)^k}{\Gamma\left(a_1 - A_1 \dfrac{b_1 + k}{B_1}\right)} \frac{z^{\frac{b_1+k}{B_1}}}{k!\, B_1}.$$ (B.3)

Summation in this work is performed numerically with multiple-precision arithmetic.

The derivation of the Fox H-function is required to calculate the fluctuation ratio for the FFPEs of type A and B. This can be performed using the following relation [72]:

$$\frac{\mathrm{d}^r}{\mathrm{d}x^r} H_{p,q}^{m,n}\left[(cx + d)^h \left|\begin{array}{c} \left(a_j, A_j\right)_{1,p} \\ \left(b_j, B_j\right)_{1,q} \end{array}\right.\right]$$

$$= \left(\frac{c}{cx + d}\right)^r H_{p+1,q+1}^{m,n+1}\left[(cx + d)^h \left|\begin{array}{c} (0, h), \left(a_j, A_j\right)_{1,p} \\ \left(b_j, B_j\right)_{1,q}, (r, h) \end{array}\right.\right].$$ (B.4)

For large arguments the Fox H-functions of type $H_{p,q}^{q,0}(z)$ decay as stretched exponential functions. The asymptotics of the PDF in equation (25) is given for large z by [72, 73]

$$f_{A,B}(z, t) \sim \frac{z^{-\frac{1-\alpha}{2-\alpha}}}{\sqrt{4\pi(2-\alpha)K_\alpha t^\alpha}} \left(\frac{2}{\alpha}\right)^{\frac{1-\alpha}{2-\alpha}} \exp\left\{ -\left(\frac{2-\alpha}{2}\right)\left(\frac{2}{\alpha}\right)^{\frac{\alpha}{\alpha-2}} z^{\frac{2}{2-\alpha}} \right\}$$ (B.5)

for $z \to \infty$ and $z = |x - v_0 t| \big/ \sqrt{K_\alpha t^\alpha}$.

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