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TOPICAL REVIEW

The restaurant at the end of the random walk: recent developments in the description of anomalous transport by fractional dynamics

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Abstract

Fractional dynamics has experienced a firm upswing during the past few years, having been forged into a mature framework in the theory of stochastic processes. A large number of research papers developing fractional dynamics further, or applying it to various systems have appeared since our first review article on the fractional Fokker–Planck equation (Metzler R and Klafter J 2000a, *Phys. Rep.* **339** 1–77). It therefore appears timely to put these new works in a cohesive perspective. In this review we cover both the theoretical modelling of sub- and superdiffusive processes, placing emphasis on superdiffusion, and the discussion of applications such as the correct formulation of boundary value problems to obtain the first passage time density function. We also discuss extensively the occurrence of anomalous dynamics in various fields ranging from nanoscale over biological to geophysical and environmental systems.

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(Some figures in this article are in colour only in the electronic version)

1. Introduction

For seven and a half million years, Deep Thought computed and calculated, and in the end announced that the answer was in fact Forty-two—and so another, even bigger, computer had to be built to find out what the actual question was³.

The notions and concepts of anomalous dynamical properties, such as long-range spatial or temporal correlations manifested in power laws, stretched exponentials, $1/f^\alpha$ -noises,

³ Douglas Adams, *The Restaurant at the End of the Universe*, Tor Books, 1988.

or non-Gaussian probability density functions (PDFs), have been predicted and observed in numerous systems from various disciplines including physics, chemistry, engineering, geology, biology, economy, meteorology, astrophysics and others. Apart from other standard tools to describe anomalous dynamics such as continuous time random walks (Blumen *et al* 1986a, Bouchaud and Georges 1990, Hughes 1995, Klafter *et al* 1996, Shlesinger *et al* 1993), fractional dynamical equations have become increasingly popular to model anomalous transport (Barkai 2001, Hilfer 2000, Metzler and Klafter 2000a, 2001, Sokolov *et al* 2002). In the presence of an external force field, in particular, the fractional Fokker–Planck equation provides a direct extension of the classical Fokker–Planck equation, being amenable to well-known methods of solution.

This review updates and complements with new and different perspectives the ‘Random walk’s guide to anomalous diffusion’ (Metzler and Klafter 2000a). Since its publication, a large volume of research covering recent developments in the fractional dynamics framework and its applications has been conducted, most of which is brought together herein. We refrain from a repetition of the historical context and the mathematical details presented in Metzler and Klafter (2000a, 2001), and we build on the material and notation introduced there. What we wish to point out is the framework character: just as the regular Fokker–Planck equation renders itself to the description of a plethora of processes, so does the fractional analogue in all those systems whose statistics is governed by the ubiquitous power laws. The breadth of such potential applications, at the same time, may indeed encourage the usage in many new fields.

We start with a collection of systems, in which have been observed anomalous processes with long-range correlations, covering both experimental and theoretical evidence. Having set the scene, we divide the introduction of fractional dynamics concepts between subdiffusive and superdiffusive processes, and for the latter we distinguish between Lévy flights and walks (Klafter *et al* 1996, Shlesinger *et al* 1993). Finally, we discuss various applications of the framework, in particular, the formulation and solution of first passage time problems and fractional diffusion–reaction processes. In the appendix, we collect some important concepts and definitions on fractional operators.

2. Processes of anomalous nature

As already mentioned, processes deviating from the classical Gaussian diffusion or exponential relaxation patterns occur in a multitude of systems. The anomalous features usually stretch over the entire data window, but there exist examples when they develop after an initial period of sampling (finite size/time effects), or they may be transient, i.e., eventually the anomalous process nature turns into normal transport or relaxation dynamics. In the anomalous regime, possibly the most fundamental definition of anomaly of the form we have in mind is the deviation of the mean squared displacement

$$\langle(\Delta\mathbf{r})^2\rangle = \langle(\mathbf{r} - \langle\mathbf{r}\rangle)^2\rangle = 2dK_\alpha t^\alpha \quad (1)$$

from the ‘normal’ linear dependence $\langle(\Delta\mathbf{r})^2\rangle = 2dK_1t$ on time. Here, d is the (embedding) spatial dimension, and K_1 and K_α are the normal and generalized diffusion constants of dimensions $\text{cm}^2 \text{s}^{-1}$ and $\text{cm}^2 \text{s}^{-\alpha}$, respectively. The anomalous diffusion exponent $\alpha \neq 1$ determines whether the process will be categorized as *subdiffusive* (dispersive, slow) if $0 < \alpha < 1$, or *superdiffusive* (enhanced, fast) if $1 < \alpha$. Usually, the domain $1 < \alpha \leq 2$ is considered, $\alpha = 2$ being the ballistic limit described by the wave equation, or its forward and

Table 1. Comparison of different anomalous diffusion models to normal Brownian motion (BM) (Lévy 1965, van Kampen 1981): PDFs of fractional Brownian motion (FBM) (Mandelbrot and van Ness 1968, Lim and Muniandy 2002, Lutz 2001b, Kolmogorov 1940), generalized Langevin equation with power-law kernel (GLE) (Kubo *et al* 1985, Lutz 2001b, Wang *et al* 1994, Wang and Tokuyama 1999); continuous time random walk (CTRW) of types subdiffusion (SD), Lévy flights (LF) and Lévy walks (Klafter *et al* 1987, 1996, Shlesinger *et al* 1993); as well as time-fractional dynamics (TFD), which covers both subdiffusion (in this case it corresponds to SD) and sub-ballistic superdiffusion (Metzler and Klafter 2000a, 2000d). The c_i are constants.

	PDF	Comments
BM	$P(x, t) = (4\pi Kt)^{-1/2} \exp(-x^2/(4Kt))$	
FBM ^a	$P(x, t) = (4\pi K_\alpha t^\alpha)^{-1/2} \exp(-x^2/(4K_\alpha t^\alpha))$	$0 < \alpha \leq 2$
GLE ^b	$P(x, t) = (4\pi K_\alpha t^\alpha)^{-1/2} \exp(-x^2/(4K_\alpha t^\alpha))$	$0 < \alpha < 2, \alpha \neq 1$
SD ^c	$P(x, t) \sim c_1 t^{-\alpha/2} \xi^{-(1-\alpha)/(2-\alpha)} \exp(-c_2 \xi^{1/(1-\alpha/2)}),$ $\xi \equiv x /t^{\alpha/2} \therefore \psi(t) \sim \tau^\alpha/t^{1+\alpha}$	$0 < \alpha \leq 1$
LF ^d	$P(x, t) = \mathcal{F}^{-1}\{\exp(-K^\mu t x ^\mu)\} \sim K^\mu t/ x ^{1+\mu}$	$0 < \mu \leq 2$
LW ^e	$P(k, u) = \frac{1}{u} \psi(u)/[1 - \psi(k, u)] \therefore$ $\psi(x, t) = \frac{1}{2} x ^{-\mu} \delta(x - v_\nu t^\nu)$	$\nu\mu > 1$
TFD ^f	$P(x, t) \sim c_1 t^{-\alpha/2} \xi^{-(1-\alpha)/(2-\alpha)} \exp(-c_2 \xi^{1/(1-\alpha/2)}),$ $\xi \equiv x /t^{\alpha/2}$	$0 < \alpha < 2$

^a Note that there are various definitions of FBM. However, the Gaussian nature is common to all versions. The behaviour of FBM is antipersistent for $0 < \alpha < 1$, and persistent for $1 < \alpha \leq 2$ (Mandelbrot 1982).

^b The GLE is in some sense more fundamental than the FBM. For instance, it occurs naturally in hydrodynamic backflow (Kubo *et al* 1985, Landau and Lifshitz 1987), and generally includes an external force. The case $\alpha = 1$ leads to a logarithmic correction of the form $\langle x^2(t) \rangle \sim t \log t$ in the GLE formulation chosen in Wang and Tokuyama (1999).

^c Same (asymptotic) PDF as in the TFD case with $0 < \alpha \leq 1$.

^d The symmetric Lévy stable law of index μ , with diverging variance $\langle x^2(t) \rangle = \infty$. LFs correspond to the space-fractional diffusion equation (34).

^e For appropriate exponents μ and ν , LWs lead to the SD and to the TFD, while for superdiffusion they exhibit δ -spikes that spread apart (with constant velocity for $\nu = 1$), continuously spanning a Lévy stable-like propagator between them (Klafter and Zumofen 1994a). Superdiffusive LWs are described in terms of the fractional material derivative (55). Compare also section 4.3.

^f Stretched ($0 < \alpha < 1$) and compressed ($1 < \alpha < 2$) Gaussian governed by equation (9) for $0 < \alpha < 1$, and by a fractional wave equation for $1 < \alpha < 2$ (Schneider and Wyss 1989, Metzler and Klafter 2000a, 2000d).

backward modes (Landau and Lifshitz 1984)⁴. Processes with $\alpha > 2$ are known, such as the Richardson pair diffusion ($\langle R^2(t) \rangle \sim t^3$) in fully developed turbulence (Richardson 1926). However, we will restrict our discussion to sub-ballistic processes with $\alpha < 2$ explicitly⁵. An exception to equation (1) is unconfined Lévy flights, for which we observe a diverging mean squared displacement⁶. We will concentrate on the one-dimensional case, to keep notation simple, in particular, for the case of Lévy flights.

Before we continue, we stop to highlight the parallels and main differences between fractional dynamics and other dynamical models, as compiled in table 1 for force-free

⁴ Note that the diffusion equation can be rephrased with a half-order derivative in time and first-order derivative in space. This is exact in $d = 1$ and $d = 3$ (Oldham and Spanier 1972), and asymptotically correct in general fractal dimension (Metzler *et al* 1994).

⁵ It was shown that the restriction to sub-ballistic motion guarantees for all non-pathological processes fulfilling equation (1) that the fluctuation–dissipation theorem holds (Costa *et al* 2003, Morgado *et al* 2002).

⁶ As will be discussed below, in the presence of steep external potentials, the mean squared displacement of Lévy flights becomes finite.

anomalous diffusion. Thus, Brownian motion (BM) can be generalized using the continuous time random walk (CTRW) model to subdiffusion or dispersive transport (SD), to Lévy flights (LF), or to Lévy walks (LW); see the table caption for more details. All of these models can be mapped onto the corresponding fractional equations, as discussed in the following sections. These descriptions differ from fractional Brownian motion (FBM) or the generalized Langevin equation (GLE). Despite being Gaussian in nature such as the PDF in Brownian dynamics, fractional Brownian motion, and generalized Langevin equation descriptions, the subdiffusion PDF has an asymptotic stretched Gaussian shape, Lévy flights are characterized by a long-tailed Lévy stable law, and Lévy walks exhibit spikes of finite propagation velocity, in between which an approximate Lévy stable PDF is being spanned continuously. As we will see from the fractional dynamical equations corresponding to SD, LFs and LWs, they are highly non-local, and carry far-reaching correlations in time and/or space, represented in the integro-differential nature (with slowly decaying power-law kernels) of these equations. In contrast, FBM and GLE on the macroscopic level are local in space and time, and carry merely time- or space-dependent coefficients. We also note that anomalous diffusion can be modelled in terms of non-linear Fokker–Planck equations based on non-extensive statistical approaches (Borland 1998). However, we intend to consider linear equations in what follows.

To build our case, let us continue by presenting a list of examples from different areas for which the anomalous character has been demonstrated.

2.1. Geophysical and geological processes

The seasonal variations of rivers, and the water balance in general, have been studied extensively over many decades, in particular, due to their environmental importance. Thus, for the water discharge variations of Lake Albert during his studies of the time variations of the Nile river, Hurst found that they cannot fall into the class of statistically independent processes, but can only be explained by a process which is correlated in time. Similar effects were reported on rainfall statistics and tree rings (Hurst 1951, Hurst *et al* 1951, Feder 1988). More recently, drought duration and rain duration as well as rain size of localized rain events have in fact been confirmed to obey power-law statistics (Dickman 2003, Peters *et al* 2002), which also enter earthquake aftershocks dynamics (Helmstetter and Sornette 2002). But also the ‘products’ of dynamical processes are often non-trivial, such as the fractal nature of coastline, also known as the ‘Coastline-of-Britain’ phenomenon based on data collected by Richardson (Mandelbrot 1967a), or the anomalous scaling between drainage area and river network length discovered originally by Hack (1957).

Considerable attention is paid to the investigation of tracer diffusion in subsurface hydrology, primarily for its obvious environmental implications. Thus, large-scale field experiments were undertaken, such as at the Borden site in Ontario, Canada (Sudicky 1986), at Cape Cod, Massachusetts (LeBlanc *et al* 1991), or during the MAcro-Dispersion Experiment (MADE) at Columbus Air Force Base, Mississippi (Boggs *et al* 1993, Adams and Gelhar 1992, Rehfeldt *et al* 1992), indicating that tracer dispersion is controlled by strong non-locality causing highly non-Gaussian PDFs (in this context often called plumes) seen as ‘scale-dependent dispersion’ (Gelhar *et al* 1992). It has been shown that long-tailed waiting time distributions with a comparably small number of fit parameters can well account for the observed behaviour (Berkowitz *et al* 2002, Berkowitz and Scher 1995, 1997, Scher *et al* 2002a); however, also space-fractional models were used to account for the anomalies (Benson *et al* 2001). In a similar study, long-time catchment data of chloride tracer in rainwater recorded in Wales, UK, were shown to follow $1/f$ statistics in the power spectrum (Kirchner *et al* 2000), which might indicate strongly non-local correlations in time, which

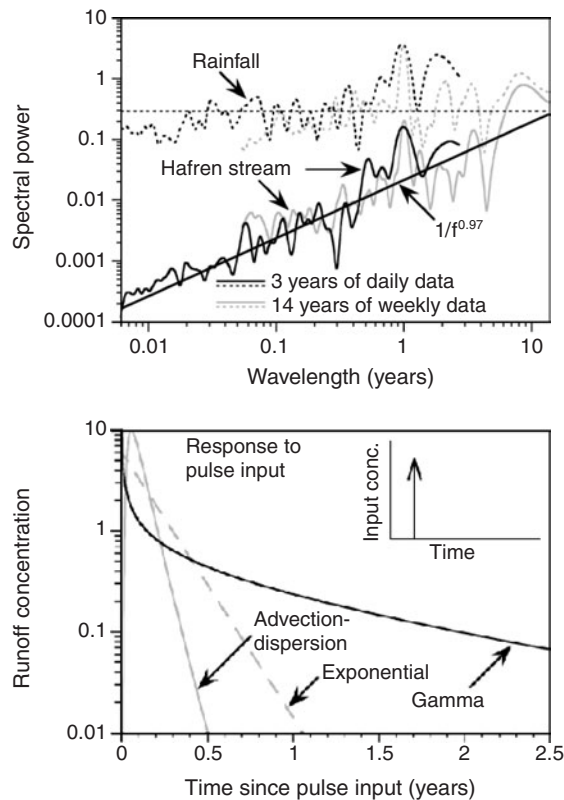


Figure 1. Top: power spectra of chloride tracer originally contained in rainwater, and as measured at the outflow to the Hafren stream after crossing the catchment. Chloride spectra of rainfall (dotted lines) resemble white noise; those of stream flow (solid lines) resemble $1/f$ -noise, with spectral power increasing proportionally to wavelength across the entire range of scales (data measured daily for 3 years, and weekly for 14 years). Bottom: response of stream-flow concentrations to a δ -function pulse input of contaminants. Because of the long-tailed nature in comparison to conventional models, contaminant concentrations are sustained substantially for much longer time spans. The logarithmic concentration scale emphasizes the persistence of low-level contamination of the Gamma-fit $\propto t^{\alpha-1} e^{-t/\tau}$ ($\alpha \simeq 0.5$; $\tau \simeq 1.9$ years is close to the edge of the data window such that essentially all data follow the power law $t^{-1/2}$) used in the original work (Kirchner *et al* 2000). The inset depicts δ -function contaminant input.

was later interpreted from an anomalous dynamics point of view, indicating that the data are perfectly consistent with a power-law form for the sticking time distribution of tracer particles in the catchment, causing extremely long retention times (Scher *et al* 2002a). Any contaminant getting into an aquifer fostering such anomalous dynamics will take considerably longer to leave the aquifer than the advecting water, in which it diffuses. This is, for instance, illustrated in figure 1, contrasting the drift-dominated behaviour of the water with the tracer outflow. According to the modelling brought forth in Scher *et al* (2002a), the mean retention time for the tracer becomes infinite, and is possibly due to sticking effects or trapping of the tracer in side channels off the aquifer backbone. We note that even on the laboratory scale, fairly simple systems were found to exhibit anomalous tracer dispersion (Berkowitz *et al* 2000a, 2000b), a problem still lacking a deeper understanding. In a similar manner, on-bed particle diffusion in gravel bed flows was recently shown to exhibit different transport regimes, ranging from ballistic to subdiffusion (Nikora *et al* 2002).

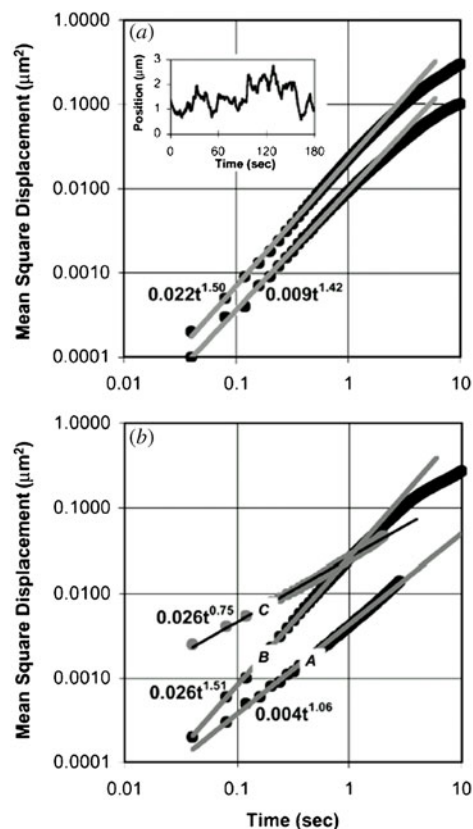


Figure 2. Mean squared displacement of engulfed microspheres in the cytoskeleton of a living cell. Active, motor-driven transport with exponent $3/2$ turns to subdiffusion with exponent $3/4$ (occasionally normal diffusion) (Caspi *et al* 2000).

2.2. Biological systems

Within a single biological cell, the motion of microspheres was found to have a transient superdiffusive behaviour with $\alpha = 3/2$ (motor-driven motion), an exponent suspiciously close to the motion in random velocity fields (Matheron and de Marsily 1980, Zumofen *et al* 1990)⁷. This active superdiffusion is followed by a subdiffusive (in some instances also normal diffusive) scaling (Caspi *et al* 2000, 2002, 2001). Some typical experimental results are depicted in figure 2. Similar subdiffusive behaviour in cells is known from lipid granular inclusions in the cytoskeleton of *E. coli* cells (Tolic-Nørrelykke *et al* 2003). We note that such presumably cytoskeleton-mediated anomalous diffusion patterns are consistent with findings from diffusion assays of microspheres in polymer networks (Amblard *et al* 1996, 1998a, 1998b), and time anomalies are also known from fluorescence video-microscopy assays (LeGoff *et al* 2002), and microrheology experiments on semiflexible polymers (Wong *et al* 2003, Tseng and Wirtz 2002), and from regular polymer melts (Fischer *et al* 1996, Kimmich 1997). The subdiffusive phenomenon may in fact be related to caging-caused subdiffusion (Weeks and Weitz 2002, Weeks *et al* 2000).

⁷ We note that anomalous diffusion-assisted ratchet transport was studied to some detail in Bao (2003) and Bao and Zhuo (2003); compare the fractional generalization of the Kramers problem, which was originally formulated in Metzler and Klafter (2000f), see also So and Liu (2004).

In proteins, a detailed analysis based on Fourier transform infrared spectroscopy data (Iben *et al* 1989, Austin *et al* 1974, 1975) demonstrated that ligand rebinding to myoglobin follows an asymptotic power-law decay. Data analysis showed that the entire measured rebinding curve follows fractional dynamics with Vogel–Fulcher-type temperature activation (Glöckle and Nonnenmacher 1995). Measurements of single ion gating channels using the patch clamp technique show logarithmic oscillations around a power-law trend (Blatz and Magleby 1986), which was demonstrated to correspond to a power-law distribution of characteristic times and amplitudes of individual exponential relaxation contributions (Nonnenmacher and Nonnenmacher 1989). Similarly, the passage of a single bio-oligo- or macromolecule through a membrane pore (Meller 2003) was recently shown to have *a priori* unexpected long-time contributions (Bates *et al* 2003, Metzler and Klafter 2003, Flomenbom and Klafter 2003, 2004). While for short chains anomalous time behaviour is most likely caused by chain–pore interactions (sticking) (Bates *et al* 2003), in the case of long chains the anomalous nature follows *a fortiori* from the polymer relaxation time (Chuang *et al* 2002). We note that long passage times were also found in fluorescence microscopy single-molecule assays of DNA uptake into the cell nucleus (Salman *et al* 2001). Compelling evidence for broad time-scale distributions in protein conformational dynamics was reported recently, and modelled on the basis of the fractional Fokker–Planck equation (Yang *et al* 2003, Yang and Xie 2002). In figure 3, we reproduce the fluorescence autocorrelation function fitted by various functions, showing the superior quality of the anomalous diffusion model, as well as a reconstruction of the energy landscape of the protein conformation, see figure 3 for details.

In a double-stranded DNA heteropolymer made up of the nucleotides (bases) A(denine), G(uanine), C(ytosine) and T(hymine), the entropy-carrying, flexible single-stranded bubbles, which open up due to thermal fluctuations, are preferentially located in areas rich in the weaker AT bonds (Altan-Bonnet *et al* 2003, Hanke and Metzler 2003). On diffusion along the DNA backbone, the bubbles have to cross tighter GC-rich regions, an effect which was shown to produce subdiffusion (Hwa *et al* 2003). Similarly, the motion of DNA-binding proteins along DNA due to differences in the local structure is subdiffusive (Slutsky *et al* 2003). In contrast, the points at which a random walker on a polymer chain can jump to another chain segment, which is close by in 3D (three-dimensional) space but distant in terms of the chemical coordinate, are distributed like an LF (Brockmann and Geisel 2003b, Sokolov *et al* 1997), which may contribute to fast target localization of (regulatory) proteins along DNA (Berg *et al* 1981); in particular, with respect to situations of overwhelming non-specific binding (Bakk and Metzler 2004a, 2004b), compare the on-DNA investigation in Slutsky and Mirny (2004). We note that such dynamical features may be employed for DNA sequencing, which is in turn related to Lévy signatures (Scafetta *et al* 2002). A particle attached to a (biological) membrane and confined to an harmonic potential (as fulfilled to good approximation in an optical tweezers field) displays anomalous relaxation behaviour related to Mittag-Leffler functions (Granek and Klafter 2001). Also the boundary layer thickness around a membrane exhibits subdiffusive behaviour (Dworecki *et al* 2003, Kosztołowicz and Dworecki 2003).

On somewhat larger scales, NMR field gradient measurements of biological tissues (Köpf *et al* 1996) could be shown to reveal anomalous diffusion behaviour in cancerous regions in both time and space resolution (Köpf *et al* 1998). Finally, the trajectories between turning or resting points of biological species within their habitats have been found to follow power-law statistics, such observations pertaining from bacteria (Shlesinger and Klafter 1990, Levandowsky *et al* 1997) and plankton (Visser and Thygesen 2003), over spider-monkeys (Ramos-Fernandez *et al* 2003) and jackals (Atkinson *et al* 2002), to the famed flight of an

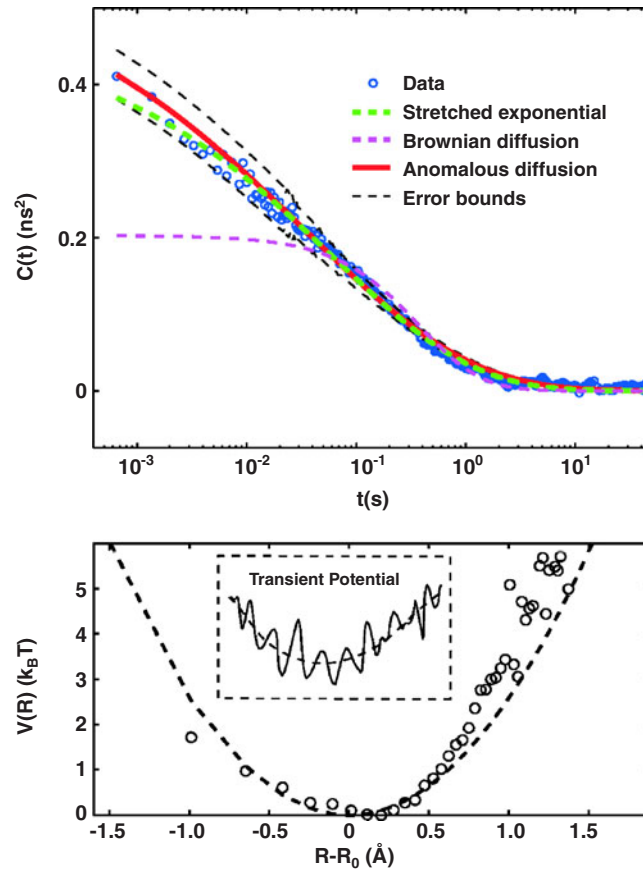


Figure 3. Top: fit of the experimental autocorrelation function of fluorescence lifetime fluctuations by stretched exponential and anomalous diffusion models, in comparison to the rather bad fits by a Brownian diffusion model. Bottom: potential of mean force calculated from measurements. The dashed line is a fit to a harmonic potential with variance of 0.19 \AA^2 . Inset: a sketch of a rugged ‘transient’ potential resulting from the short-time projection of 3D motions of the protein to the experimentally accessible coordinate (Yang *et al* 2003).

albatross (Viswanathan *et al* 1996, 1999). As an example, we show typical trajectories of spider-monkeys in the forest of the Mexican Yucatan peninsula in figure 4. In parts (c) and (d) of this figure, a zoom into the trajectory reveals a self-similar behaviour. Statistical analysis reveals a Lévy walk with an exponent in the mean squared displacement (1) of magnitude $\alpha \approx 1.7$ (Ramos-Fernandez *et al* 2003).

2.3. Small and large: other systems with anomalous dynamics

In subrecoil laser cooling, ‘velocity selective coherent population trapping’ leads to a broadly distributed waiting time of particles close to zero momentum, the Lévy stable nature of which can, in principle, be measured. Moreover, its dynamical description exactly leads to a Riemann–Liouville fractional operator (Kondrashin *et al* 2002, Schaufler *et al* 1999a, 1999b, Bardou *et al* 2002). Similarly, anomalous diffusion occurs in optical lattices (Lutz 2003). Power-law statistics were observed for the histograms of on- and off-times in single quantum dots (Shimizu *et al* 2001), see figure 5. Signatures of Lévy statistics were impressively

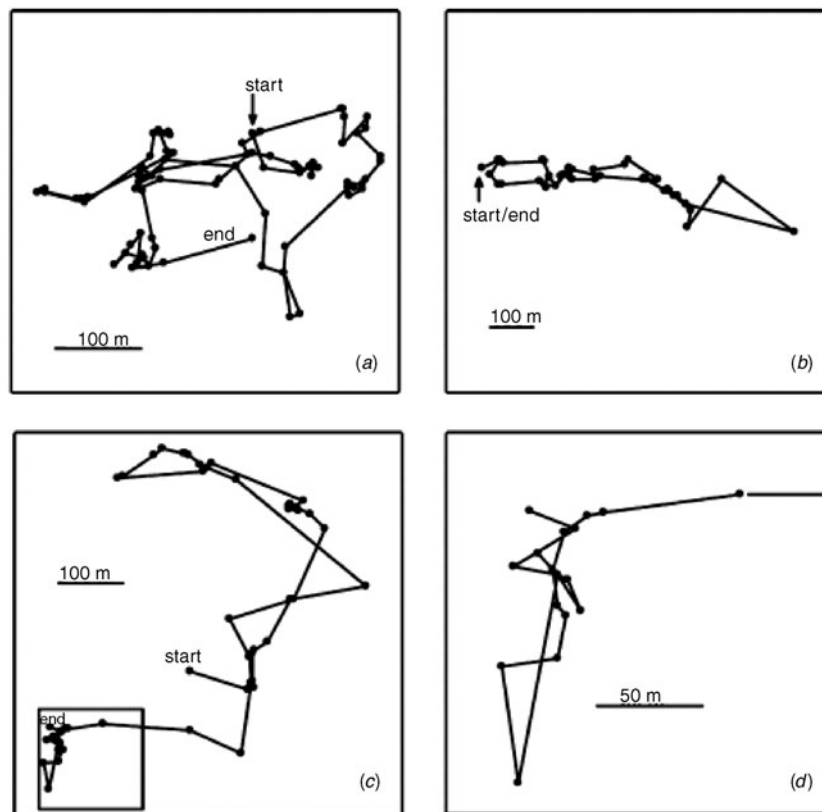


Figure 4. Daily trajectories of adult female (a), (b) and male (c) spider monkeys. In panel (d), a zoom into the inset of (c) is shown (Ramos-Fernandez *et al* 2003).

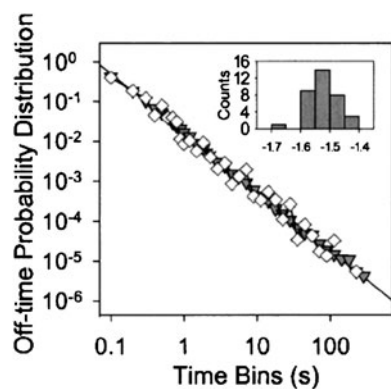


Figure 5. Off-time statistics of blinking (on-off cycles) quantum dots, exhibiting power-law statistics over several decades. For details see Shimizu *et al* (2001).

documented in the study of the position of a single ion in a one-dimensional optical lattice, in which diverging fluctuations could be observed in the kinetic energy (Katori *et al* 1997). Lévy statistics have been identified in random single-molecule line shapes in glass-formers (Barkai *et al* 2000a, 2003). Already in Kenty (1932) it was concluded that in radiation

diffusion the rapidity of escape of resonance radiation from a gas leads to anomalous statistics according to which the fraction of emitted quanta traversing at least a given distance before absorption decays approximately linearly with the distance. Classical intermittency, expressed in terms of continuous time random walks and the fractional Fokker–Planck equation for LFs can be related to the quantum Anderson transition (García-García 2003), and LF signatures were proposed to underlie the fracton excitations in certain ‘unconventional’ superconductors (Milovanov and Rasmussen 2002).

Lévy-type random walks were recognized in the evolution of comets from the Oort cloud (Zhou *et al* 2002, Zhou and Sun 2001), and anomalous diffusion was diagnosed in the cosmic ray spectrum (Lagutin and Uchaikin 2003). It has recently been argued that the terrestrial temperature anomalies are inherited through a Lévy walk memory component from intermittent solar flares (Scafetta and West 2003). From radio signals received from distant pulsars, it has been proposed that the interstellar electron density fluctuations obeys Lévy statistics (Boldyrev and Gwinn 2003). A non-linear fractional equation was proposed for the kinetic description of turbulent plasma and fields at the nonequilibrium stationary states of the magnetotail of Earth (Milovanov and Zelenyi 2002, 2001). Finally, ion motion along the direction normal to the magnetopause has been diagnosed to be of Lévy walk nature (Greco *et al* 2003).

Anomalous diffusion was proposed to account for the hydrogen effect on the morphology of silicon electrodes under electrochemical conditions (Goldar *et al* 2001), as well as in the context of non-linear electrophoresis (Baskin and Zilberstein 2002). Fractional analysis tools were applied in the analysis of anomalous diffusion patterns found in amorphous electroactive materials (Bisquert *et al* 2003, Bisquert 2003). Anomalous diffusion of cations was found as the mechanism in the growth of surface molybdenum oxide patterns (Lugomer *et al* 2002), and similarly the electron transfer kinetics in PEDOT⁸ films (Randriamahazaka *et al* 2002) and atomic transport and chemical reaction processes in high-*k* dielectric films (de Almeida and Baumvol 2003).

Fractional dynamics may underlie the statistics of the joint velocity–position PDF of a single particle in turbulent flow (Friedrich 2003). A fractional generalization of Richardson’s law was proposed for the description of water transport in unsaturated soils (Pachevsky *et al* 2003). Lévy-type PDFs of particle velocities in soft-mode turbulence were studied in electroconvection (Tamura *et al* 2002). From a phenomenological point of view, LFs have been used to describe the dynamics observed in plasmas (Chechkin *et al* 2002b, Gonchar *et al* 2003, Bakunin 2003), or in molecular collisions (Carati *et al* 2003). Stochastic collision models and their natural relation to Lévy velocity laws are discussed in Barkai (2003a). A fractional diffusion approach to the force distribution in static granular media was brought forth recently (Vargas *et al* 2003). Anomalous diffusion properties of heat channels have been investigated in Denisov *et al* (2003) and Reigada *et al* (2002)⁹. Surface growth under certain circumstances requires a generalization of the classical Kardar–Parisi–Zhang model. Recent discussion involves a space-fractional KPZ equation (Katzav 2003, Mann and Woyczynski 2001); compare the discussion of anomalous surface diffusion in Naumovets and Zhang (2002) and Vega *et al* (2002), fractal growth (Leith 2003), and of travelling fronts in the presence of non-Markovian processes (Feodotov and Mendez 2000).

Dielectric susceptibilities in glassy systems are of strong non-Debye form (compare Déjardin 2003, Metzler and Klafter 2002) and can in some systems be studied over some 15 decades in frequency¹⁰ (Hilfer 2002a, 2002b, Schneider *et al* 1999, Lunkenheimer and Loidl

⁸ Poly-3,4-ethylenedioxythiophene.

⁹ Also compare to Li and Wang (2003) and the Comment on that paper (Metzler and Sokolov 2004).

¹⁰ Similar to master curves from rubbery systems (Glöckle and Nonnenmacher 1991, Metzler *et al* 1995).

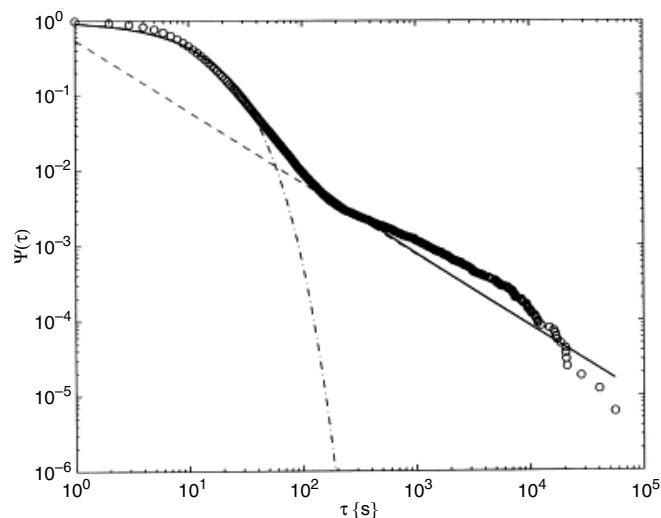


Figure 6. Survival probability for BUND futures from September 1997. The Mittag-Leffler function (full line) is compared with a stretched exponential (dashed-dotted line) and a power law (dashed line) (Mainardi *et al* 2000).

2002), and by NMR both subdiffusion in percolation clusters and Lévy walks in porous media have been verified (Kimmich 2002, Stapf 2002, 1995). In Klemm *et al* (2002), the PDF of fractional diffusion is shown to account for the measured, projected self-diffusion profiles on a fractal percolation structure. $1/f$ -noise and correlated intermittent behaviour were reported from molecular dynamics simulations of water freezing (Matsumoto *et al* 2002).

In economical contexts, it has been revealed that Lévy statistics are present in the distribution of trades (Mandelbrot 1963, 1966, 1967b, Mantegna and Stanley 1996, 2000, Bouchaud and Potters 2000, Matia *et al* 2002). Similarly, it was shown that ‘fat tails’ appear in the return of the value of a given asset to a fixed level (Jensen *et al* 2003, Simonsen *et al* 2002). For the waiting time between two transactions power-law statistics were observed (Kim and Yoon 2003, Raberto *et al* 2002, Scalas *et al* 2000, Mainardi *et al* 2000). Figure 6 shows a Mittag-Leffler fit to the BUND futures traded in September 1997, in which the initial ≈ 2.5 decades in time are nicely fitted by the Mittag-Leffler function (which has a point of inflection on the log–log scale for index larger than $1/2$, afterwards the data appear to oscillate around the Mittag-Leffler trend).

Finally, we note that ageing in glasses and other disordered systems (Monthus and Bouchaud 1996, Rinn *et al* 2000, Pottier 2003) as well as in dynamical systems (Barkai and Cheng 2003, Barkai 2003b) involves power-law non-locality in time; compare the discussion in Sokolov *et al* (2001) and Allegrini *et al* (2003).

3. Subdiffusive processes

Subdiffusive dynamics is characterized by strong memory effects on the (fluctuation-averaged) level of the PDF $P(x, t)$, i.e., unlike in a Markov process the now-state of the system depends on the entire history from its preparation (Barkai 2001, Hughes 1995, Metzler and Klafter 2000a). This contrasts generalized Langevin equations, whose fluctuation average produces equations for $P(x, t)$, which carry time-dependent transport coefficients but are local in time (Wang *et al* 1994, Wang and Tokuyama 1999, Lutz 2001b, Bazzani *et al* 2003). Subdiffusion

is classically described in terms of the CTRW (see the appendix) with a long-tailed waiting time PDF of the asymptotic form

$$\psi(t) \sim \tau^\alpha / t^{1+\alpha} \quad 0 < \alpha < 1 \quad (2)$$

for $t \gg \tau$. In fact, subdiffusive processes are directly subordinated to their analogous Markovian system through a waiting time PDF $\psi(t)$ of the above form. Such waiting times are distinguished by the divergence of the characteristic waiting time, $\mathfrak{T} = \int_0^\infty \psi(t)t \, dt = \infty$, and they reflect the existence of deep traps, which subsequently immobilize the diffusing particle. The seminal case study for such processes is amorphous semiconductors (Pfister and Scher 1977, 1978, Scher and Montroll 1975).

3.1. Fractional diffusion equation

From expression (2), we can immediately obtain the equation for $P(x, t)$ in the force-free case. To this end, we combine the long-tailed $\psi(t)$ with a short-range jump length PDF $\lambda(x)$ and the known expression (A.1) for the PDF $P(x, t)$ in the continuous time random walk model (see Metzler and Klafter 2000a and the appendix). With the asymptotic behaviour

$$\psi(u) \equiv \mathcal{L}\{\psi(t); u\} = \int_0^\infty \psi(t) \exp(-ut) \, dt \sim 1 - (u\tau)^\alpha \quad (3)$$

of the Laplace transform $\mathcal{L}\{\psi(t); u\}$ of $\psi(t)$, and the analogous expansion of a typical, short-range jump length PDF, $\lambda(k) \sim 1 - \sigma k^2$ ($k \rightarrow 0$) for the Fourier transform of $\lambda(x)$, we obtain

$$P(k, u) \simeq \frac{1/u}{1 + u^{-\alpha} K_\alpha k^2} \quad (4)$$

where we identified the anomalous diffusion constant as $K_\alpha \equiv \sigma/\tau^\alpha$. By the symbol \simeq we indicate that the result for $P(k, u)$ is based on expansions for $\psi(u)$ and $\lambda(k)$. However, similar to the limit in going from the master equation to the continuum limit, we can choose τ and σ small enough (keeping K_α finite), such that $P(k, u)$ essentially covers the entire time–space range. In this sense, we will drop the \simeq sign in the following.

In the Brownian limit $\alpha = 1$, expression (4) after multiplication by the denominator leads to the standard diffusion equation $\partial P(x, t)/\partial t = K_1 \partial^2 P(x, t)/\partial x^2$, making use of the integral theorem $\mathcal{L}\{\int_0^t f(t) \, dt\} = u^{-1} f(u)$ and the differentiation theorem $\mathcal{F}\{d^2 g(x)/dx^2\} = -k^2 g(k)$ of the Laplace and Fourier transformations, respectively (Wolf 1979). By partial differentiation of the obtained integral equation, the diffusion equation yields. For the subdiffusive case $0 < \alpha < 1$, in contrast, a term of the form $u^{-\alpha} f(u)$ occurs. Its Laplace inversion is indeed feasible, due to the property

$$\mathcal{L}\{{}_0 D_t^{-\alpha} f(t)\} \equiv \int_0^\infty dt e^{-ut} {}_0 D_t^{-\alpha} f(t) = u^{-\alpha} f(u) \quad (5)$$

of the Riemann–Liouville fractional integral,

$${}_0 D_t^{-\alpha} f(t) \equiv \frac{1}{\Gamma(\alpha)} \int_0^t dt' \frac{f(t')}{(t-t')^{1-\alpha}} \quad (6)$$

defined for any sufficiently well-behaved function $f(t)$ (Miller and Ross 1993, Oldham and Spanier 1974, Podlubny 1998, Samko *et al* 1993). Thus, from equation (4) we obtain the fractional diffusion equation in the so-called integral form (Balakrishnan 1985, Schneider and Wyss 1989),

$$P(x, t) - P_0(x) = {}_0 D_t^{-\alpha} K_\alpha \frac{\partial^2}{\partial x^2} P(x, t) \quad (7)$$

where we have written a general initial condition $P_0(x)$ instead of the δ -condition $P_0(x) = \delta(x)$ corresponding to (4). By partial differentiation and with the Riemann–Liouville fractional differential operator

$${}_0D_t^{1-\alpha} \equiv \frac{\partial}{\partial t} {}_0D_t^{-\alpha} \quad (8)$$

we arrive at the usual form of the fractional diffusion equation

$$\frac{\partial}{\partial t} P(x, t) = {}_0D_t^{1-\alpha} K_\alpha \frac{\partial^2}{\partial x^2} P(x, t). \quad (9)$$

The solution of this equation can be obtained in closed form in terms of the Fox H -function (Hilfer 1995, Metzler and Klafter 2000a, 2000d, Schneider and Wyss 1989). Moreover, due to the definition of this H -function as a Mellin–Barnes integral, the spectral functions of $P(x, t)$ such as $P(k, t)$, $P(k, \omega)$ etc can be obtained in closed form, as well (Metzler and Nonnenmacher 1997). As we are mainly interested in processes in the presence of an external force field, we only stop to note that the asymptotic behaviour of the propagator $P(x, t)$ of the fractional diffusion equation (9) corresponds to the stretched Gaussian shape listed in table 1. The PDF of such a subdiffusive diffusion process has a softer decay than that of normal diffusion. In return, the Fourier transform, $P(k, t)$, bears asymptotic power-law decay characteristics of a Lévy stable law (Metzler and Klafter 2000a, Metzler and Nonnenmacher 1997).

We should point out that it is important to keep track of the initial condition in the fractional diffusion equation (9). Thus, one can by the standard property ${}_0D_t^\alpha 1 = t^{-\alpha} / \Gamma(1 - \alpha)$ of the Riemann–Liouville operator retrieve the equivalent equation to (9) in the form

$${}_0D_t^\alpha P(x, t) - \frac{P_0(x)}{\Gamma(1 - \alpha)} t^{-\alpha} = K_\alpha \frac{\partial^2}{\partial x^2} P(x, t). \quad (10)$$

Neglecting the initial condition would lead to a wrong equation, as can easily be seen by calculating the average on both sides.

We can now also compare the memory form of equation (9) with the dynamical equation for FBM (Lutz 2001b),

$$\frac{\partial}{\partial t} P_{\text{FBM}}(x, t) = \alpha K_\alpha t^{\alpha-1} \frac{\partial^2}{\partial x^2} P_{\text{FBM}}(x, t) \quad (11)$$

which is perfectly local in time. This equation for FBM can be derived from the force-free GLE (Lutz 2001b)

$$m \frac{d^2}{dt^2} x(t) + m \eta_\alpha {}_0D_t^\alpha x(t) = \Gamma(t) \quad (12)$$

where $\Gamma(t)$ is Gaussian random noise with variance $\langle \Gamma(t)\Gamma(0) \rangle \propto t^{-\alpha}$. This GLE then also gives rise to Mittag-Leffler-type correlation functions, see Lutz (2001b) for more details.

3.2. Fractional Fokker–Planck equation

The incorporation of an external force can be achieved by choosing an explicitly space-dependent form for the jump length PDF, such that one can account for the spatial inhomogeneity due to a general force field $F(x) = -dV(x)/dx$. From this model one infers the fractional Fokker–Planck equation, as detailed in Barkai *et al* (2000b) and Metzler *et al* (1999b). However, here we prefer to present a somewhat more fundamental derivation leading to a fractional Fokker–Planck equation in phase space.

To this end, we come back to the idea of interpreting the subdiffusion process as a subordination to a Brownian process, in the following sense. This subordination is intuitively

described by the adjunct microscopic multiple trapping process. As detailed in Metzler and Klafter (2000b, 2000c) and Metzler (2000), based on the continuous time version of the Chapman–Kolmogorov equation, the motion events in this multiple trapping picture are based on a regular, Markovian random walk process, governed through the Langevin equation (Langevin 1908, Chandrasekhar 1943, van Kampen 1981)

$$m \frac{d^2x}{dt^2} = -\eta m \frac{dx}{dt} + F(x) + m\Gamma(t) \quad (13)$$

where $\Gamma(t)$ denotes a δ -correlated Gaussian noise, i.e., $\overline{\Gamma(t)\Gamma(t')} = 2K\delta(t-t')$, and the noise characteristic function is $\varphi(k) = \int_{-\infty}^{\infty} \exp(ik\Gamma)p(\Gamma) d\Gamma = \exp(-Kk^2)$.¹¹ Each motion event governed through the Langevin equation (13) is supposed to last an average time span τ^* , and each such single motion event is interrupted by immobilization (trapping) of a duration governed by the broad waiting time PDF (2). Averaging over many such motion–immobilization events, one obtains the multiple trapping scenario leading to subdiffusion in the external field $F(x)$ (Metzler and Klafter 2000b, 2000c, Metzler 2000), which may be viewed as a direct consequence of the generalized central limit theorem (Gnedenko and Kolmogorov 1954, Lévy 1954). In the Markov limit, the waiting time PDF $\psi(t)$ possesses a finite characteristic waiting time \mathfrak{T} , and may for instance be given by the exponential $\psi(t) = \tau^{-1} \exp(-t/\tau)$, or a sharp distribution such as $\psi(t) = \delta(t-\tau)$. Note that in Laplace space, $\psi(u) \simeq 1 - (u\tau)^\alpha$ ($u\tau \ll 1$), both subdiffusive ($0 < \alpha < 1$) and Markov ($\alpha = 1$) limits appear unified.

In phase space spanned by velocity v and position x , a test particle governed by the above multiple trapping process is described in terms of the fractional Klein–Kramers equation (FKKE)

$$\frac{\partial P}{\partial t} = {}_0D_t^{1-\alpha} \left(-v^* \frac{\partial}{\partial x} + \frac{\partial}{\partial v} \left(\eta^* v - \frac{F^*(x)}{m} \right) + \frac{\eta^* k_B T}{m} \frac{\partial^2}{\partial v^2} \right) P(x, v, t) \quad (14)$$

with the abbreviations $v^* \equiv v\tau^*/\tau^\alpha$, $\eta^* \equiv \eta\tau^*/\tau^\alpha$ and $F^*(x) \equiv F(x)\tau^*/\tau^\alpha$ (Metzler and Klafter 2000b, 2000c, Metzler 2000). Note that the Stokes operator $\left(\frac{\partial}{\partial t} + v \frac{\partial}{\partial x}\right)$ from the standard Klein–Kramers equation (Chandrasekhar 1943) is replaced by the operator $\left(\frac{\partial}{\partial t} + {}_0D_t^{1-\alpha} v^* \frac{\partial}{\partial x}\right)$ which shows the non-local drift response due to trapping.

For both the Langevin equation (13) and the FKKE (14) one can consider the under- (velocity equilibration) and overdamped (large friction constant) limits. The former limit corresponds to the fractional version of the Rayleigh equation (van Kampen 1981),

$$\frac{\partial P}{\partial t} = {}_0D_t^{1-\alpha} \eta^* \left(\frac{\partial}{\partial v} v + \frac{k_B T}{m} \frac{\partial^2}{\partial v^2} \right) P(v, t) \quad (15)$$

in the force-free limit (Metzler and Klafter 2000b, 2000c). This is the subdiffusive generalization of the Ornstein–Uhlenbeck process, see also below. Conversely, in the overdamped case, the FKKE (14) corresponds in position space to the fractional Fokker–Planck equation (FFPE) (Metzler *et al* 1999a, 1999b, Metzler and Klafter 2000b, 2000c)

$$\frac{\partial P}{\partial t} = {}_0D_t^{1-\alpha} \left(-\frac{\partial}{\partial x} \frac{F(x)}{m\eta_\alpha} + K_\alpha \frac{\partial^2}{\partial x^2} \right) P(x, t), \quad (16)$$

where $\eta_\alpha \equiv \eta\tau^\alpha/\tau^*$ and $K_\alpha \equiv k_B T/(m\eta_\alpha)$. We note that all fractional equations (14)–(16) reduce to their Markov counterparts in the limit $\alpha \rightarrow 1$, which can be seen from both the reduction of the multiple trapping process to the regular random walk, and the properties of the Riemann–Liouville fractional operator. We also note that in the general case of $0 < \alpha < 1$,

¹¹ We denote fluctuation averages by an overline, $\bar{\cdot}$, and coordinate averages by angular brackets, $\langle \cdot \rangle$.

initial conditions are strongly persistent due to the slow decay of the sticking probability of not moving $\phi(t) = 1 - \int_0^t \psi(t) dt$, i.e., one observes characteristic cusps at the location of a sharp initial PDF, e.g., $P(x, 0) = \delta(x - x_0)$; compare figure 8 and Metzler and Klafter (2000a) for more details. The fractional equations following from the multiple trapping model with broad waiting time PDF give rise to a generalized Einstein–Stokes relation

$$K_\alpha = k_B T / (m \eta_\alpha) \quad (17)$$

and fulfil linear response in the presence of a constant field F_0 (Metzler *et al* 1999a, Metzler and Klafter 2000a):

$$\langle x(t) \rangle_{F_0} = \frac{k_B T}{2} \langle x^2(t) \rangle_{F=0}. \quad (18)$$

The calculation of moments from fractional equations of the FFPE (16) kind can be straightforwardly obtained by multiplying the dynamical equation with the moment variable and integration over the coordinate, e.g., calculating $\int x^m \cdot dx$ where \cdot acts on the dynamical equation (Metzler and Klafter 2000a). More-point correlation functions are somewhat more difficult to obtain due to the strongly non-local character in time. Three-point correlation functions have recently been obtained on the basis of the FFPE by introducing the associated backward equation (Barsegov and Mukamel 2004).

Fractional equations of the above linear, uncoupled kind can be solved by the method of separation of variables. Thus, for instance, the FFPE (16) can be separated through the ansatz $P(x, t) = X(x)T(t)$ to produce a spatial eigenequation, which has the same structure as its Markov analogue, and a temporal eigenequation,

$$\frac{dT_n(t)}{dt} = -\lambda_n {}_0D_t^{1-\alpha} T_n(t) \quad (19)$$

for a given eigenvalue λ_n (Metzler *et al* 1999a, Metzler and Klafter 2000a). Its solution yields in terms of the Mittag-Leffler function (Mittag-Leffler 1903, 1904, 1905, Erdélyi 1954)

$$E_\alpha(-\lambda_n t^\alpha) \equiv \sum_{j=0}^{\infty} \frac{(-\lambda_n t^\alpha)^j}{\Gamma(1 + \alpha j)} \sim \begin{cases} \exp\left(-\frac{\lambda_n t^\alpha}{\Gamma(1+\alpha)}\right) & t \ll \lambda^{1/\alpha} \\ (\lambda_n t^\alpha \Gamma(1 - \alpha))^{-1} & t \gg \lambda^{1/\alpha} \end{cases} \quad (20)$$

where we also indicated the interpolation property of the Mittag-Leffler function, connecting between an initial stretched exponential (KWW) pattern and a terminal inverse power-law behaviour (Metzler and Klafter 2000a, Glöckle and Nonnenmacher 1991, 1994); compare figure 7. As for a non-trivial external field $F(x)$, the lowest eigenvalue vanishes, $\lambda_1 = 0$, and thus $0 < \lambda_2 < \dots$, the PDF $P(x, t)$ relaxes towards the equilibrium solution given by the lowest eigenvalue λ_1 which is identical to the Boltzmann solution and fulfils the stationarity condition $\partial P(x, t) / \partial t = 0$ (Metzler *et al* 1999a, Metzler and Klafter 2000a). Finally, we note that there exists a Laplace space scaling relation (Metzler *et al* 1999a, Metzler and Klafter 2000a)

$$P(x, u) = \frac{1}{u} \frac{\eta_\alpha u^\alpha}{\eta_1} P_M\left(x, \frac{\eta_\alpha u^\alpha}{\eta_1}\right) \quad (21)$$

for the same initial condition $P_0(x)$ between the solution of the FFPE (16), $P(x, t)$, and its Markov counterpart $P_M(x, t)$ ($\alpha = 1$). Equation (20) is equivalent to the integral transformation (Barkai and Silbey 2000),

$$P(x, t) = \int_0^\infty ds E_\alpha(s, t) P_M(x, s) \quad (22)$$

which corresponds to a generalized Laplace transformation from t to $\frac{\eta_\alpha u^\alpha}{\eta_1}$. The kernel $E_\alpha(s, t)$ is defined in terms of the inverse Laplace transformation $E_\alpha(s, t) = \mathcal{L}^{-1}\left\{\frac{\eta_\alpha}{\eta_1 u^{1-\alpha}} \exp\left(-\frac{\eta_\alpha}{\eta_1} u^\alpha s\right)\right\}$, the result being the modified one-sided Lévy distribution¹² $L_{1-\alpha/2}^+$

$$E_\alpha(s, t) = \frac{t}{(1 - \alpha/2)s} L_{1-\alpha/2}^+ \left(\frac{t}{(s^*)^{1/(1-\alpha/2)}} \right) \quad s^* \equiv \eta_\alpha s / \eta_1 \quad (23)$$

which is everywhere positive definite. Consequently, the transformation (22) guarantees the existence and positivity of $P_\alpha(x, t)$ if (and only if) the Brownian counterpart, $P_M(x, t)$, is a proper PDF. We note that the solution of certain classes of fractional equations is intimately related to the Fox H -function and related special functions (Mathai and Saxena 1978, Srivastava *et al* 1982, Saxena and Saigo 2001). Also the kernel $E_\alpha(s, t)$ can be expressed as an H -function (Metzler and Klafter 2000a). It should be noted once more that fractional diffusion in the above-defined Riemann–Liouville sense is fundamentally different from fractional Brownian motion (Mandelbrot and van Ness 1968, Lévy 1953,

¹² In this review, we use symmetric Lévy stable laws with characteristic function $\varphi(z) = \int_{-\infty}^{\infty} e^{ikx - \sigma^\mu |k|^\mu} dk / (2\pi)$. The definition of Lévy laws is more general. Thus, one-sided Lévy stable laws exist, which are defined only on the positive semidefinite axis, i.e., in our case on the causal time line $t \geq 0$. In terms of the general characteristic function φ of a Lévy stable law, defined through

$$\log \varphi(z) = -|z|^\alpha \exp\left(i \frac{\pi\beta}{2} \text{sign}(z)\right)$$

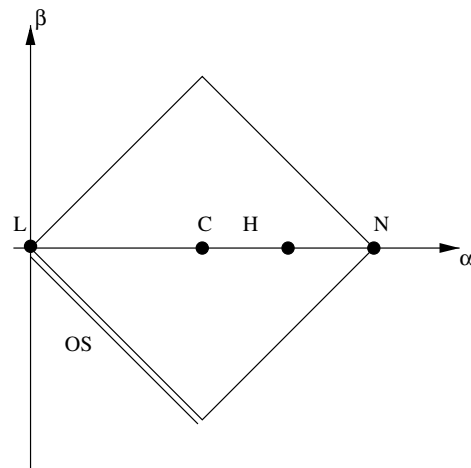
the one-sided laws exist for $0 < \alpha < 1$ and $\beta = -\alpha$. For instance, the one-sided stable law for $\alpha = 1/2$ and $\beta = -1/2$ is given by

$$f_{1/2, -1/2} = \frac{1}{2\sqrt{\pi}} x^{-3/2} \exp(-1/[4x])$$

where, in general, we have

$$f_{\alpha, \beta} \equiv \frac{1}{\pi} \text{Re} \int_0^\infty \exp(-ixz - z^\alpha \exp\{i \frac{\pi\beta}{2}\}) dz$$

The parameter space of Lévy stable laws can be represented by the ‘Takayasu diamond’ (Takayasu 1990):



All pairs of indices inside and on the edge of the diamond shape refer to proper stable laws. The double line denotes one-sided stable laws (OS). The letters represent the normal or Gaussian law (N), the Holtsmark distribution (H) and the Cauchy–Lorentz distribution (C) (Feller 1968, Gnedenko and Kolmogorov 1954, Lévy 1954, Takayasu 1990). We note that the connection of the fractional integral with stable distributions was recently investigated explicitly in (Stanislavsky 2004).

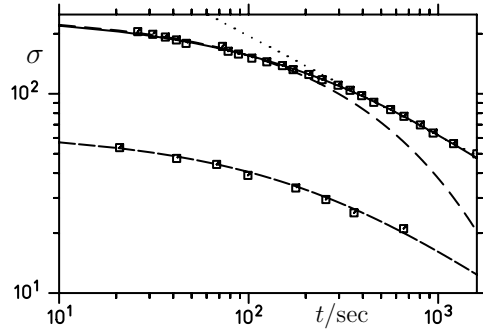


Figure 7. Interpolative nature of the Mittag-Leffler function, in an example from stress relaxation at constant strain (the image shows two different initial conditions). In the upper curve, we compare the Mittag-Leffler function (full line), with the initial stretched exponential and the terminal inverse power-law behaviour. (From Nonnenmacher (1991).)

Lim and Muniandy 2002, Kolmogorov 1940, Lutz 2001b) and generalized Langevin equation approaches (Kubo *et al* 1985, Wang *et al* 1994, Wang and Tokuyama 1999); compare table 1, as well as non-linear (fractional) Fokker–Planck equations (Borland 1998, Lenzi *et al* 2003b, Tsallis and Lenzi 2002).

3.3. The fractional Ornstein–Uhlenbeck process

The Ornstein–Uhlenbeck process corresponds to the motion in a harmonic potential $V(x) = \frac{1}{2}m\omega^2x^2$ giving rise to the restoring force field $F(x) = -m\omega^2x$, i.e., to the dynamical equation

$$\frac{\partial}{\partial t} P(x, t) = \left(\frac{\partial}{\partial x} \frac{\omega^2 x}{\eta_\alpha} + K_\alpha \frac{\partial^2}{\partial x^2} \right) P(x, t). \tag{24}$$

From separation of variables, and the definition of the Hermite polynomials (Abramowitz and Stegun 1972), one finds the series solution for the fractional Fokker–Planck equation with the Ornstein–Uhlenbeck potential (Metzler *et al* 1999a),

$$P(x, t) = \sqrt{\frac{m\omega^2}{2\pi k_B T}} \sum_{n=0}^{\infty} \frac{1}{2^n n!} E_\alpha \left(-\frac{n\omega^2 t^\alpha}{\eta_\alpha} \right) H_n \left(\frac{\sqrt{m\omega} x_0}{\sqrt{2k_B T}} \right) \times H_n \left(\frac{\sqrt{m\omega} x}{\sqrt{2k_B T}} \right) \exp \left(-\frac{m\omega^2 x^2}{2k_B T} \right) \tag{25}$$

plotted in figure 8. Individual position space modes follow the ordinary Hermite polynomials of increasing order, while their temporal relaxation is of Mittag-Leffler form, with decreasing internal time scale $(\eta_\alpha/[n\omega^2])^{1/\alpha}$. Numerically, the solution (25) is somewhat cumbersome to treat. In order to plot the PDF $P(x, t)$ in figure 8, it is preferable to use the closed form solution (we use dimensionless variables)

$$P(x, t) = \frac{1}{\sqrt{2\pi(1 - e^{-2t})}} \exp \left(-\frac{(x - x_0 e^{-t})^2}{2(1 - e^{-2t})} \right) \tag{26}$$

of the Brownian case, and the transformation (22) to construct the fractional analogue.

Figure 8 shows the distinct cusps at the position of the initial condition at $x_0 = 1$. The relaxation to the final Gaussian Boltzmann PDF can be seen from the sequence of three consecutive times. Only at stationarity, the cusp gives way to the smooth Gaussian shape

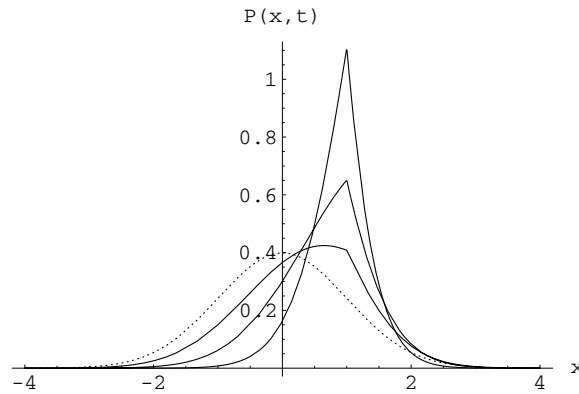


Figure 8. Time evolution of the PDF of the fractional Ornstein–Uhlenbeck process ($\alpha = 1/2$). The initial condition was chosen as $\delta(x - 1)$. Note the strongly persistent cusp at the location of the initial peak. Dimensionless times: 0.02, 0.2, 2. The dashed line corresponds to the Boltzmann equilibrium, to which the PDF relaxes.

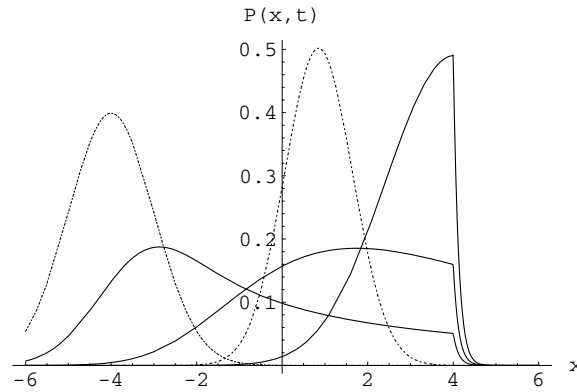


Figure 9. Time evolution of the PDF of the fractional Ornstein–Uhlenbeck process with superposed constant force of dimensionless strength $V = -4$ ($\alpha = 1/2$). The initial condition was chosen as $\delta(x - 4)$. Dimensionless times: 0.02, 0.2, 2. The dashed lines correspond to the Brownian solution at times 0.5 and 50 (in essence, the stationary state). Again, note the cusps due to the initial condition, causing a strongly asymmetric shape of the PDF in contrast to the Gaussian nature of the Brownian counterpart.

of the Boltzmann equilibrium PDF. By adding an additional linear drift V to the harmonic restoring force, the drift term in the FFPE (17) changes to $-\partial(x - V)P(x, t)/\partial x$, and the exponential in expression (26) takes the form $\exp(-[x - V - (x_0 - V)e^{-t}]/[2(1 - e^{-2t})])$. As displayed in figure 9, the strong persistence of the initial condition causes a highly asymmetric shape of the PDF, whereas the Brownian solution shown in dashed lines retains its symmetric Gaussian profile. Let us note again that a generalized Langevin picture would give rise to time-dependent coefficients, but would not change the Gaussian nature of the connected process in the harmonic potential.

Let us finally address the moments of the fractional Ornstein–Uhlenbeck process, equation (25). These can be readily obtained either from the Brownian result with the integral

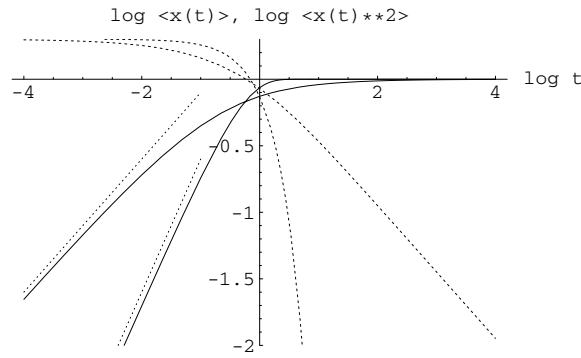


Figure 10. First ($x_0 = 2$, dashed line) and second ($x_0 = 0$, full line) moment of the fractional Ornstein-Uhlenbeck process ($\alpha = 1/2$), in comparison to the Brownian case. \log_{10} - \log_{10} scale. The dotted straight lines show the initial (sub)diffusive behaviour with slopes $1/2$ and 1 , in the special case $x_0 = 0$ chosen for the second moment.

transformation (22), or from integration $\int dx x^n$ of the FFPE (17). For the first and second moments one obtains

$$\langle x(t) \rangle = x_0 E_\alpha \left(-\frac{\omega^2 t^\alpha}{\eta_\alpha} \right) \tag{27}$$

and

$$\langle x(t)^2 \rangle = x_{\text{th}}^2 + (x_0^2 - x_{\text{th}}^2) E_\alpha \left(-\frac{2\omega^2 t^\alpha}{\eta_\alpha} \right) \tag{28}$$

respectively. The first moment starts off at the initial position, x_0 , and then falls off in a Mittag-Leffler pattern, reaching the terminal inverse power law $\sim t^{-\alpha}$. The second moment turns from the initial value x_0^2 to the thermal value $x_{\text{th}}^2 = k_B T / (m\omega^2)$. In the special case $x_0 = 0$, the second moment measures initial force-free diffusion due to the initial exploration of the flat apex of the potential. We graph the two moments in figure 10 in comparison to their Brownian counterparts.

We note in passing that by optical tracking methods, it is, in principle, possible to obtain precise results for the Gaussian PDF of a single random walker in the equilibrium state, as demonstrated by Oddershede *et al* (2002). It should therefore be possible to obtain more information also on anomalous processes than through measurements of the mean squared displacement alone (particularly, due to the slow power-law relaxation of FFPE-governed processes it might be possible to monitor transient PDFs during the relaxation towards the Boltzmann equilibrium). We also note that for a particle connected to a membrane and experiencing in addition optical tweezers potential, the relaxation dynamics is closely related to the Mittag-Leffler decay (Granek and Klafter 2001). Finally, we mention that the fractional Ornstein-Uhlenbeck process was investigated from the point of view of a time-dependent potential in Tofighi (2003).

3.4. Fractional diffusion equations of distributed order

There exist physical systems with the so-called ultraslow diffusion of the logarithmic form

$$\langle x^2(t) \rangle \sim \log^\kappa t \quad \kappa > 0 \tag{29}$$

such as the famed Sinai diffusion ($\kappa = 4$) of a particle moving in a quenched random force field (Sinai 1982), the motion of a polyampholyte hooked around an obstacle (Schiesse *et al*

1997), and similarly in aperiodic environments (Igloi *et al* 1999), in a family of iterated maps (Dräger and Klafter 2000), as well as in a parabolic map (Prosen and Žnidarič 2001).

Within the continuous time random walk theory, such ‘strong anomalies’ (Dräger and Klafter 2000) can be described in terms of a waiting time PDF of the form (Havlin and Weiss 1990)

$$\psi(t) \sim \frac{\tau}{t \log^{\kappa+1}(t/\tau)}. \quad (30)$$

Obviously, the characteristic waiting time \mathfrak{T} for this $\psi(t)$ diverges, although it is normalized. The corresponding propagator exhibits asymptotic exponential flanks of the form

$$P(x, t) \sim \exp\left(-A \frac{|x|}{\log^{\kappa/2} t}\right). \quad (31)$$

For such strongly anomalous processes running off under the influence of an external potential, one would again like to have a description in terms of a dynamical equation. In fact, on the basis of distributed-order fractional operators (Caputo 1969, 2001, Chechkin *et al* 2003d), the fractional equation

$$\int_0^1 \tau^{\beta-1} p(\beta) {}_0D_t^\beta P(x, t) = \left(\frac{\partial}{\partial x} \frac{V'(x)}{m\eta_{\text{do}}} + K_{\text{do}} \frac{\partial^2}{\partial x^2} \right) P(x, t) \quad (32)$$

was shown to lead to the desired logarithmic behaviour (30) in the force-free limit and with $p(\beta) = \kappa\beta^{\kappa-1}$ (Chechkin *et al* 2002c). Note that in the generalized Fokker–Planck operator we include properly generalized units of friction and diffusion coefficient.

The model equation (32), by construction, controls system relaxation towards the Boltzmann equilibrium, and fulfils the generalized Einstein–Stokes relation $K_{\text{do}} = k_B T / (m\eta_{\text{do}})$. Moreover, one can show that it fulfils the linear response behaviour (18). We note that the mode relaxation, which is of Mittag-Leffler nature in the case of the regular (non-distributed) FFPE (16), for the distributed order case includes a logarithmic time dependence (Chechkin *et al* 2002c)¹³.

4. Superdiffusive processes

Subdiffusive processes of the above kind can be physically understood in terms of the subordination to the corresponding Markov process, immanent in the multiple trapping model with long-tailed waiting time PDF of the form (2). The solution corresponds to re-weighting of the Brownian solution with a sharply peaked kernel. In particular, the obtained PDFs relax towards the Boltzmann equilibrium, and they possess all moments if only the Brownian counterpart does (i.e., constant or confining potentials). Thus, the presence of the diverging characteristic waiting times does not change the quality (basin of attraction in a generalized central limit theorem sense) of the process in position (x) space. In contrast, we will show in this section that for random processes with non-local jump lengths of the Lévy type, *a priori* surprising multimodal PDFs may arise and one observes a breakdown of the method of images. If the external potential is not steep enough, the moments diverge. Questions about the physical and thermodynamic interpretation of such processes arise. These points are addressed in the following. We will first introduce the concept of Lévy flights (LFs) and discuss their formulation in terms of fractional equations. We then proceed to elaborate on some details concerning the above-mentioned surprising features of LFs, before briefly addressing first results of a dynamic formulation of Lévy walks (LWs), the spatiotemporally coupled version of superdiffusive random processes, and their fractional formulation.

¹³ We should stress that the physics of equation (32) differs from Sinai diffusion, cf Chechkin *et al* (2002c).



Figure 11. Lévy flight (right) of index $\mu = 1.5$ and Gauss walk (left) trajectories with the same number ($\simeq 7000$) of steps. The long sojourns and clustering appearance of the LF are distinct.

4.1. Lévy flights

LFs are Markov processes with broad jump length distributions with the asymptotic inverse power-law behaviour

$$\lambda(x) \sim \frac{\sigma^\mu}{|x|^{1+\mu}} \quad (33)$$

such that its variance diverges, $\mathfrak{X}^2 = \int_{-\infty}^{\infty} \lambda(x)x^2 dx = \infty$. This scale-free¹⁴ form gives rise to the characteristic trajectories of LFs as shown in figure 11: in contrast to the ‘area-filling’ nature of a regular (Gaussian) random walk, an LF has a fractal dimension with exponent μ (Blumenthal and Gettoor 1960, Hughes 1995, Rocco and West 1999), and consists of a self-similar clustering of local sojourns, interrupted by long jumps, at whose end a new cluster starts, and so on. This happens on all length scales, i.e., zooming into a cluster in turn reveals clusters interrupted by long sojourns. Thus, LFs intimately combine the local jump properties stemming from the centre part of the jump length distribution around zero jump length with strongly non-local, i.e., long-distance jumps, thereby creating slowly decaying spatial correlations, a signature of non-Gaussian processes with diverging variance (Hughes 1995, Bouchaud and Georges 1990, Lévy 1954, van Kampen 1981). Of course, also the Gaussian trajectory is self-similar, however, its finite variance prohibits the existence of long jumps separating local clusters.

Following along the lines pursued in the case of force-free subdiffusion, we describe LFs with a sharply peaked waiting time PDF $\psi(t)$ ($\alpha = 1$) with finite characteristic waiting time \mathfrak{T} and $\psi(u) \sim 1 - u\tau$. The Fourier transform $\lambda(k) = \exp(-\sigma^\mu |k|^\mu) \sim 1 - \sigma^\mu |k|^\mu$ of a Lévy stable jump length PDF $\lambda(x)$ with asymptotic form (33) by means of expression (A.1) produces a dynamical equation in Fourier–Laplace space, in which occurs the expression $|k|^\mu P(k, u)$ instead of the standard term $k^2 P(k, u)$ in Gaussian diffusion. Let us for the moment define the fractional derivative in space through $\mathcal{F}\left\{\frac{d^\mu g(x)}{d|x|^\mu}\right\} \equiv -|k|^\mu g(k)$ for $1 \leq \mu < 2$,¹⁵ such that we infer the Lévy fractional diffusion equation (Compte 1996, Fogedby 1994a, Honkonen 1996, Saichev and Zaslavsky 1997)¹⁶

$$\frac{\partial}{\partial t} P(x, t) = K^\mu \frac{\partial^\mu}{\partial |x|^\mu} P(x, t) \quad (34)$$

where we define in the analogous sense as above the generalized diffusion constant $K^\mu \equiv \sigma^\mu / \tau$ of (formal) dimension $\text{cm}^\mu \text{s}^{-1}$.¹⁷ Again, equation (34) can be solved in closed form in

¹⁴ In the sense that there does not exist a variance of the jump length distribution.

¹⁵ We do not pursue the case $0 < \mu < 1$ in what follows, although it follows the same reasoning.

¹⁶ Note that here we differ from our previous notation ${}_{-\infty}D_x^\mu$ used in Metzler and Klafter (2000a). We follow here the convention which seems to have become a standard for the space-fractional case, the Riesz–Weyl operators.

¹⁷ Nb: the waiting time PDF has the Laplace transform $\psi(u) \sim 1 - u\tau$ for this Markovian case.

terms of Fox H -functions (Jespersen *et al* 1999, Metzler and Klafter 2000a). Let us show that indeed equation (34) defines a Lévy stable law: Fourier transforming leads to the equation $\partial P(k, t)/\partial t = -|k|^\mu K^\mu P(k, t)$, which is readily integrated to yield $P(k, t) = \exp(-K^\mu |k|^\mu t)$, the characteristic function of a Lévy stable law (Gnedenko and Kolmogorov 1954, Lévy 1954). From the fractional operator (defined below) in equation (34), the symmetric, strongly non-local character of LFs becomes obvious. LFs were originally described by Mandelbrot, and formally the Fourier space analogue of equation (34) was discussed in Seshadri and West (1982) on the basis of a Langevin equation with Lévy noise, see below.

We note that due to the Markovian nature of LFs, a constant force/velocity V can immediately be incorporated in terms of a moving wave variable, i.e., the solution of the LF in the presence of the drift V defined by the equation

$$\frac{\partial}{\partial t} P(x, t) = \left(\frac{\partial}{\partial x} V + K^\mu \frac{\partial^\mu}{\partial |x|^\mu} \right) P(x, t) \quad (35)$$

is the solution $P_{V=0}(x, t)$ of equation (34) taken at position $x - Vt$, i.e., $P_V(x, t) = P_{V=0}(x - Vt, t)$ (Jespersen *et al* 1999, Metzler and Compte 2000, Metzler and Klafter 2000a).

4.1.1. Lévy fractional Fokker–Planck equation. LFs in the presence of an external potential $V(x) = -\int^x F(x') dx'$ are described in terms of a different fractional Fokker–Planck equation, which we will call the Lévy fractional Fokker–Planck equation (LFFPE) in the following. It has the simple form (Fogedby 1994a, 1994b, 1998, Peseckis 1987)

$$\frac{\partial P}{\partial t} = \left(-\frac{\partial}{\partial x} \frac{F(x)}{m\eta} + K^\mu \frac{\partial^\mu}{\partial |x|^\mu} \right) P(x, t) \quad (36)$$

where we encounter the fractional Riesz derivative defined through (Podlubny 1998, Samko *et al* 1993)

$$\frac{d^\mu f(x)}{d|x|^\mu} = \begin{cases} -\frac{\mathbf{D}_+^\mu f(x) + \mathbf{D}_-^\mu f(x)}{2 \cos(\pi\mu/2)} & \mu \neq 1 \\ -\frac{d}{dx} \mathbf{H}f(x) & \mu = 1 \end{cases} \quad (37)$$

$$(\mathbf{D}_+^\mu f)(x) = \frac{1}{\Gamma(2-\mu)} \frac{d^2}{dx^2} \int_{-\infty}^x \frac{f(\xi, t) d\xi}{(x-\xi)^{\mu-1}} \quad (38)$$

and

$$(\mathbf{D}_-^\mu f)(x) = \frac{1}{\Gamma(2-\mu)} \frac{d^2}{dx^2} \int_x^\infty \frac{f(\xi, t) d\xi}{(\xi-x)^{\mu-1}} \quad (39)$$

for, respectively, the left and right Riemann–Liouville derivatives ($1 \leq \mu < 2$); for $\mu = 1$, the fractional operator reduces to the Hilbert transform operator (Mainardi *et al* 2001)

$$(\mathbf{H}f)(x) = \frac{1}{\pi} \int_{-\infty}^\infty \frac{f(\xi) d\xi}{x-\xi}. \quad (40)$$

The Riesz operator has the convenient property

$$\mathcal{F} \left\{ \frac{\partial^\mu}{\partial |x|^\mu} f(x); k \right\} \int_{-\infty}^\infty e^{ikx} \frac{\partial^\mu}{\partial |x|^\mu} f(x, t) dx \equiv -|k|^\mu f(k). \quad (41)$$

It should be noted that according to the LFFPE (36), it is only the diffusive term which is affected by the Lévy noise. In contrast, the character of the drift remains unchanged, i.e., the external force is additive (Fogedby 1994a, 1994b, 1998, Metzler *et al* 1999b, Metzler 2000), as noted above for the case of a constant drift V .

Starting from the Feynman–Vernon path integral formulation of the influence functional (Feynman and Vernon 1963), a characteristic functional, whose classical analogue corresponds to the Caldeira–Leggett equation for quantum Brownian motion, was established. By a Wigner transform for a Lévy source in the influence functional, the following Lévy fractional Klein–Kramers equation emerges (Lutz 2001a)¹⁸:

$$\frac{\partial}{\partial t} P(x, v, t) = -\frac{v}{m} \frac{\partial P}{\partial x} + V'(x) \frac{\partial P}{\partial v} + \frac{\gamma}{m} \frac{\partial}{\partial v} v P + \gamma k_B T \frac{\partial^\mu P}{\partial |v|^\mu}. \quad (42)$$

A similar equation was derived from a Langevin equation with Lévy noise in Peseckis (1987). On the basis of a so-called quantum Lévy process, a Lévy fractional Klein–Kramers equation was obtained through random matrix methods in Kusnezov *et al* (1999), however, it carries a different friction term, as discussed in Lutz (2003). Equation (42) was derived in Metzler (2000) from the generalized Chapman–Kolmogorov equation.

In equation (42), the fractional derivative is attached to the velocity v of the particle. Thus, the corresponding LF-Rayleigh equation corresponds to a Lévy motion in a harmonic potential. As obvious from the result for the Lévy Ornstein–Uhlenbeck process reported in the next subsection, the solution $P(v, t)$ features a diverging mean squared displacement and in fact always remains a Lévy law with the same exponent μ (Jespersen *et al* 1999). In particular, the stationary state is a Lévy stable law of the same index μ (Seshadri and West 1982). We note, however, that this extreme Lévy behaviour is solely due to the linear friction inherent in equation (42). Due to the extremely large velocities attained by a Lévy flyer described by equation (42), i.e., to the divergence of the kinetic energy (Seshadri and West 1982), the linear friction is in fact non-physical, and should be replaced by an expansion of the friction to higher order terms, $\gamma = \gamma(v) = \gamma_0 + \gamma_1 v^2 + \dots$ (Chechkin *et al* 2004). Such a velocity-dependent friction was already discussed by Klimontovich and called dissipative non-linearity (Klimontovich 1991). As we will see below, already the next higher term would produce a finite mean squared displacement of this more physical Lévy process. We also mention that another way to regularize Lévy flights is the spatiotemporal coupling of Lévy walks, which are briefly discussed below. We finally note that the velocity average of equation (42) reduces directly to the LFFPE (36).

Although for the diverging moments, LFs may appear somewhat artificial¹⁹, processes with diverging kinetic energy have been identified (Katori *et al* 1997), and from a physics point of view are permissible in certain connections such as diffusion in energy space (Barkai and Silbey 1999, Zumofen and Klafter 1994), or as a description for the random path (trajectory) of such a random process. Moreover, LFs may be considered paradigmatic in the generalized central limit theorem sense and therefore deserve investigation. Not least, they correspond to approximate schemes to more complex processes, like LWs.

4.1.2. Novel features of Lévy flights in superharmonic potentials. In Jespersen *et al* (1999), it was derived that the solution of the LFFPE in a harmonic potential field, $V(x) = \frac{1}{2}\omega x^2$, in Fourier space takes the form

$$P(k, t) = \exp\left(-\frac{\eta m K^\mu |k|^\mu}{\mu \omega} [1 - e^{-\mu \omega t / (\eta m)}]\right) \quad (43)$$

i.e., it is still a Lévy stable density, with the identical Lévy index μ as in the corresponding solution without external potential, and the stationary solution is $P_{\text{st}}(x) \sim K^\mu \eta m / (\mu \omega |x|^{1+\mu})$,

¹⁸ In fact, the equation derived in Lutz (2001a) also contains a term appearing in the case of asymmetric Lévy laws, which we do not consider herein.

¹⁹ Note, however, that fractional moments $\langle |x|^\delta \rangle$ of order $\delta < \mu$ can be defined, and easily obtained from H -function properties in the case of free LFs (Metzler and Nonnenmacher 2002).

in particular. Thus, a harmonic potential is not able to confine an LF such that its variance becomes finite, pertaining both to the time evolution and the stationary behaviour of the PDF. Equation (43) corresponds to the Lévy analogue of the Ornstein–Uhlenbeck process defined in equation (42) in phase space.

From this perspective, it might seem *a priori* surprising that as soon as the external potential becomes slightly steeper than harmonic, the variance of the underlying LF becomes finite. However, this was demonstrated by Chechkin *et al* (2002a, 2003b, 2003c) both analytically and numerically. We now briefly review the main features connected to such confined LFs. Before addressing these features for general $1 < \mu < 2$, we regard the case of a Cauchy flight ($\mu = 1$) in a quartic potential

$$V(x) = \frac{b}{4}x^4 \quad (44)$$

whose stationary solution is exactly analytically solvable (for the more general cases we will draw on asymptotic arguments corroborated by numerical results). Thus, rewriting equation (36) with the quartic potential (44) in dimensionless coordinates (Chechkin *et al* 2002a),

$$\frac{\partial P}{\partial t} = \left(\frac{\partial}{\partial x}x^3 + \frac{\partial^\mu}{\partial |x|^\mu} \right) P(x, t) \quad (45)$$

one can immediately derive the stationary ($dP_{\text{st}}(x)/dt = 0$) solution

$$P_{\text{st}}(x) = \pi^{-1} \frac{1}{1 - x^2 + x^4} \quad (46)$$

which is remarkable in two respects: (i) the asymptotic power law $P_{\text{st}}(x) \sim x^{-4}$ falls off steeper than the Lévy stable density with index μ , and the variance in fact *converges*, i.e., the process leaves the basin of attraction of the generalized central limit theorem; and (ii) the PDF (46) is *bimodal*, i.e., it exhibits two maxima at $x_{\text{max}} = \pm 1/\sqrt{2}$ (Chechkin *et al* 2002a), see figure 12. This bimodality becomes increasingly pronounced when the Lévy index approaches the Cauchy case, $\mu = 1$ (Chechkin *et al* 2002a). We note that the numerical solution procedures we employ for determining the PDF defined by the LFFPE are mainly based on the Grünwald–Letnikov representation of the fractional Riesz derivative (Podlubny 1998, Gorenflo 1997, Gorenflo *et al* 2002a), details of which are also described in Chechkin *et al* (2003b); see also Lynch *et al* (2003)²⁰.

Let us pursue somewhat further the incapability of a harmonic potential to confine an LF in contrast to a quartic potential and give rise to multimodal states, some of which are transient. To this end, consider the combined potential (Chechkin *et al* 2003c)

$$V(x) = \frac{a}{2}x^2 + \frac{b}{4}x^4. \quad (47)$$

By rescaling the LFFPE (36) with the potential (47) according to $x \rightarrow x/x_0$, $t \rightarrow t/t_0$, with $x_0 \equiv (m\eta D/b)^{1/(2+\mu)}$ and $t_0 \equiv x_0^\mu/D$, and $a \rightarrow at_0/m\eta$, we obtain the normalized form

$$\frac{\partial P}{\partial t} = \left(\frac{\partial}{\partial x}[x^3 + ax] + \frac{\partial^\mu}{\partial |x|^\mu} \right) P(x, t) \quad (48)$$

for the LFFPE (36). A detailed analysis, both analytically and numerically, reveals that there exists a critical magnitude of the relative harmonicity strength a , $a_c \simeq 0.794$, below which a bimodal state exists (Chechkin *et al* 2003c, 2003b).

²⁰ We note that bifurcation to a bimodal state may be obtained from a linear Langevin equation with multiplicative noise term and time-dependent drift (Fa 2003), another, yet different scenario towards multimodal states. Compare also section 4.2.

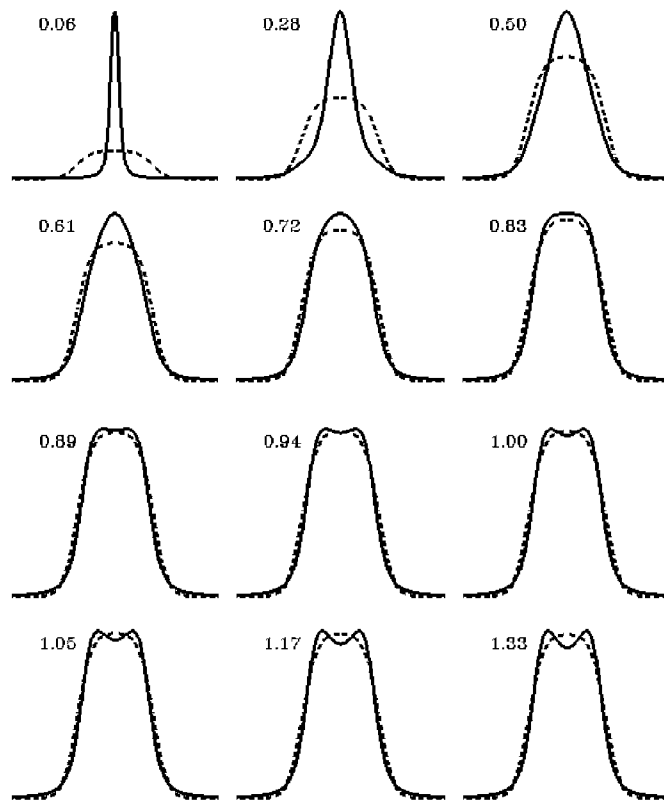


Figure 12. Time evolution of the PDF governed by the LFFPE (36) in a quartic potential, starting from $P(x, 0) = \delta(x)$, with Lévy index $\mu = 1.2$. The dashed line indicates the Boltzmann distribution from the Gaussian process in a harmonic potential.

Dynamically, starting from a monomodal initial condition such as a δ -peak in the centre of the potential, it turns out that there exists a critical time t_c at which the PDF develops bimodality (figure 12). This turnover can be studied similarly as the critical harmonicity strength, and the result is shown in figure 13 at the top: for times $t > t_c$, a bimodal state spontaneously comes into existence, corresponding to a dynamical bifurcation. However, for all potentials $V(x)$ containing a term $\propto |x|^{2+c}$ with $c > 0$, the variance becomes finite for all $t > 0$ (Chechkin *et al* 2002a).

Given the potential

$$V(x) = \frac{a}{c}|x|^c \quad \therefore \quad c \geq 2 \tag{49}$$

there exists an additional, transient trimodal state in the case $c > 4$, an example of which is depicted in figure 14. In this case, the relaxation of the peak of the initial condition overlaps with the building up of the two side-maxima, which will eventually give rise to the terminal bimodal PDF. In figure 13 at the bottom, the size of the maxima and their temporal evolution are shown.

Similar *a priori* surprising effects of LFs were found in periodic potentials, in which LFs turn out to be delicately sensitive. For instance, there has been revealed a rich band structure in the Bloch waves described by an LFFPE and its associated fractional Schrödinger-type equation (Brockmann and Geisel 2003a).

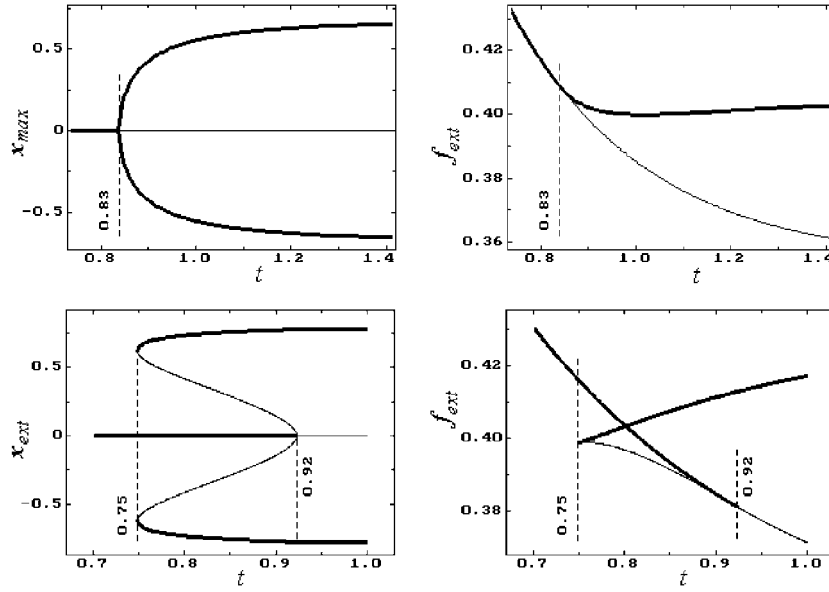


Figure 13. Bifurcation diagrams. Upper panel: for the case $c = 4.0$, $\mu = 1.2$, on the left the thick lines show the location of the maximum, which at the bifurcation time $t_{12} = 0.84 \pm 0.01$ turns into two maxima; right part: the value of the PDF at the maxima location (thick line) and the value at the minimum at $x = 0$ (thin line). Lower panel: for the case $c = 5.5$ and $\mu = 1.2$, on the left positions x_{\max} of the maxima (global and local, thick lines); the thin lines indicate the positions of the minima (at the first bifurcation time, there is a horizontal tangent at the site of the two emerging off-centre maxima). The bifurcation times are $t_{13} = 0.75 \pm 0.01$ and $t_{32} = 0.92 \pm 0.01$. Right: values of the PDF at the maxima (thick lines); the thin line indicates the value of the PDF at the minima.

4.1.3. Lévy flights and thermal (Boltzmann) equilibrium. The above definition (36) of the LFFPE describes a process far from thermal (Boltzmann) equilibrium. In particular, it does not fulfil the linear response theorem in the form (18) known from Gaussian and subdiffusive processes, due to the divergence of the second moment. However, it still underlies the physical concept of additivity of drift and diffusive terms manifested in the fact that for a constant force field $F(x) = m\eta V$, the solution of the LFFPE (36) is given by the propagator at zero force, $P_{F=0}(x - Vt, t)$, taken at the wave variable $x - Vt$ (Jespersen *et al* 1999, Metzler *et al* 1998, Metzler and Klafter 2000a). Above, it was shown that this additive combination of drift and diffusivity produces solutions with converging variance and multimodal properties in superharmonic potentials.

It is interesting to see that one can consistently obtain an equation that describes LFs in the absence of a force field, but which relaxes towards classical thermal equilibrium for *any* non-trivial external field. This equation is based on a different weighting, introduced through a subordination, leading to the ‘exponent-fractional’ equation (Sokolov *et al* 2001)²¹

$$\frac{\partial}{\partial t} P(x, t) = -K^\mu \left(-\frac{\partial^2}{\partial x^2} + \frac{\partial}{\partial x} \frac{F(x)}{k_B T} \right)^\mu P(x, t) \quad (50)$$

²¹ At least when $F(x)$ is a constant, the stochastic process corresponding to equation (49) is $X(T(t))$ where $X(t)$ is the process behind equation (49) when $\mu = 1$ and $T(t)$ is the μ -stable subordinator, compare Bochner 1949; see also the application in Baeumer *et al* 2001.

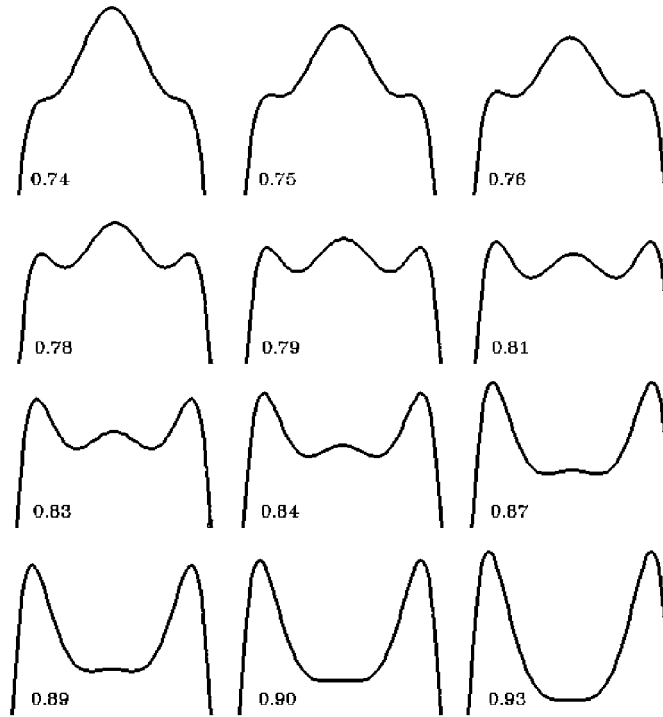


Figure 14. Same as in figure 12, in a superharmonic potential (49) with exponent $c = 5.5$, exhibiting a trimodal structure.

where the μ th power of the Fokker–Planck operator is interpreted in Fourier–Laplace space, i.e., equation (50) acquires the form $\partial P/\partial t = -K^\mu (ikf/[k_B T] + k^2)^\mu P(k, t)$ (Sokolov *et al* 2001). As shown in Sokolov *et al* (2001), the PDF $P(x, t)$ relaxes exponentially towards the regular Boltzmann equilibrium PDF, and is therefore qualitatively different from the LFFPE (36) discussed above. The eigenvalues of λ_n^{ef} of equation (50) are related to those of the regular Fokker–Planck equation (λ_n) by $\lambda_n^{\text{ef}} = -(-\lambda_n)^\mu$, the eigenfunctions coinciding, and the relaxation of moments is exponential, $\propto \exp(-\text{const}|\lambda_n^{\text{ef}}|^\mu t)$ (Sokolov *et al* 2001). In particular, the process leading to the modified LF equation (50) does not possess a direct interpretation of continuous time random walks (but it can be related to the Chapman–Kolmogorov equation). An interesting question will be to determine the corresponding Langevin picture of such a process.

4.2. Bi-fractional transport equations

The coexistence of long-tailed forms for both jump length and waiting time PDFs was investigated within the CTRW approach in Zumofen and Klafter (1995b), discussing in detail the laminar-localized phases in chaotic dynamics. In a similar way, the combination of the long-tailed waiting time PDF (2) with its jump length analogue (33) leads to a dynamical equation with fractional derivatives with respect to both time and space (Luchko *et al* 1998, Mainardi *et al* 2001, Metzler and Nonnenmacher 2002, West and Nonnenmacher 2001):

$$\frac{\partial}{\partial t} P(x, t) = K_\alpha^\mu {}_0 D_t^{1-\alpha} \frac{\partial^\mu}{\partial |x|^\mu} P(x, t). \quad (51)$$

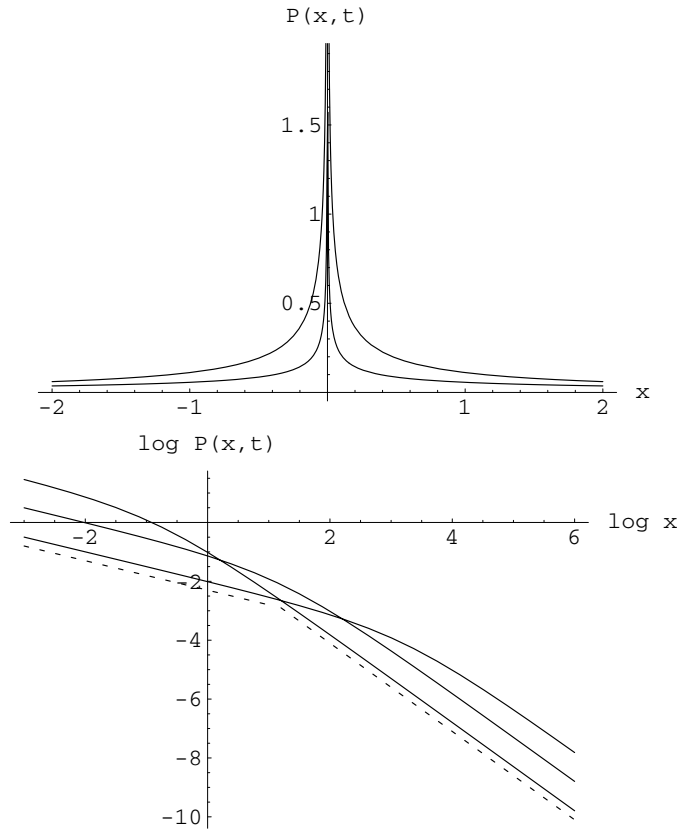


Figure 15. Subdiffusion for the neutral-fractional case $\alpha = \mu = 1/2$. Top: linear axes, dimensionless times $t = 2, 20$. Bottom: double-logarithmic scale, dimensionless times, $t = 0.1, 10, 1000$. The dashed lines in the bottom plot indicate the slopes $-1/2$ and $-3/2$. Note the divergence at the origin.

This equation can in fact be extended to cover the superdiffusive, sub-ballistic domain up to the wave equation (for $\mu = 2$) (Metzler and Klafter 2000d), and under the condition $1 \leq \alpha \leq \mu \leq 2$ in general (Mainardi *et al* 2001). A closed form solution can be found in terms of Fox H -functions (Metzler and Nonnenmacher 2002).

A special case of equation (51) is the ‘neutral-fractional’ case $\alpha = \mu$ (Mainardi *et al* 2001). In this limit, one can obtain simple reductions of the H -function solution, in the following three cases (Metzler and Nonnenmacher 2002):

- (i) Cauchy propagator $\alpha = \mu = 1$,

$$P(x, t) = \frac{1}{2\pi K_1^1 t} \frac{1}{1 + x^2 / (K_1^1 t)} \tag{52}$$

with the long-tailed asymptotics $P(x, t) \sim (2\pi)^{-1} x^{-2}$. The Cauchy propagator converges to $(2\pi K_1^1 t)^{-1}$ at $x = 0$.

- (ii) The case $\alpha = \mu = 1/2$ (figure 15),

$$P(x, t) = \frac{2}{|x| \sqrt{2\pi + 2\pi z^{1/2} + \sqrt{2\pi} z}} \quad \therefore \quad z = \frac{|x|}{2(K_{1/2}^{1/2})^2 t} \tag{53}$$

At the origin, this PDF diverges $\sim |x|^{-1/2}$, while for large $|x|$, it decays like $\sim |x|^{-3/2}$.

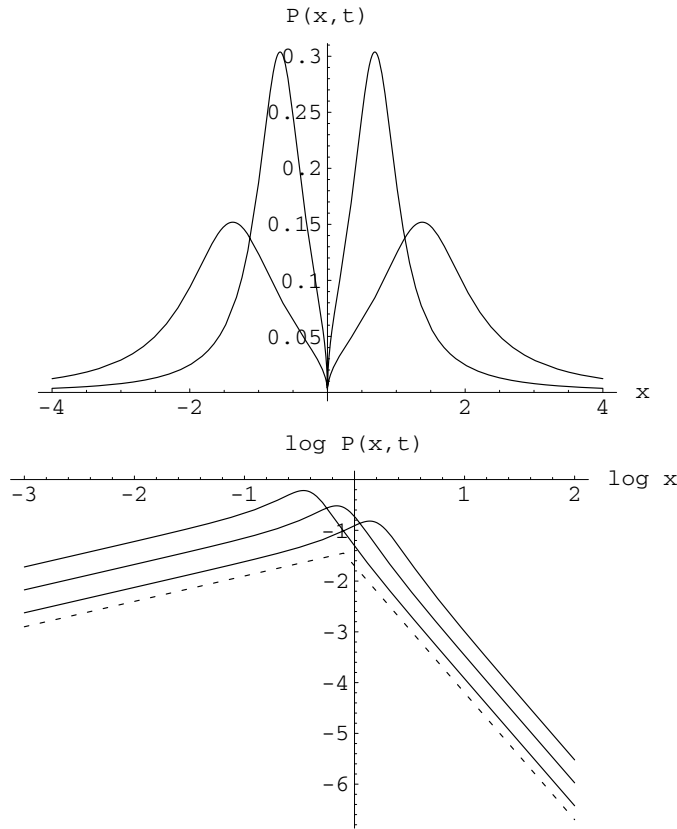


Figure 16. Superdiffusion for the neutral-fractional case $\alpha = \mu = 3/2$. Top: linear axes, dimensionless times $t = 1$ and 2 . Bottom: double-logarithmic scale drawn for the dimensionless times $0.5, 1$ and 2 . The dashed lines in the bottom plot indicate the slopes $1/2$ and $-5/2$. Note the complete depletion at the origin.

(iii) The case $\alpha = \mu = 3/2$ (figure 16):

$$P(x, t) = \frac{\sqrt{2}}{3\pi|x|} \frac{z^{3/2} + z^3 + z^{9/2}}{1 + z^6} \quad \therefore \quad z = \frac{2^{1/3}|x|}{(K_{3/2}^{3/2})^{2/3}t}. \quad (54)$$

In this case, the PDF shows complete depletion at the origin, exhibiting a $\sim |x|^{1/2}$ square root behaviour close to $|x| = 0$, and it has the inverse power-law decay $\sim |x|^{-5/2}$ for $|x| \rightarrow \infty$. This solution also exhibits a bimodal PDF, known from equation (9) in the superdiffusive sub-ballistic regime ($1 < \alpha < 2$) (Metzler and Klafter 2000d, West *et al* 1997). This superdiffusive feature comes from the interpretation of the process as a memory version of the wave equation, i.e., to a propagative contribution (Metzler and Klafter 2000d). It is therefore completely different from the bimodal structure in confined LFs discussed above.

We note that bi-fractional diffusion equations were also discussed in Barkai (2002), Saichev and Zaslavsky (1997), Baeumer *et al* (2003), Hughes (2002), Gorenflo *et al* (2002b) and Uchaikin (2002); see also Meerschaert *et al* 2002a with a discussion of the connection between fractional time derivatives and subordination to a process with Mittag-Leffler PDF.

A bi-fractional Fokker–Planck equation with a power-law dependence $\propto |x|^{-\theta}$ ($\theta \in \mathbb{R}$) of the diffusion coefficient was studied in Fa and Lenzi (2003) and Lenzi *et al* (2003a).

4.3. Lévy walks

Lévy walks (LWs) correspond to the spatiotemporally coupled version of continuous time random walks. The waiting time and jump length PDFs are no longer decoupled but appear as conditional in the form $\psi(x, t) \equiv \lambda(x)p(t|x)$ (or $\psi(t)\tilde{p}(x|t)$) (Klafter *et al* 1987). In particular, through the coupling $p(t|x) = \frac{1}{2}\delta(|x| - vt^\nu)$, one introduces a generalized velocity v , which penalizes long jumps such that the overall process, the LW, attains a finite variance and a PDF with two spiky fronts successively exploring space (Zumofen and Klafter 1993, Klafter and Zumofen 1994a). Thus, LWs have properties similar to generalized Cattaneo/telegraphers' equation-type models (Compte and Metzler 1997, Metzler and Compte 1999, Metzler and Nonnenmacher 1998). In particular, for $\nu = 1$, the coupling $p(t|x) = \frac{1}{2}\delta(|x| - vt)$ introduces a proper velocity v , and the PDF $P(x, t)$ can be expressed explicitly in the velocity model (Zumofen and Klafter 1993), see also Metzler and Compte 1999. For the enhanced, sub-ballistic regime $\langle x^2(t) \rangle \sim t^{3-\gamma}$ with $1 < \gamma < 2$, the asymptotic behaviour of this PDF is $P(x, t) \simeq t^{-1/\gamma} f(\xi)$ with the scaling function (Zumofen *et al* 1999):

$$f(\xi) \sim \begin{cases} t^{-1/\gamma} L_\gamma(-c\xi), & |x| < vt \\ \delta(|x| - vt)t^{-\gamma}, & |x| \simeq vt \\ 0, & |x| > vt, \end{cases}$$

where $\xi = |x|/t^{1/\gamma}$ and $L_\gamma(-c\xi)''$ is the symmetric Lévy stable density of index γ . The cutoff at $|x| = vt$ is a consequence of the constant velocity v introduced in $p(t|x)$.

On the basis of the latter fractional equations, formulations were obtained for the description of LWs in the presence of non-trivial external force fields, with the same restriction to lower order moments with respect to an LW process (Barkai and Silbey 2000, Metzler and Sokolov 2002). Recently, however, a coupled fractional equation was reported (Sokolov and Metzler 2003), which describes a force-free LW exactly. Thus, it was shown that the fractional version of the material derivative $\partial/\partial t \pm \partial/\partial x$,

$$d_\pm^\beta P(x, t) \equiv {}_0D_t^\beta P(x \pm t, t) \quad (55)$$

defined in Fourier–Laplace space through

$$\mathcal{F}\{\mathcal{L}\{d_\pm^\beta f(x, t); u\}; k\} \equiv (u \pm ik)^\beta f(k, u) \quad (56)$$

(\mathcal{F} acts on x and \mathcal{L} on t) replaces the uncoupled fractional time operators introduced in the previous work; compare also the detailed discussion of LW processes in Zumofen and Klafter (1993). Although one may argue for certain forms (Sokolov and Metzler 2003), there is so far no derivation for the incorporation of general external force fields in the coupled formalism. A question of particular interest is whether LWs in non-trivial external fields relax towards a stationary solution or not; compare Metzler and Sokolov (2002). We note that a very similar fractional approach to LWs was suggested by Meerschaert *et al* (2002) virtually simultaneously.

5. Applying fractional dynamics

The interesting quantity of many dynamical processes is the PDF and its associated mean time, to arrive at or cross a certain point after having started somewhere else in the system.

This is the problem of first passage, which for Brownian motion is identical to the problem of first arrival. In the presence of subdiffusion, the first passage time problem corresponds to a subordination of the analogous Brownian problem, and can be solved with the same tools. The major difference is the divergence of the mean first passage time (MFPT) T . In contrast, for LFs, we will report that the method of images breaks down, and the details of the process (especially the Lévy index) enter only marginally into the first passage time density (FPTD). Apart from the first passage, the nature of the underlying diffusion process naturally defines the detailed properties of the associated diffusion–reaction problem. We will discuss these two fundamental applications of (anomalous) diffusion processes in the following.

5.1. First passage time processes and boundary conditions

The firing of neurons (Ben-Yacov 2002, Gerstner and Kistler 2002), diffusion-limited aggregation (Vicsek 1991), hydrological breakthrough (Kirchner *et al* 2000, Scher *et al* 2002a), the passage of a biomolecule through a membrane nanopore (Bates *et al* 2003, Metzler and Klafter 2003), the fluctuation behaviour of single-strand bubbles in DNA double helices (Hanke and Metzler 2003), the encounter of two independently diffusing particles (von Smoluchowski 1916a, 1916b), the rebinding of a ligand to a protein (Iben *et al* 1989), the switching of topological molecules (Metzler 2001b), or the electrical current caused by anomalously moving charge carriers in amorphous semiconductors (Pfister and Scher 1977, 1978) etc, can be mapped onto the problem of calculating the FPTD, and the associated MFPT (Redner 2001), an approach originally dating back to Schrödinger (1915).

In the regular, Brownian domain of diffusion, the unbiased passage for a process with initial condition $P_0(x) = P(x, 0) = \delta(x - x_0)$ is described by the FPTD

$$p(t) = \frac{x_0}{\sqrt{4\pi K t^3}} \exp\left(-\frac{x_0^2}{4Kt}\right) \quad (57)$$

which defines the probability $p(t) dt$ for the particle to arrive at $x = 0$ during the time interval $t, \dots, t + dt$. Its long-time behaviour corresponds to the 3/2 power-law behaviour

$$p(t) \sim \frac{x_0}{\sqrt{K}} t^{-3/2}. \quad (58)$$

In particular, we note that even for Brownian processes there are natural cases when the characteristic time diverges. Here, the MFPT $T = \int_0^\infty p(t)t dt = \infty$.

In a finite box of size L , the Brownian first passage time problem has an exponential tail, with increasingly quicker decay on increasing mode number,

$$p_L(t) = \frac{\pi K}{L^2} \sum_{n=0}^{\infty} (-1)^n (2n+1) \exp\left(-K \frac{(2n+1)^2 \pi^2}{4L^2} t\right). \quad (59)$$

This solution for the FPTD corresponds to an initial condition in the centre of a box with two absorbing boundaries. The MFPT becomes $T = L^2/(2K)$.

The third case of interest here is the first passage in a semi-infinite domain in the presence of a constant bias V . In the corresponding FPTD

$$p_V(t) = \frac{x_0}{\sqrt{4\pi K t^3}} \exp\left(-\frac{(x_0 - Vt)^2}{4Kt}\right) \quad (60)$$

the drift term $V^2 t^2$ in the exponential outweighs the diffusive $1/(Kt)$ part, and causes an exponential decay. One finds the MFPT $T = x_0/V$, i.e., the average first passage time matches exactly the classical motion with constant velocity V .

Let us now investigate what changes come about in the presence of transport anomalies of subdiffusive and LF nature.

5.1.1. Subdiffusion. The first passage of a diffusing particle through a point x_f in processes characterized by a jump length PDF with finite variance \mathfrak{X}^2 can always be mapped on the problem of putting an absorbing boundary at that point x_f and calculating the negative time derivative of the survival probability. A natural way of constructing the FPTD in such a case is the method of images (Redner 2001) going back to Lord Kelvin. For instance, on the semi-infinite domain, this method prescribes to mirror the unrestricted propagator with initial condition x_0 at the point x_f , and turning this image negative. The image solution is then the sum of both PDFs. Thus, the part of the PDF which has imaginarily crossed the absorbing boundary at x_f at some time t , is subtracted from the original PDF. In our example of the semi-infinite domain with $x_f = 0$, the image solution becomes $Q(x, t) = P(x, t; x_0) - P(x, t; -x_0)$. Thus, one readily obtains $Q(0, t) = 0$, as it should. The integral $S(t) = \int_0^\infty Q(x, t) dx$ is the survival probability. Its negative derivative $p(t) = -dS(t)/dt$ is then the FPTD. In a similar manner, the images method can be employed to solve the (sub)diffusion in a box, or in the presence of a constant bias (Metzler and Klafter 2000e, Redner 2001).

Another method is based on the generalized Laplace transformation (22), which is equally valid for the position average of $P(x, t)$, in the present language the survival probability. Thus, we can relate the Markovian (S_M) and subdiffusive (S) results through

$$S(u) = u^{\alpha-1} S_M(u^\alpha) \quad (61)$$

corresponding to the relation through equations (21) and (22). As the FPTD is the (negative) time derivative of the survival probability S , the Brownian and subdiffusive FPTDs are not connected by relation (22). However, it is straightforward to show that the latter fulfil the scaling relation

$$p_\alpha(u) = p_M(u^\alpha) \quad (62)$$

in Laplace space. In the time domain, this corresponds to another generalized Laplace transformation of the kind (22), but with the one-sided Lévy density $\mathcal{L}^{-1}\{\exp(-su^\alpha); t\} \sim t^{-1-\alpha}$ as kernel.

The essential property of subdiffusive first passage time problems lies in the fact that the long-tailed nature of the waiting time PDF translates into the FPTD itself. The MFPT diverges both in the absence of a bias and under a constant drift, pertaining to both finite as well as semi-infinite domains (Metzler and Klafter 2000e, Rangarajan and Ding 2000a, 2000b, 2003, Scher *et al* 2002a, Barkai 2001)²². For the three cases of first passage time problems, we obtain the following subdiffusive generalizations:

- (i) For subdiffusion in the semi-infinite domain with an absorbing wall at the origin and initial condition $P(x, 0) = \delta(x - x_0)$ it was found that (Metzler and Klafter 2000e)

$$p(t) \sim \frac{x_0}{|\Gamma(-\alpha/2)| K_\alpha^{1/2}} t^{-1-\alpha/2} \quad (63)$$

i.e., the decay becomes a flatter power law than in the Markovian case (58).

- (ii) Subdiffusion in the semi-infinite domain in the presence of an external bias V falls off faster, but still in power-law manner (Barkai 2001, Metzler and Klafter 2003, Scher *et al* 2002a):

$$p(t) \sim t^{-1-\alpha}. \quad (64)$$

²² Note that for both subdiffusive (\mathfrak{T} diverges) and LF (\mathfrak{X}^2 diverges) systems it may be dangerous to invoke results from the x, t -scaling found in the mean squared displacement $\langle \Delta x^2(t) \rangle \sim t^\alpha$, from which erroneously a finite MFPT could be predicted, or inferred at all, respectively. Compare Gitterman (2000), Yuste and Lindenberg (2004) and Li and Wang (2003), Metzler and Sokolov (2004), as well as Denisov *et al* (2003), Reigada *et al* (2002).

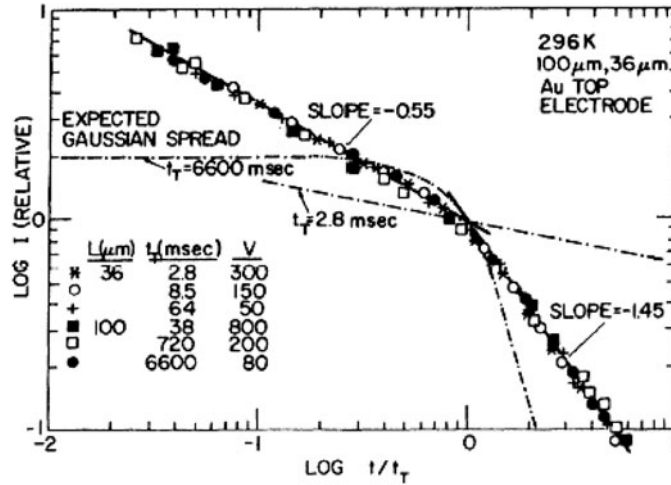


Figure 17. Universality of the electrical current shape as a function of time, for various driving voltages as sample sizes. The slopes of the two power laws add up to 2. Note the impressive data collapse after rescaling (Pfister and Scher 1977).

In strong contrast to the biased Brownian case, we now end up with a process whose characteristic time scale diverges. This is exactly the mirror of the multiple trapping model, i.e., the classical motion events become repeatedly interrupted such that the immobilization time dominates the process.

(iii) Subdiffusion in a finite box (Metzler and Klafter 2000e)²³:

$$p(t) \sim t^{-1-\alpha} \tag{65}$$

i.e., this process leads to the same scaling behaviour for longer times as found for the biased semi-infinite case (ii). Effectively, the drift towards the absorbing boundary outweighs the diffusion, as remarked in the Brownian case above, and acts as a reflecting boundary.

The latter two results should be compared to the classical Scher–Montroll finding for the FPTD of biased motion in a finite system of size L with absorbing boundary condition. In that case, the FPTD exhibits two power laws (compare figure 17)

$$p(t) \sim \begin{cases} t^{\alpha-1} & t < \tau \\ t^{-1-\alpha} & t > \tau \end{cases} \tag{66}$$

the sum of whose exponents equals -2 (Pfister and Scher 1977, 1978, Scher *et al* 2002a, 2002b). Here, τ is a system parameter dependent time scale (Pfister and Scher 1978). In figure 17 we also find the anomalous diffusion modelling interestingly proved by the experimental data (Pfister and Scher 1977, 1978, Scher and Montroll 1975). As shown in figure 17, a Brownian drift–diffusion model could not reasonably reproduce the data (which in that case would also be non-universal in the sense that it would not exhibit the observed data collapsing). Note that the anomalous transport behaviour also correctly describes other system features, such as the size-dependent mobility. These investigations were the first, and highly successful, application of continuous time random walk dynamics.

²³ It does not matter whether both boundaries are absorbing, or one is reflective.

5.1.2. *Lévy flights.* The first passage time problem for LFs might naively be considered simpler than the corresponding subdiffusive problem, keeping in mind that LFs are Markovian. Moreover, their dynamical equation is linear. It might therefore be tempting to apply the method of images to infer the FPTD for an LF. Such an approach was in fact pursued in Montroll and West (1976) for a finite domain, and with similar methods in Gitterman (2000). In Buldyrev *et al* (2001), eigenfunctions of the full-space LFFPE were obtained, from which the FPTD was determined. These methods lead to results for the FPTD in the semi-infinite domain, whose long-time behaviour is explicitly dependent on the Lévy index, such that the FPTD would actually decay faster than under Brownian conditions. Given the much quicker exploration of space of LFs, this is an *a priori* intuitive result. In fact, it was shown by Sparre Andersen that *any* symmetric jump length PDF gives rise to a decay $\sim n^{-3/2}$ of the FPTD with number of steps n (Sparre Andersen 1953, 1954). For any Markov process, the analogous continuous time behaviour can be obtained through $n \propto t$, and is according to the Sparre Andersen theorem given by equation (58). Thus, an LF necessarily has to fulfil the Sparre Andersen universality, fully independent of the Lévy index²⁴. We note that an analogous result was proved in Frisch and Frisch (1995) for the special case in which an absorbing boundary is placed at the location of the starting point of the LF at $t > 0$, and numerically corroborated in Zumofen and Klafter (1995a).

The inadequacy of the images method can indeed be understood from the Lévy fractional diffusion equation (34). There, reflecting the strongly non-local nature of LFs the fractional Riesz operator stretches its integration from $-\infty$ to $+\infty$. However, given an absorbing boundary condition at $x = 0$ with an initial condition at $x_0 > 0$, the solution necessarily has to vanish on the negative semi-axis. The correct dynamical equation in the presence of the boundary condition should therefore read (Chechkin *et al* 2003a)

$$\frac{\partial f(x, t)}{\partial t} = \frac{D}{\kappa} \frac{\partial^2}{\partial x^2} \int_0^\infty \frac{f(x', t)}{|x - x'|^{\mu-1}} dx' \equiv \frac{\partial^2}{\partial x^2} \mathcal{F}(x, t) \quad (67)$$

in which the fractional integral is truncated to the semi-infinite interval and $\kappa = 2\Gamma(2 - \mu)|\cos \pi\mu/2|$. After Laplace transformation and integrating over x twice, one obtains²⁵

$$\int_0^\infty K(x - x', s) f(x', s) dx' = (x - x_0)\Theta(x - x_0) - xp(s) - \mathcal{F}(0, s) \quad (68)$$

where $p(t)$ is the FPTD and the kernel $K(x, s) = sx\Theta(x) - (\kappa|x|^{\mu-1})$. This equation is formally of the Wiener–Hopf type of the first kind (Gakhov 1966). After some manipulations similar to those applied in Zumofen and Klafter (1995a), we arrive at the asymptotic expression $p(s) \simeq 1 - Cs^{1/2}$, where $C = \text{const}$, in accordance with the universal behaviour (58) and with the findings in Zumofen and Klafter (1995a). Thus, the dynamic equation (67) consistently phrases the FPTD problem for LFs. A numerical analysis of an LF in the presence of an absorbing boundary is shown in figure 18, nicely corroborating the Sparre Andersen universality for various Lévy exponents (Chechkin *et al* 2003a). In these simulations, a particle was removed when its jump would move it across the absorbing boundary. The simulations also demonstrate that the measured FPTD cannot be described by the other two (in this case inadequate) approaches, the images method and the direct definition addressed now.

²⁴ It is only for processes in which time t and number of steps n are not a linear function of each other that the Sparre Andersen universality is broken (Feller 1968, Redner 2001, Sparre Andersen 1953, 1954, Spitzer 2001), for instance, for subdiffusion or LWs.

²⁵ This definition is thus significantly different from the Brownian case in which it does not matter whether the lower integration limit is 0, or actually $-\infty$.

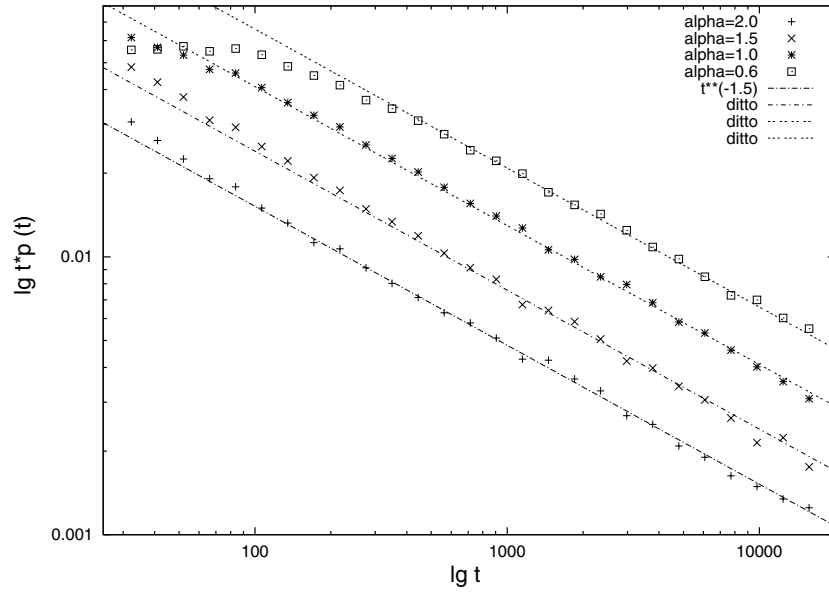


Figure 18. First passage time density for LFs with different index. The universal (Sparre Andersen) decay with power-law index $-3/2$ is nicely fulfilled.

This direct definition of the FPTD for Brownian processes uses the chain rule ($p_{fa}(\tau)$ depends implicitly on x_0) (Hughes 1995, Redner 2001)

$$P(-x_0, t) = \int_0^t p_{fa}(\tau) P(0, t - \tau). \tag{69}$$

More precisely, this convolution, which corresponds to the algebraic relation $p_{fa}(u) = P(-x_0, u)/P(0, u)$ in Laplace space, is the PDF of first arrival. However, in Gaussian processes (Brownian diffusion, subdiffusion), due to the local jumps, both notions are equivalent. This is no longer true for an LF. Here, by long jumps the LF particle can repeatedly hop across the point, until it eventually lands there. For an LF, the chain rule will therefore produce a too slowly decaying FPTD (Chechkin *et al* 2003a). Equation (69) is equivalent to a process with a δ -sink paraphrased by the dynamical equation

$$\frac{\partial}{\partial t} f(x, t) = D \frac{\partial^\mu}{\partial |x|^\mu} f(x, t) - p_{fa}(t) \delta(x). \tag{70}$$

For an LF with index μ , one would therefore expect the PDF of first arrival to decay like

$$p_{fa}(t) \sim t^{-2+1/\mu}. \tag{71}$$

This was investigated by numerical simulation, as shown in figure 19. A small interval w around the sink was fixed, and any particle jumping into this zone was removed.

We note that LWs on a finite domain and in the presence of absorbing boundary conditions were studied extensively in Drysdale and Robinson (1998). A one-sided Lévy stable jump length PDF was investigated in Eliazar and Klafter (2004); due to its asymmetric character, such a process is not subject to the Sparre Andersen theorem. Indeed, the detailed analysis of this ‘shot-noise’ type process with one-sided, long-tailed $\lambda(x)$ for the special case $\mu = 1/2$ reveals the FPTD $a(\pi x_0)^{-1/2} \exp(-a^2 t^2/(4x_0))$ where a is the amplitude of the Lévy stable law, and x_0 is the distance of the absorbing barrier from the initial location

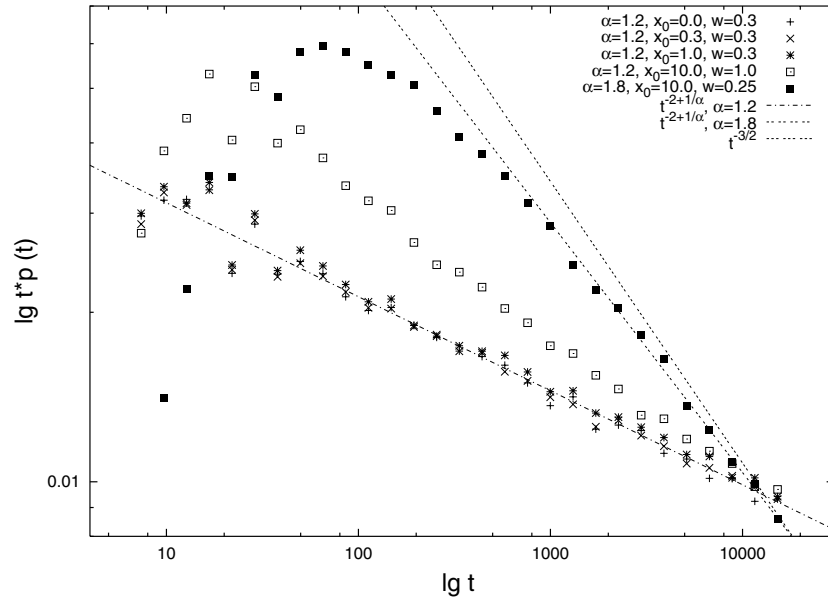


Figure 19. First arrival PDF for $\mu = 1.2$ demonstrating the $t^{-2+1/\mu}$ scaling, for trap width $w = 0.3$. For comparison, we show the same scaling for $\mu = 1.8$, and the power law $t^{-3/2}$ corresponding to the FPTD. The behaviour for too large $w = 1.0$ shows a shift of the decay towards the $-3/2$ slope. Note that on the abscissa we plot $\lg tp(t)$. Note also that for the initial condition $x_0 = 0.0$, the trap becomes activated *after* the first step, consistent with Zumofen and Klafter (1995a).

(Eliazar and Klafter 2004); compare also to the detailed discussion in Meerschaert and Scheffler 2004.

5.2. Reaction–diffusion processes

The above result that for LFs the first arrival and the first passage are in fact different from each other is expected to have profound implications for diffusion–reaction under Lévy jump length conditions. Similarly, subdiffusion conditions will change the dynamics of reactive systems. We note that the theoretical modelling of reaction–diffusion processes within a stochastic framework goes back to von Smoluchowski (1916a, 1916b) in which the encounter of two independently diffusing particles is considered. The simplest version, diffusing particles in the presence of an immobile reaction centre C, at which particles undergo an annihilation reaction $A+C \rightarrow 0$, is described by the first arrival time results obtained above: the decay of the concentration of A follows $\sim t^{-\alpha/2}$ in subdiffusion with $0 < \alpha < 1$, and $\sim t^{-1+1/\alpha}$ for an LF of Lévy index $1 < \alpha < 2$. Note that in the latter case, the reaction becomes successively stalled for α getting close to the Cauchy case $\alpha = 1$, and that it approaches the Brownian case $\sim t^{-1/2}$ for $\alpha \rightarrow 2$.

In Yuste and Lindenberg (2001, 2002), the problem of subdiffusive reaction–diffusion of the coagulation type $A + A \rightarrow A$ was considered. Thus, the quantity of interest, the density function $E(x, t)$ to find an interval of length x empty of any particle at time t , was shown to be described by the fractional diffusion equation

$$\frac{\partial}{\partial t} E(x, t) = 2 {}_0D_t^{1-\alpha} K_\alpha \frac{\partial^2}{\partial x^2} E(x, t) \quad (72)$$

where we note the occurrence of the additional factor 2. For an initially random interparticle distribution $\lambda e^{-\lambda x}$ of the Poisson type, one can, for instance, obtain the concentration of particles,

$$c(t) = \lambda E_{\alpha/2}(-\lambda\sqrt{2K_\alpha}t^{\alpha/2}) \quad (73)$$

which exhibits a temporal decay of the Mittag-Leffler kind, with scaling exponent $\alpha/2$. Similarly, an approximate expression for the time behaviour of the interparticle distance can be obtained. The analysis in Yuste and Lindenberg (2001) highlights the advantages of having at hand a formulation of the process in terms of a dynamical equation, i.e., the fractional diffusion equation. Compare the derivation in Henry and Wearne (2000).

A fractional diffusion equation for the geminate reaction of a particle B, which starts a distance r_0 away from particle A, is given as follows. B moves subdiffusively towards A, until it reaches the encounter distance within the range $R, \dots, R + dr$. This process was shown to be controlled by the fractional diffusion–reaction equation (Seki *et al* 2003a, 2003b) (compare Sung *et al* (2002))

$$\frac{\partial}{\partial t} C(r, t) = {}_0D_t^{1-\alpha} \left(K_\alpha \nabla^2 - k_\alpha \frac{\delta(r-R)}{4\pi R^2} \right) C(r, t) \quad (74)$$

for the concentration $C(r, t)$, where we retained the spherical notation from Seki *et al* (2003a, 2003b). Here, the generalized rate constant is k_α .

A systematic derivation of reaction–diffusion equations with distributed delays (memory kernels), including their connection to fractional reaction–diffusion equations, is discussed, and applied to the modelling of the (neolithic) transition from the hunting and gathering to the agricultural society in Vlad and Ross (2002). In a two-species fractional reaction–diffusion system Turing instabilities were identified by Henry and Wearne (2002).

In del-Castillo-Negrete *et al* (2003) the front dynamics in a reaction–diffusion system based on an asymmetric jump distribution, of Gaussian nature to the left, say, and Lévy stable to the right, related to the study of a birth–death process with similar jump rules (Sokolov and Belik 2003), is investigated. In a similar study, superfast reaction front propagation was observed in the presence of long-tailed increments (Mancinelli *et al* 2002), which is based on a fractional version of the Fisher–Kolmogorov equation. The efficiency of Lévy statistics in mixing of chemical reactions was shown in a numerical study of an $A+B \rightarrow 0$ reaction (Zumofen *et al* 1996a, 1996b). Accordingly, Lévy walks under certain parametric conditions can avoid the Ovchinnikov–Zeldovich segregation of the reactants, which under inefficient mixing conditions impedes speedy reactions, refer to figure 20. Finally, subdiffusive reaction dynamics was investigated in detail in Blumen *et al* (1986b).

6. Summary

Fractional dynamics ideas have been extensively explored during the past few years, both theoretically and experimentally. It was therefore timely to update our review (Metzler and Klafter 2000a), in order to provide a basic reference for colleagues who are either actively working in the field of fractional dynamics, or who would like to get a quick overview. We decided to write this new review in a self-contained manner. In particular, we gathered a number of applications, and went more deeply into Lévy flight-type processes. Finally, we discussed in detail first passage and reaction–diffusion problems under anomalous conditions. In contrast, we did not repeat the historical and mathematical context from the earlier work (Metzler and Klafter 2000a), and we refer to that work for details, in particular, how to obtain explicit solutions in terms of Fox H -functions and via the method of separation of variables, and for earlier references to papers on fractional dynamics.

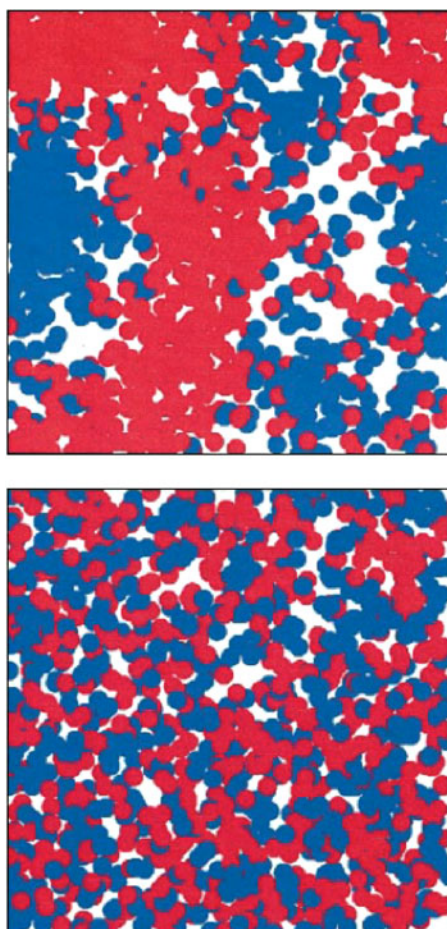


Figure 20. Top: Ovcchinnikov–Zeldovich segregation for nearest-neighbour random walks under $A + B \rightarrow 0$ reaction conditions where the two colours symbolize the two reactant species. Bottom: the same process under Lévy mixing conditions, the Ovcchinnikov–Zeldovich segregation is removed (Zumofen *et al* 1996a).

We concentrated on fractional integro-differential operators of the Riemann–Liouville type (acting on time t) and the Riesz–Weyl (position x) type. These follow straightforwardly from physical principles such as the continuous time random walk (Compte 1996, Metzler *et al* 1999b), the generalized master equation (Metzler 2001a), the continuous time Chapman–Kolmogorov equation (Metzler 2000), a multiple trapping model (Metzler and Klafter 2000b, 2000c), the Langevin equation with a Lévy noise source (Seshadri and West 1982, Fogedby 1994a, Peseckis 1987, Schertzer *et al* 2001), or subordination (Sokolov). In particular, the equations obtained through these methods (probabilistic approach!) define a proper PDF as their solution, compare the discussion in Brockmann and Sokolov (2002), Sokolov (2002). There exist numerous other definitions (Samko *et al* 1993, Srivastava and Saxena 2001, Hilfer 2000), whose potential relation to concrete physical systems has not yet been fully explored. We note that there exist other approaches to fractional equations, and connections to other physical processes. Thus, it was shown that the FFPE emerges as the master equation for a Langevin process the internal clock of which is the FPTD of a self-similar Markov process (Stanislavsky 2003)

(compare to the fractional Langevin equation (Lutz 2001b, Picozzi and West 2002). Similarly, a non-Markovian stochastic Liouville equation was recently discussed (Shushin 2003), and a statistical functional method to derive LFs was explored in Vlad *et al* (2000). New aspects of the relation between deterministic systems and anomalous transport (Zaslavsky 1999, 2002, Klafter and Zumofen 1994b) have recently been discussed (Artuso and Cristadoro 2003), also from a quantum perspective (Iomin and Zaslavsky 2002, Laskin 2002, Banerjee *et al* 2002), and effects of memory in the sense of macroscopic time are discussed in Zaburdaev (2003). LFs in quenched jump length fields are investigated in a renormalization group approach in Schulz (2002) and Schulz and Reineker (2002). We also note that the potential conflict between PDF and trajectory description of anomalous transport was considered in Bologna *et al* (2002). Finally, it should be mentioned that finite sampling effects for LFs were investigated in Condat *et al* (2002).

Apart from the solutions discussed herein, exact and approximate solutions of the FFPE were established in a double-well potential for rotational dipoles (Kalmykov *et al* 2003), for the fractional oscillator (Ryabov and Puzenko 2002), and for double-well anomalous diffusion in Shushin (2001). Similarly, fixed axes dipoles rotating in an N -fold cosine potential were investigated on the basis of the fractional Klein–Kramers equation in Coffey *et al* (2003). The fractional Fokker–Planck equation on a comb structure was explored by Zahran *et al* (2003), El-Wakil *et al* (2002). Experimentally, such predictions are being explored in Jadżyn (2003). Cole–Cole-type relaxation patterns were studied in dielectric systems (Ryabov *et al* 2003, Feldman *et al* 2002), and applied to seismic wave attenuation (Hanyga 2003). ‘Reverse engineering’, i.e., the exploration of how to design a Lévy noise-driven Langevin system, which yields a given steady state behaviour, was studied in Eliazar and Klafter (2003). Stationarity-conservation laws could be determined for fractional differential equations with variable coefficients (Klimek 2002).

From the phase space or position space fractional dynamical equations, one can by integration obtain the purely time-dependent relaxation behaviour of the system under consideration. Given the anomalous diffusion behaviour described by such dynamics, also the relaxation patterns will deviate from the classical exponential Maxwell–Debye form (Plonka 2001, Ramakrishnan and Raj Lakshmi 1987). Fractional relaxation models have *inter alia* been discussed in Blumen *et al* (2002), Richert (2002), Schiessel *et al* (1995) and Uchaikin (2003). Applications of such anomalous relaxation models have been impressively demonstrated over some 10 to 15 decades in frequency for dielectric relaxation (Hilfer 2003) and polymeric systems (Glöckle and Nonnenmacher 1991, Metzler *et al* 1995). A comparative study of different anomalous relaxation models was undertaken in Talkner (2001). Compare also to dynamical disorder models generalizing the KWW-stretched exponential form (Vlad *et al* 1998, 1996).

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Appendix A. Continuous time random walk approach to fractional dynamics

A standard textbook random walk process is assumed to perform a step of fixed length in a random direction at each tick of a system clock, i.e., having constant spatial and temporal increments, Δx and Δt . Such a process will give rise to the standard diffusion process in

the long-time limit, i.e., after a sufficient number of steps, and the associated random variable $x(t) = N^{-1/2} \sum_i^N x_i$, where x_i is the position after the i th step, will be distributed by a Gaussian due to the central limit theorem. A convenient generalization of this process is the so-called continuous time random walk (CTRW) process, originally introduced by Montroll and Weiss (1965), in which both jump length and waiting time are distributed according to two PDFs, $\lambda(x)$ and $\psi(t)$. A CTRW therefore is based on a probabilistic concept. It is easy to show that the propagator for such a CTRW process in the absence of an external force is given in terms of the simple expression

$$P(k, u) = \frac{1 - \psi(u)}{u[1 - \psi(k, u)]} \quad (\text{A.1})$$

in Fourier–Laplace space (Klafter *et al* 1987). A detailed review is the book by Hughes (1995). We note that this framework is based on a Euclidean space, and has to be modified for transport on supports with fractal dimension (Ben-Avraham and Havlin 2000, Metzler *et al* 1994, Metzler and Nonnenmacher 1997, Kobelev *et al* 2002, 2003, Ren *et al* 2003a, 2003b, Acedo and Yuste 1998).

The waiting time and jump length PDFs. A waiting time PDF of the long-tailed inverse power-law form $\psi(t) \sim \tau^\alpha/t^{1+\alpha}$ ($0 < \alpha < 1$), which enters the CTRW propagator (A.1) via the Laplace expansion $\psi(u) \sim 1 - (u\tau)^\alpha$ ($u \ll \tau$), can be completed, for instance, to a one-sided Lévy stable density $L_{\alpha,\tau}^+(t)$ whose characteristic function in Laplace space is $\varphi(u) = \int_0^\infty L_{\alpha,\tau}^+(t) e^{-ut} dt = \exp(-[u\tau]^\alpha)$ (Lévy 1954, Gnedenko and Kolmogorov 1954, Hughes 1995, Samorodnitsky and Taqqu 1994, Re *et al* 2003). This complete, explicit representation for $\psi(t)$ has the advantage that it includes the limit $\alpha = 1$, in which case $\psi(u) = e^{-u\tau} \sim 1 - u\tau$, and $\psi(t) = \delta(t - \tau)$. Another possibility is to choose $\psi(t) = (\tau^\alpha/t^{1-\alpha})E_{\alpha,\alpha}(-[t/\tau]^\alpha)$, where $E_{\alpha,\alpha}$ is the generalized Mittag-Leffler function with the series expansion $\psi(t) = (\tau^\alpha/t^{1-\alpha}) \sum_{n=0}^\infty (-[t/\tau]^\alpha)^n / \Gamma(\alpha n + \alpha)$, as derived by Hilfer and Anton (1995). In this case, the limit $\alpha = 1$ becomes $\psi(t) = e^{-t/\tau}$.

Similarly, the jump length PDF $\lambda(x)$ can be assumed to be given by a symmetric Lévy stable PDF defined in terms of its characteristic function, $\varphi(k) \equiv \int_{-\infty}^\infty \lambda(x) \exp(ikx) dx = \exp(-[\sigma|k|]^\mu)$, where $0 < \mu \leq 2$. In the case $\mu = 2$, we obviously recover the Gaussian distribution, which corresponds to a standard jump length distribution. For $0 < \mu < 2$, $\lambda(x) \sim \sigma^\mu/|x|^{1+\mu}$, and in particular, its variance diverges (Hughes 1995, Lévy 1954).

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