Non-Gaussianity in stochastic transport: phenomenology and modelling

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Abstract. Non-Gaussian shapes, despite a linear form of the mean-squared displacement, have been observed for the displacement distribution in a large range of diffusive systems. Stochastic models for such "Brownian yet non-Gaussian" diffusion will be introduced and discussed. Systems with non-Gaussian, anomalous diffusion will also be addressed.

1. Introduction

In their foreword to volume 5 of the famed Theoretical Physics textbook series, Landau and Lifshitz address the perception of Statistical Physics [1]: "Among physicists there exists the widely prevalent fallacy that Statistical Physics was the least well-founded discipline of Theoretical Physics. In doing so it is typically referred to the lack of rigorous mathematical proof of some conclusions in Statistics; one forgets that also other disciplines of Theoretical Physics contain inexact proofs, but on no account this is regarded as a signature of an insufficient validity of these disciplines."

While this perception has not fully vanished (and is partially shared by our undergraduate students) over the 80 years since this was written‡ Statistical Physics has become a success story, being continuously developed further, and it has pervaded an ever increasing range of fields, including the physics of condensed, soft, and biological

‡ To some extent, one might argue that the perception of Statistical Physics is often (too) closely connected with that of classical Thermodynamics, that in its axiomatic nature is indeed quite different from other fields of physics.
matter, geophysics, economy, ecology, or epidemiology. An outstanding acknowledgement of the role of Statistical Physics has been the 2021 Physics Nobel Prize to Giorgio Parisi for his work on the Statistical Physics of complex systems.

An avid advocate for Statistical Physics has always been our dear colleague Paolo Grigolini, whose work we honour in this book. Paolo’s work has changed many aspects of how we understand complex systems, and his most outstanding contributions are in the Statistical Physics of Non-equilibrium Systems. To quote from the foreword to volume 10 of the Landau-Lifshitz series, Physical Kinetics by Lifshitz and Pitaevski in 1978 [2]: "In contrast to the properties of statistical equilibrium systems, kinetic properties are far more connected with the microscopic interactions of specific physical objects. This is the cause for the massive diversity of these properties and the substantially higher level of difficulty of their theoretical analysis." Indeed, this diversity has been a driving force for exploration for many of us.

In the field of stochastic processes an outstanding scientist was Elliot Montroll, who came to fame when he successfully applied random walk theory to the behaviour of neutrons in the chain reaction in the Manhattan project. Montroll was central in formulating continuous time random walks and, ultimately, setting the scene for anomalous diffusion [3–6]. His heritage, continued by his collaborator Harvey Scher, his PhD student Mike Shlesinger, and his postdoc Bruce West, is still central for new developments and applications of stochastic processes. While the by-now classical textbooks of van Kampen [7] or Gardiner [8] concentrate on what we may call "classical dynamics" (Brownian motion, master equations, etc.), more recent textbooks such as those by Hughes [9], Klafter and Sokolov [10], or by West, Bologna, and Grigolini [11] make strong cases that anomalous diffusion has become "normal", as stated by Katja Lindenberg and colleagues [12].

In fact, statistical physics is an open, emerging field. One example is the recent development of stochastic thermodynamics [13] or efforts to come up with formulations for non-extensive systems, e.g., systems with long-range interactions for which the Gibbsian idea of subsystems no longer holds. One famed mathematical formulation taking such non-extensivity into account was the generalised entropy formulated by Tsallis [14]. Another field to mention are ongoing questions on the precise formulation of even classical entropy, as those posed by Stratonovich [15].

A number of concrete and experimentally relevant questions, on which Paolo has been working, include anomalous diffusion and its origins, the role of ergodicity, and ageing/non-stationarity in complex systems. In this spirit we will here address a main field of recent interest, prompted by the ongoing discovery of non-Gaussian statistics in relatively simple systems.

In the following we will use the term anomalous diffusion in the sense that the
associated mean squared displacement (MSD) of an ensemble is of the power-law form
\[ \langle x^2(t) \rangle = 2K_\alpha t^\alpha, \] (1)
where \( K_\alpha \) is the generalised diffusion coefficient of physical dimension cm\(^2\)/sec\(^\alpha\). Depending on the precise value of the anomalous diffusion exponent \( \alpha \), we typically distinguish subdiffusion (\( 0 < \alpha < 1 \)) and superdiffusion (\( \alpha > 1 \)) [16]. Specific cases are normal ("Brownian" or "Fickian") diffusion for \( \alpha = 1 \) and ballistic motion for \( \alpha = 2 \). The case \( \alpha = 3 \) is that of Richardson relative diffusion in turbulence [17].

When the increments of a stochastic process are independent and identically distributed (iid) variables \( x_i \) with a finite variance, as originally considered by Einstein [18] and Smoluchowski [19], their normalised sample average \( \sqrt{n}\sum x_i/n - \mu \) with mean \( \mu \) for \( n \) approaching infinity, converges in distribution to the normal or Gaussian probability density function (PDF) [20–22]
\[ P(x, t) = \frac{1}{\sqrt{4\pi K_1 t}} \exp\left(-\frac{x^2}{4K_1 t}\right), \] (2)
as written here for the case of normal Brownian diffusion with diffusivity \( K_1 \). Mathematically, this is due to the law of large numbers or, more stringently, the central limit theorem [22]. Violations of one or more of these conditions lead to anomalous diffusion and non-Gaussian statistics. Leaving the basin of attraction of the central limit theorem [23] causes the loss of the universality of the Gaussian law, in the sense that details of the considered process become more relevant and lead to a variety of emerging dynamics [24]. We could also say, things are getting interesting.

2. Non-Gaussian diffusion

While in his work Perrin used Einstein’s and Smoluchowski’s prediction of the Gaussian PDF (2) to evaluate his diffusion experiments of an ensemble of unbiased tracer particles, Kappler employed a torsional balance setup to measure the angular Brownian motion of a small mirror to map, with remarkable accuracy, the resulting Gaussian angle distribution [25]. With the much better experimental resolution of modern experiments, especially those using single particle tracking [26], and simulations, mapping out the PDF of stochastically or actively moving entities has become routine. However, more recently a number of systems report significant deviations from the Gaussian statistic, and such non-Gaussian diffusion has attracted significant attention from both experimentalists and modellers. After a brief summary of different systems reporting non-Gaussian PDFs we will introduce several possible stochastic approaches to explain such non-Gaussianity.

\( \S \) We will mostly use a one-dimensional notation, generalisations to higher dimensions are fairly straightforward.
2.1. Phenomenology

While one tacitly assumes that the occurrence of the Brownian (or Fickian) linear scaling \( \langle x^2(t) \rangle \simeq K_1 t \) in time of the MSD implies a Gaussian shape of the PDF, intermittent departure from a Gaussian statistic was reported from disordered solids (glasses, supercooled liquids) [27–29] and interfacial dynamics [30,31]. The ubiquitousness of "Brownian yet non-Gaussian" (BnG) diffusion was popularised in the field of soft and biological matter by Granick in their mini-review [32]. Two specific systems addressed there are colloidal beads diffusing along tubes made up of phospholipid bilayer (the simplest building blocks for biological membranes) and colloidal beads moving in entangled actin hydrogels. In the actin gel system they observe an exponential shape ("Laplace distribution") of the PDF of the form [32,33]

\[
P(x, t) = \frac{1}{2\lambda(t)} \exp\left( -\frac{|x|}{\lambda(t)} \right),
\]

with the decay length \( \lambda(t) \simeq t^{1/2} \) scaling like the square root of time. As can be easily checked this PDF is normalised on the interval \( x \in (-\infty, \infty) \) and indeed encodes the Brownian scaling \( \langle x^2(t) \rangle \simeq \lambda^2(t) \simeq t \). In contrast, in the lipid bilayer tube system a crossover was observed: for times longer than some characteristic time the Laplace distribution reverts to a Gaussian shape. In both systems the Fickian scaling \( \langle x^2(t) \rangle \simeq t \) with a stationary prefactor is observed at all times. Similar examples for diffusive motion yet exponential tails were reported for tracer diffusion in suspensions of swimming microorganisms [34] and colloidal nanoparticles adsorbed to fluid interfaces [35–37]. Similarly, BnG was observed for the motion of nematodes [38]. In a detailed analysis using single particle tracking of fluorescently labelled colloidal particles in an array of micropillars, the non-Gaussianity of the resulting ensemble PDF was shown to be due to an apparent position-dependent, heterogeneous particle diffusivity, plus a heterogeneous particle distribution [39]. While the overall MSD was linear (i.e., Fickian), depending on the micropillar density and the randomness of their placement in space, the study mapped out the PDFs of the diffusivities and the apparent anomalous diffusion exponent \( \alpha \) for individual particles. Especially the PDFs for \( \alpha \) turn out to be quite broad. A recent example for BnG dynamics is nanoparticle transport in a graphene liquid cell [40].

Gaussianity is also a hallmark of certain anomalous diffusion processes. The best known is fractional Brownian motion (FBM), going back to Kolmogorov [41] as well as Mandelbrot and van Ness [42]. FBM is described in terms of an overdamped Langevin equation \( \dot{x}(t) = \xi_\alpha(t) \), where \( \xi_\alpha(t) \) is the driving noise. While Brownian motion corresponds to a white Gaussian form for \( \xi_1(t) \), in FBM the noise remains Gaussian yet is long-ranged correlated. Specifically, the autocorrelation of the noise is stationary and of the power-law form \( \langle \xi_\alpha(t)\xi_\alpha(t+\tau) \rangle = D_\alpha \alpha(\alpha - 1)\tau^{\alpha - 2} \), at long times [24,43,44]. For subdiffusion (0 < \( \alpha < 1 \)), the noise-noise correlator has a negative sign, mirroring so-called antipersistence, while for superdiffusion (1 < \( \alpha < 2 \)) a persistent, positively correlated power-law in \( \tau \) is followed. FBM of subdiffusive type is particularly found to
characterise diffusion in viscoelastic systems in the overdamped limit.\[\]

In a range of systems viscoelastic diffusion with a non-Gaussian PDF was observed. Thus, single particle tracking in bacteria and yeast cells demonstrated clearly an exponential distribution of apparent particle diffusivities along with a Laplace shape of the particle PDF \[47\] (compare also the results in \[48, 49\]). Non-Gaussian diffusion along with pronounced subdiffusion was also found for the motion of phospholipid molecules in bilayer membranes at sub-nanosecond times \[50\]. A dynamic crossover from subdiffusion to normal diffusion and an approximately exponential displacement PDF was reported for the motion of acetylcholine receptors in the membranes of living biological cells \[51\]. Both lipids and membrane proteins were shown to exhibit pronounced non-Gaussian PDFs in protein-decorated lipid bilayer membranes at higher crowding fractions \[52\]: here the PDF was of a stretched Gaussian form, \( P(x, t) \approx \exp(-c|x|^{\kappa}) \) with \( 1 < \kappa < 2 \).

Similar stretched Gaussian shapes were also observed in the (mostly) superdiffusive, active motion of dictyostelium discoideum amoeba cells spreading on a surface \[53\]; an interesting observation here is that, within the experimentally accessible time window, the displacement PDF becomes more exponential with increasing lag time, i.e., the stretching exponent \( \kappa \) tends to unity, contrasting the crossover to a Gaussian in the colloids-on-nanotube experiment of \[32\] or in the graphene liquid cell experiments in \[40\].

A surprisingly rich dynamic behaviour was found for the lateral diffusion of doxorubicin drug molecules in between two silica slabs \[54\]: the displacement PDF is pronouncedly non-Gaussian, but the motion is also antipersistent and non-ergodic. Crossover from subdiffusion to a plateau of the MSD along with pronounced non-Gaussianity was reported for tracers in an active DNA gel with FtsK50C molecular motors \[55\]. A rich dynamic behaviour was also reported from glass-forming Lennard-Jones systems \[56\]: here the scaling of the length scale \( \lambda(t) \) in the Laplace PDF has the diffusive scaling \( \lambda(t) \simeq t^{1/2} \) in the 2D case but exhibits \( \lambda(t) \simeq t^{1/3} \) in 3D and \( \lambda(t) \simeq t^{1/4} \) in 4D. A crossover from early subdiffusion to BnG was found in quasi-2D suspensions of colloidal beads in a spatially random yet static optical force field \[57\].

From simulations of the motion of particles in 2D static disordered landscapes non-Gaussian behaviour were rationalised in \[58\], and especially the role of an additional peak in the centre of the displacement PDF discussed \[59\]. In fact, the precise relaxation dynamics of the central peak in heterogeneous systems was scrutinised in detail recently \[60\]. From extensive computer simulations the role of the system initiation, i.e., randomly initiated versus equilibrated particle positions, in BnG was revealed in \[61\]. In the context of non-Gaussian diffusion in quenched landscapes we finally note that random walks visiting traps with exponentially distributed depths, as well as the annealed continuous time random walk limit with scale-free waiting time PDFs always have stretched exponential displacement PDFs \[6, 16, 23, 62–65\].

\[\]

\( ^{\|}\) Strictly, viscoelastic diffusion should be described in terms of the generalised Langevin equation with power-law memory kernel \[43, 45, 46\], but we here base our discussion on the simpler FBM.

\[\]
2.2. Quantifying non-Gaussianity

Before turning to concrete models to describe BnG we first mention how BnG can be quantified. One concrete piece of information already mentioned is the diffusion length $\lambda(t)$, from which the $x$-$t$ scaling can be inferred. Non-Gaussianity as such, of course, needs to be identified from higher order moments. Typically, for a centred process one uses the kurtosis $K = \langle x^4(t) \rangle / \langle x^2(t) \rangle^2$. For a Gaussian, for instance, $K = 3$ in 1D and $K = 2$ in 2D. Platykurtic PDFs with $K$ values smaller than that for a Gaussian, have thinner tails than a Gaussian and are sometimes termed sub-Gaussian [66]. Leptokurtic PDFs, in contrast, have a higher $K$ value than a Gaussian and thus thicker tails (“super-Gaussians” [67]), for instance, the Laplace PDF has $K = 9$ in 1D and $K = 4$ in 2D. An alternative to the kurtosis is the non-Gaussian parameter $[68,69] (d-2)!d^2K/(2+d)!! - 1$ in $d$ dimensions

A recently introduced measure for non-Gaussianity is the codifference [70]

$$\tau_\theta(t) = \frac{1}{\theta^2} \ln \frac{\langle \exp(i\theta[x_{s+t} - x_s]) \rangle}{\langle \exp(i\theta x_s + t) \rangle \langle \exp(-i\theta x_s) \rangle}$$

(4)

of a random process $x_t$ with the continuous parameter $\theta$. It can be argued that the codifference pays more weight to the bulk of the underlying PDF, in contrast to the covariance. For Gaussian variables with the variance $\sigma^2$ it has the simple form $\exp(-\theta^2\sigma^2)/2$ and has the characteristic behaviour of a memory function. From detailed analysis it can be shown that the codifference (4) is in fact a suitable measure to detect non-Gaussianity (and non-ergodic behaviour) [70]. We also mention the availability of methods based on random coefficient autoregressive approaches, that can be mapped on BnG-style models (see below) [71].

2.3. Superstatistical approaches

Granick and coworkers proposed that the non-Gaussian displacement statistic $P(x,t)$ can be understood in terms of the superposition

$$P(x,t) = \int \int \ldots \int G(x,t|D)p(D)dD,$$

(5)

where $G(x,t|D)$ represents a Gaussian of the form (2) for a specific value $D$ of the diffusion coefficient, and $p(D)$ is a PDF of $D$ values. In this view each particle performs normal diffusion but with a different $D$. This scenario could simply represent particles of different sizes or particles moving in patches of different local properties. The MSD of this process reads

$$\langle x^2(t) \rangle = 2t \int \int \ldots \int Dp(D)dD = 2\langle D \rangle t,$$

(6)

that is, the MSD is linear with the effective diffusion coefficient $\langle D \rangle$. The approach (5) is in fact the "superstatistics" approach by Beck and Cohen [73], in the sense that

¶ In the latter case the formulations only holds as long as the particles do not cross boundaries between patches with other $D$ values—in contrast to the annealed transit time model [72].
the statistical behaviour of the Gaussian PDF $G(x,t|D)$ is modulated by additional averaging over the diffusivity PDF $p(D)$. In this superstatistical approach one can show that the Laplace PDF (3) corresponds uniquely to an exponential form of $p(D)$ [74]. In [38] a gamma distribution for $p(D)$ was used, compare also [75]. Power-law forms of $p(D)$ lead to superstatistical forms of the PDF (5) with power-law tails [74, 76], another relevant form for a superstatistical PDF are stretched Gaussians [74, 77]. Superstatistical approaches were also formulated for non-Fickian, anomalous diffusion dynamics, especially extensions of the generalised Langevin equation with random parameters [78, 79]. In particular, in [79] it is shown that this formulation may also lead to more exotic shapes for the PDF, such as a Cauchy law.

Historically, concepts similar to superstatistics were used before, notably, in the field of turbulence, where the refined similarity hypotheses take into account fluctuations of energy dissipation and lead to intermittent corrections to the famous -5/3 spectrum of energy in the inertial interval. In this setting non-Gaussian cascades were considered to emerge from "statistical mixing" by Obukhov and Kolmogorov [80–82]. This idea was further developed by Castaing and coworkers [83, 84] and is related to the scale dependence of velocity increments in turbulent systems [85, 88]. Other fields using concepts similar to superstatistics include financial mathematics ("compounding") [86] and the study of fracture processes [87].

A process closely related to superstatistics is generalised grey Brownian motion (ggBM), defined in terms of the stochastic equation [89–93]

$$x_{ggBM}(t) = \sqrt{2DB(t)}$$

for the particle trajectory $x_{ggBM}(t)$, see also the discussions in [70, 75]. $D$ here is now a random diffusivity. The core idea is that different yet physically identical particles move in disjointed areas in which their diffusivity is different—in other words, the essential view of superstatistics. We could also think of physically different particles, each with a different diffusivity, in a homogeneous environment. GgBM includes anomalous diffusion scenarios, as detailed further in [90, 92].

### 2.4. Diffusing-diffusivity models

In typical (anomalous) diffusion models we assume that a particle is characterised by a given diffusion coefficient or noise strength. This includes the superstatistical approach, in which each particle has its own diffusion coefficient, that does not change in time. This is not always justified. For instance, lipid molecules in crowded bilayer membranes are found to exhibit intermittent dynamics switching between high and low diffusivity modes, and this behaviour can be mimicked by hard core particles moving through a fixed obstacle environment [52]. Another scenario is that a particle is moving in a hydrogel with (sufficiently fast) local fluctuations of mesh sizes, such that on its way the particle experiences a varying degree of obstacle densities [94].

Another scenario emerges for certain proteins moving freely in aqueous solution. These protein molecules can then be shown to exhibit continuous changes between
conformations, some of which are compact while others are extended. These conformations correspond to different local minima of the free energy landscape of the protein. Significant temporal fluctuations of the effective size (measured in terms of the gyration radius $R_g$) and thus the hydrodynamic radius of such a perpetually shape-shifting protein molecule were demonstrated to effect a stochastic instantaneous diffusivity. This instantaneous diffusivity fulfils a Stokes-Einstein-type relation with $R_g$ [95]. As further detailed in section 2.6 tracer particles may also exhibit a stochastically changing diffusivity due to ongoing (de)polymerisation size-variations.

The motion of a tracer particle with a stochastically evolving diffusivity can be modelled in terms of a Langevin equation in the so-called "diffusing diffusivity" approach [96]. This model was further developed in [76, 94, 97–99]. In a minimal formulation the diffusing-diffusivity model is captured by the set of coupled stochastic equations [74]+

$$\frac{d}{dt} \tau(t) = \sqrt{2D(t)}\xi(t),$$  \hspace{1cm} (8a)
$$D(t) = y^2(t),$$  \hspace{1cm} (8b)
$$\frac{d}{dt} y(t) = -\frac{1}{\tau} y + \sigma \eta(t).$$  \hspace{1cm} (8c)

Equation (8a) here represents the Langevin equation for the particle position $x(t)$, driven by the white Gaussian noise $\xi(t)$. In contrast to the standard Langevin equation, however, the diffusion coefficient $D(t)$ is explicitly time-dependent. The dynamics of this noise strength $D(t)$ is specified by equations (8b) and (8c). First, to vouchsafe positivity of the diffusivity, we write $D(t)$ as the square of the auxiliary variable $y(t)$. The dynamics of $y(t)$ in expression (8c) is that of an Ornstein-Uhlenbeck process [7], that is, diffusion driven by the white Gaussian noise $\eta(t)$ in the presence of a Hookean restoring force. The motion $y(t)$ is thus confined by an harmonic potential and will reach equilibrium beyond the correlation time $\tau$. The physical dimension of the noise strength in the Ornstein-Uhlenbeck process for the auxiliary variable $y(t)$ here is $[\sigma] = \text{cm/sec}$.

The diffusing-diffusivity dynamics encoded in equations (8) can be shown to reproduce the superstatistical exponential tails (with dimension dependent power-law correction) at times shorter than $\tau$,

$$P(x, t) \sim \frac{1}{\sqrt{2\pi|x|\sigma(\tau t)^{1/2}}} \exp \left(-\frac{|x|}{\sigma(\tau t)^{1/2}}\right).$$  \hspace{1cm} (9)

At times $t \gg \tau$, when correlations in the diffusivity process $D(t)$ (or, better, in the auxiliary variable $y(t)$) are relaxed, the Gaussian

$$P(x, t) \sim \frac{1}{\sqrt{4\pi\langle D \rangle t}} \exp \left(-\frac{x^2}{4\langle D \rangle t}\right)$$  \hspace{1cm} (10)

is recovered. Concurrently to the crossover from Laplace-type PDF to a Gaussian shape, the MSD of the minimal diffusing-diffusivity process (8) is given by the law

$$\langle x(t) \rangle = 2\langle D \rangle t, \quad \langle D \rangle = \frac{1}{2} \sigma^2 \tau$$  \hspace{1cm} (11)

+ In [74] the model is considered in arbitrary dimensions for position $x$ and auxiliary variable $y$. 
at all times, as long as an equilibrium initial condition for \( y(t) \) is chosen. Moreover, the initial non-Gaussianity can be monitored in terms of the kurtosis \( K = \frac{\langle x^4(t) \rangle}{\langle x^2(t) \rangle^2} \), that shows a crossover from the short-time value \( K \sim 9 \), the value for an exponential PDF, to \( K \sim 3 \), the value for a Gaussian, in the 1D case. All these results were also shown to be in full agreement with stochastic simulations [74].

We note that more technically, the minimal diffusing-diffusivity model (8) can be shown to correspond to a dynamic subordinated to a Brownian diffusion process, and it can be formulated in terms of a bivariate Fokker-Planck equation [74]. In [75] the model was extended to a generalised gamma distribution in the superstatistical, short-time limit, and the initial condition for \( y(t) \) was generalised to non-equilibrium initial conditions, leading to distinct crossover behaviours in the MSD. We finally note that diffusing-diffusivity models analogous to the minimal model exist, in different language, in mathematical finance. Notably, the Heston model [100] with its stochastic volatility. In turn the Heston model is a specific case of the Cox-Ingersoll-Ross model [101]. We also note that the crossover behaviour governed by equations (8) with the stationary dynamics for the stochastic diffusion coefficient \( D(t) \) is generically different from formulations with diffusion coefficients \( D(t) \) based on deterministic dynamics such as scaled Brownian motion, effective FBM models in absence of boundaries, or combinations based on such processes [102–107].

The generic behaviour encoded in the minimal diffusing-diffusivity approach of equations (8) is quite robust and matches (up to minor details) the short- and long-time behaviour of the other diffusing-diffusivity models developed in [76, 94, 96–99]. What happens when we combine the diffusing-diffusivity idea with Gaussian anomalous diffusion of the FBM type? This was analysed in [108] where the Ornstein-Uhlenbeck dynamic (8c) for the time-dependent diffusivity \( D(t) \) in (8b) was combined with a Langevin equation \( \dot{x}(t) = \sqrt{2D(t)} \xi_\alpha(t) \) driven by fractional Gaussian noise chosen in the Mandelbrot-van Ness smoothed form \( \langle \xi_\alpha(t) \xi_\alpha(t+\tau) \rangle = (2\delta^2)^{-1}(|\tau+\delta\alpha-2|\tau\alpha+|\tau-\delta\alpha) \) for the noise autocorrelation and small constant \( \delta \). For antipersistent noise \( \xi_\alpha \) with \( 0 < \alpha < 1 \) a crossover from initial subdiffusive scaling of the MSD to normal-diffusive scaling was obtained. In contrast, for persistent noise \( (1 < \alpha < 2) \) both short- and long-time scaling of the MSD was found to be superdiffusive. As desired, the PDF crosses over from a Laplace-type distribution at short times to a long-time Gaussian. The effective diffusivity as function of the anomalous diffusion exponent exhibits a discontinuity around \( \alpha = 1 \). This behaviour was demonstrated to be different from the generalised model of [98] and the two-state model developed in [109]. Thus, the interplay of the long-ranged noise correlations and the crossover dynamics of the diffusion coefficient effect a quite intricate, non-universal dynamics. In fact, the often unexpected behaviour of FBM was recently also highlighted in its behaviour in the presence of reflecting boundaries with possible connection to brain fibre density fields [110–116].

We finally mention that diffusing-diffusivity dynamics in association with active Brownian motion was recently analysed in [117], finding a crossover from a short-time exponential shape of the PDF to a long-time Gaussian form.
2.5. Large-deviation approach

While we mentioned observations of non-Gaussian statistics in the form of stretched Gaussian shapes, the majority of cases reported involve Laplace PDFs with their characteristic exponential tails. In terms of a random walk approach the emergence of such unconventional tails can be understood by extreme-value arguments, as demonstrated in [118, 119] for general continuous-time random walk processes. The central argument goes as follows: when we derive the Gaussian limit PDF using the law of large numbers or the central limit theorem, the main condition is the limit of many jumps, that is, the long-time limit. What is the shape of the tails when the number of jumps or time is finite? In fact, the tails of the PDF $P(x, t)$ are then dominated by extreme events. Applying the theorems of large-deviation statistics, the leading contribution to the PDF in the limit $|x|/t \to \infty$ is given by [118]

$$P(x, t) \sim \exp \left( -\kappa \log \left( \frac{|x|}{t} \right)^{1-1/\beta} \frac{|x|}{t} + C \right) t.$$ (12)

i.e., leading exponential tails with power-law corrections. The authors state that "the exponential tails for the positional PDF are rather a rule and not an exception. The exponential decay of the tails is a general feature exactly like the Gaussian behaviour (that is dictated by the CLT [central limit theorem]) at the centre." Note the somewhat different form of the large-deviation result (12) compared to the BnG form (3): in (12) no scaling between $x$ and $t$ occurs, in analogy to the results reported in [28].

2.6. Polymerisation models

In the above example of the perpetually shape-shifting protein molecule [95] the stochastic diffusivity dynamics was effected by the change of the effective protein size as measured by the gyration radius. A similar effect can be observed when the tracer particle is growing or shrinking. In fact, ARG3 messenger RNA molecules were reported to occur with a broad distribution of diffusivities, associated, inter alia, with conglomeration with one or multiple mRNA and other associated entities such as RNA-binding proteins [120].

In a simple polymerisation dynamics approach $A_N + A_1 \overset{k_+}{\underset{k_-}{\rightleftharpoons}} A_{N+1}$ describing the addition of a monomer $A_1$ to a polymer $A_N$ containing $N$ monomers with rate $k_+$ and the backwards reaction, chains with a time-dependent, fluctuating polymerisation degree are being formed [121]. If a monomer has diffusivity $D_0$ then a Stokes-Einstein-type relation emerges in the form $D_N \simeq D_0/N^\nu$, where the scaling exponent depends on the specific polymer model. Thus, for the Rouse limit (Gaussian chain), $\nu = 1/2$, $\nu = 1$ for the Zimm model when hydrodynamic interactions are included, and $\nu = 2$ in the reptation model when the tagged polymer moves in a solution of entangled polymers [122]. Detailed analysis and simulations then demonstrate that the resulting diffusion of such a polymer with fluctuating size is non-Gaussian at short times and
crosses over to a Gaussian diffusion at long times, corresponding to the equilibrium size distribution of the polymer chain. It is shown in [121] that the kurtosis in the non-Gaussian state can assume extremely high values. A complementary analysis based on simulations and analytical arguments is presented in [123].

2.7. Random-diffusivity models

The concept of diffusing-diffusivity is further carried on in [124] based on a Langevin equation of the form \( \dot{x}(t) = \sqrt{2D_0 \Psi_t} \xi(t) \), where \( \xi \) is white Gaussian noise and \( \Psi_t \) a positive-definite random function of time. A squared Ornstein-Uhlenbeck process for \( \Psi_t \) leads back to the minimal diffusing-diffusivity model (8), while other choices discussed in [124] are jump processes with Gamma and Lévy-Smirnov distributions, as well as the cases of squared Brownian motion (\( \Psi_t = B(t)^2 \)), rectified Brownian motion (\( \Psi_t = \Theta(B(t)) \), where \( \Theta(\cdot) \) is the Heaviside step function), and geometric Brownian motion (\( \Psi_t = \exp(-B(t)/a) \)). The emerging non-Gaussian shapes for the displacement PDF and the corresponding single trajectory power spectra (see [125,126]) are calculated for each case.

2.8. Random-coefficient autoregressive models

The Langevin equation approach is a standard approach in non-equilibrium statistical physics. In other fields such as financial mathematics, so-called autoregressive models are widespread [127]. The connection between both can be directly established in certain cases. It can then be shown that if we start from the generalised Langevin equation \( dv(t) = -\Lambda(t)v(t) + \sqrt{D(t)}dB(t) \) with the time-dependent friction and diffusion coefficients \( \Lambda(t) \) and \( D(t) \), and where \( B(t) \) represents Brownian motion (Wiener process), this dynamics can be mapped onto a random-coefficient autoregressive model (rcAR). Namely, discretisation using \( v_k = v(k\Delta t) \) with small but finite time delay \( \Delta t \) and integer \( k \), leads to the approximation

\[
 v_k - (1 - \Lambda_k \Delta t)v_{k-1} = \sqrt{D_k \Delta B_k}. \tag{13}
\]

This is indeed an autoregressive process of the class rcAR(1) with AR coefficient \( 1 - \Lambda_k \Delta t \) [71]. Based on such mappings one can then use the well-established methods of data analysis in autoregressive modelling to identify measured data exhibiting BnG [71].

2.9. Mobile-immobile models

We finally mention a simple model describing relaxation in a system with two populations of particles, mobile and immobile ones, that mimics some features of BnG. As was demonstrated in [128] Brownian diffusion in a dilute field of traps is Fickian but non-Gaussian. Briefly, the starting point are the coupled equations for the mobile \( (n_m) \)
and immobile \((n_{im})\) particle concentrations,
\[
\frac{d}{dt}n_m(r,t) = -\beta n_m(r,t) + \frac{1}{\tau} n_{im}(r,t) + D \nabla^2 n_m(r,t),
\]
\(14a\)
\[
\frac{d}{dt}n_{im}(r,t) = \beta n_m(r,t) - \frac{1}{\tau} n_{im}(r,t)
\]
\(14b\)
where \(r(t) = (x(t), y(t))\) is a two-dimensional vector. The initial conditions are \(n_m(r,0) = N_0\delta(r)\) and \(n_{im}(r,0)\). Note than in [128] equilibrium initial conditions were used. Such a choice may be more natural for some systems, however, this choice weakens the net effect. For that reason we here choose the non-equilibrium condition that all particles initially are mobile.

The Laplace transform \(P(r,s) = \mathcal{L}\{P(r,t)\} = \int_0^\infty P(r,t)\exp(-st)dt\) of the PDF is obtained after Fourier-Laplace transformation of equations \((14)\) in terms of the modified Bessel function \(K_0\) as
\[
P(r,t) = \frac{1}{N_0} \left( n_m(r,s) + n_{im}(r,s) \right) = \frac{1}{2\pi D}s \varphi(s) K_0 \left( r \sqrt{\varphi(s)/D} \right),
\]
\(15\)
where
\[
\varphi(s) = \frac{s}{s + 1/\tau} \left( s + \beta + \frac{1}{\tau} \right).
\]
\(16\)
The MSD encoded in this PDF reads
\[
\langle r^2(t) \rangle = \frac{4D}{1 + \beta\tau} \left( t + \frac{\beta\tau^2}{1 + \beta\tau} \left[ 1 - \exp \left( -\frac{1 + \beta\tau}{\tau} t \right) \right] \right).
\]
\(17\)
As can be seen from equations \((15)\) and \((17)\), for \(\beta\tau \gg 1\) the MSD grows linearly at short and long times while the PDF is Gaussian. However, at intermediate times \(\beta^{-1} \ll t \ll \tau\) the MSD exhibits a plateau-like behaviour while the PDF has the time-independent shape
\[
P(r,t) \approx P(r) = \frac{\beta}{2\pi D} K_0 \left( \sqrt{\frac{\beta}{D}} r \right) \simeq \exp \left( -\sqrt{\frac{\beta}{D}} r \right),
\]
\(18\)
with exponential tails. Thus, the crossover from the plateau- to normally diffusive-regime in the MSD is accompanied by a crossover from exponential to Gaussian behaviour in the shape of the PDF.

3. Conclusions

Non-Gaussianity has been observed in a large number of systems in many different systems. Similar to the occurrence of non-Brownian, anomalous diffusion, non-Gaussianity breaks with one of the dogmas of conventional statistical physics, the predominance of the central limit theorem. The MSD in these processes can be Fickian or exhibit anomalous scaling, while the displacement PDFs can assume a Laplace PDF with exponential tails, stretched Gaussian shapes, or even power-laws. In a given
experiment or simulation, the non-Gaussian character is either preserved within the accessible experimental window of probed time scales, or crossover behaviours can be observed. The latter typically show the emergence of a Gaussian PDF beyond some correlation time, although other observations exist. A growing number of stochastic processes are being devised to describe this non-Gaussian behaviour.

The approaches reviewed here correspond to annealed models, in which the instantaneous value of the particle diffusivity is unconnected from the particle’s specific position in space. This is justified when the particles of an ensemble themselves have a distribution of diffusivities, and then the superstatistical description with its time-independent diffusivity PDF serves as the apt physical description. Other scenarios for annealed descriptions are those of shape-shifting or polymerising tracers, or when the environment changes rapidly enough such that the particle experiences a renewed value of its local mobility compared to its previous visit to that position. In three dimensions the annealed models provide reasonable approximations even if the environment does not change rapidly, as here the probability to revisit specific positions is relatively low. Models with explicit quenched disorder, when correlations in the particle motion build up while the particle samples the same mobility values each time it returns to previously visited points, investigating the emerging non-Gaussianity of the diffusive spreading include predominantly simulations-based information [56, 58, 59, 61, 129]. Finding analytical access to the quantitative modelling in such cases remains a challenge. Equally elusive is the understanding of the peculiar dimension dependence of the scaling $\lambda(t) \simeq t^{1/d}$ with spatial dimension $d = 2, 3, 4$ in the Lennard-Jones system in [56].

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