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Non-Markovian effects on the Brownian motion of a free particle

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ABSTRACT

Non-Markovian effects on the Brownian movement of a free particle in the presence as well as in the absence of inertial force are investigated under the framework of generalized Fokker–Planck equations (Rayleigh and Smoluchowski equations). More specifically, it is predicted that non-Markovian features can diminish the values of both the root mean square displacement and the root mean square momentum, thereby assuring the mathematical property of analyticity of such physically observable quantities for all times $t \geq 0$. Accordingly, the physical concept of non-Markovian Brownian trajectory turns out to be mathematically well defined by differentiable functions for all $t \geq 0$. Another consequence of the non-Markovicity property is that the Langevin stochastic equations underlying the Fokker–Planck equations should be interpreted as genuine differential equations and not as integral equations according to a determined interpretation rule (Doob's rule, for instance).

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1. Introduction

A particle with mass m immersed in a generic environment (e.g., a non-thermal fluid) undergoes a jittering movement dubbed Brownian motion. The motion of this tagged particle may be mathematically described by the Langevin equation [1–10]

$$m \frac{d^2 X}{dt^2} = -\frac{dV(X)}{dX} - \gamma m \frac{dX}{dt} + b\psi(t), \quad (1)$$

where the displacement $X = X(t)$ of the Brownian particle depends on the evolution time t . In the Langevin equation (1), the inertial force $m d^2 X / dt^2$ offsets a set of three sorts of forces: a conservative force derived from a potential energy $V(X)$ inherent in the tagged particle, and two environmental forces: a linearly velocity-dependent dissipative force $F_d = -\gamma m dX / dt$, which accounts for stopping the particle's motion due to the damping parameter $\gamma \geq 0$, as well as an anti-dissipative force $L(t) = b\psi(t)$, dubbed Langevin's force, responsible for activating the particle's movement. The parameter $b \geq 0$ controls the environmental influence (fluctuations) on the Brownian particle and is in dimensions of [mass \times length \times time $^{-3/2}$] provided that the function $\psi(t)$ has dimensions of [time $^{-1/2}$].

From the mathematical viewpoint, the quantities $X = X(t)$ and $\Psi = \Psi(t)$ in the Langevin equation (1) are interpreted as random variables [1–10] in the sense that there exists a probability distribution function, $\mathcal{F}_{X\Psi}(x, \psi, t) \geq 0$, associated with the stochastic system $\{X, \Psi\}$, expressed in terms of the possible values (realizations) $x = \{x_i\}$ and $\psi = \{\psi_i\}$, with $i \geq 1$, distributed about the sharp values q and φ of X and Ψ , respectively. In addition, it is assumed that the average value of any physical quantity $A(X, \Psi, t)$ can be calculated as

$$\langle A(X, \Psi, t) \rangle = \iint_{-\infty}^{+\infty} a(x, \psi, t) \mathcal{F}_{X\Psi}(x, \psi, t) dx d\psi, \quad (2)$$

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where the function $a(x, \psi, t)$ stands for the possible realizations of the random function $A(X, \Psi, t)$. Definition (2) fulfills the normalization condition

$$\langle 1 \rangle = \iint_{-\infty}^{+\infty} \mathcal{F}_{X\Psi}(x, \psi, t) dx d\psi = 1. \quad (3)$$

As far as a free particle $V(X) = 0$ is concerned, the stochastic equation (1) may be written in terms of the linear momentum $P \equiv P(t) = m dX/dt$ as

$$\frac{dP}{dt} = -\gamma P + b\Psi(t). \quad (4)$$

On the other hand, in the absence of inertial force, i.e., $m d^2X/dt^2 = 0$, the free Brownian particle turns out to be described by the random equation

$$\frac{dX}{dt} = \frac{b}{\gamma m} \Psi(t). \quad (5)$$

In the literature on Brownian motion [1–15] it is used to assume that the Langevin force $L(t)$ has the following Gaussian statistical properties

$$\langle L(t) \rangle = b \langle \Psi(t) \rangle = 0, \quad (6)$$

$$\langle L(t')L(t'') \rangle = b^2 \langle \Psi(t')\Psi(t'') \rangle = 2D\delta(t' - t''), \quad (7)$$

where $D = b^2/2$ is a constant measuring the noise intensity and $\langle \dots \rangle$ denotes the average value evaluated over the probability distribution function associated with the environmental random function $L(t)$ or $\Psi(t)$, according to the prescription (2). Property (6) is meant to characterize the irregularity feature of the Langevin force so implying that there is no averaging effect on the Brownian motion; whereas assumption (7) stands for that the interaction between the tagged particle and a generic environment is deemed to be Markovian in the sense that the autocorrelation function of the Langevin fluctuating force is delta-correlated (white noise), i.e., $L(t')$ and $L(t'')$ are assumed to be completely independent for arbitrarily small time $|t' - t''|$.

For a free Brownian particle immersed in a thermal reservoir characterized by the temperature T and the Boltzmann constant k_B , the Ornstein–Uhlenbeck process (4) gives rise in the Gaussian approximation (6) and (7) to the following Fokker–Planck equation (the so-called Markovian Rayleigh equation [5,13])

$$\frac{\partial \mathcal{F}(p, t)}{\partial t} = \gamma \frac{\partial}{\partial p} [p \mathcal{F}(p, t)] + \gamma m k_B T \frac{\partial^2 \mathcal{F}(p, t)}{\partial p^2}, \quad (8)$$

which in turn leads to the root mean square momentum

$$\Delta P(t) \equiv \sqrt{\langle p^2 \rangle - \langle p \rangle^2} = \sqrt{m k_B T (1 - e^{-2\gamma t})}, \quad (9)$$

whereas the stochastic equation (5) together with the statistical properties (6) and (7) yields the Markovian Smoluchowski equation [16–18] (or diffusion equation)

$$\frac{\partial f(x, t)}{\partial t} = \frac{k_B T}{\gamma m} \frac{\partial^2 f(x, t)}{\partial x^2}, \quad (10)$$

which provides the Einstein's root mean square displacement [1,17,18]

$$\Delta X(t) \equiv \sqrt{\langle X^2 \rangle - \langle X \rangle^2} = \sqrt{\frac{2k_B T}{\gamma m} t}. \quad (11)$$

Both Markovian results (9) and (11) are non-differentiable functions at $t = 0$. Hence, it has been claimed that physically there is no force, $F(t) = \frac{d\Delta P(t)}{dt}$, acting on a free Brownian particle in the presence of inertial forces, as well as no velocity, $\mathcal{V}(t) = \frac{d\Delta X(t)}{dt}$, in the absence of inertial force [2,15]. In other words, such a non-analyticity property implies that the concept of trajectory of a free Brownian particle seems to be an elusive feature in the Markovian regime [2,9,15,18]. Accordingly, from the mathematical standpoint the Langevin stochastic equation (1) [or Eqs. (4) and (5)] should properly be interpreted not as a differential equation but as an integral equation according to Doob's rule [2,5–9,15]. Yet, it has been argued that the Markov property (7) is a highly idealized feature [3,6,13,14,19] because any physical interaction between the Brownian particle and environment actually comes about for a finite correlation time. By the same token, a decade ago van Kampen [14] had laconically pointed out: “*Non-Markov is the rule, Markov is the exception*”. In brief, hypothesis (7) seems to be a flawed starting point for fathoming the true physical nature of Brownian movement.

The inadequacy of Markov hypothesis, perhaps, accounts for the huge proliferation of papers that cope with the problem of non-Markovian behavior of Brownian motion. A possible approach is to derive generalized master equations and examine

the equivalence with the continuous-time random walks framework [20]. Another account is to study non-Markovian effects brought about by quadratic noise [21]. Moreover, molecular derivations of generalized Fokker–Planck equations with retarded kernels has been developed in Refs. [22–25]. More recently, stochastic processes described by non-Markovian Fokker–Planck equations have been investigated in Refs. [26,27].

Another way of looking at the non-Markovian Brownian motion is by means of the generalized Langevin equation [28–30]

$$m \frac{d^2X}{dt^2} = -\frac{dV(X)}{dX} - \int_0^t \beta(t' - t'') \frac{dX(t')}{dt'} dt' + L(t), \tag{12}$$

put originally forward by Mori [28] on the grounds of the assumption that there exists a close relationship between memory effects ingrained in the friction kernel $\beta(t' - t'')$ and non-Markovian effects (colored noise) showing up in the autocorrelation function $\langle L(t')L(t'') \rangle$ given by $\langle L(t')L(t'') \rangle = \frac{b}{\gamma} \beta(t' - t'')$. In the Markovian case, i.e., $\beta(t' - t'') = 2\gamma \delta(t' - t'')$, the Markovian Langevin equation (1) is recovered from (12).

The non-Markovian Fokker–Planck equation corresponding to (12) has been built up in Refs. [31–36] for both the free particle and the harmonic oscillator. Furthermore, generalizations of the Adelman’s non-Markovian Fokker–Planck equation [31] have been achieved in Refs. [37–40].

It is worth stressing that such non-Markovian approaches [20–40] to Brownian motion do not show explicitly how non-Markovian effects affect both the root mean square momentum (9) and displacement (11). Further, even for the case of the non-Markovian Langevin equation (12) it has been reported that the concept of force $m \frac{d^2X}{dt^2}$ does not exist because the Brownian paths are assumed to be non-differentiable. Consequently, like Eq. (1), the generalized Langevin equation (12) should be rigorously interpreted as an Itô–Doob integral equation and not as a differential equation [41].

The purpose of the present paper is to show explicitly how non-Markovian effects on the Brownian motion undergone by a free particle can be responsible for the differentiability property of the root mean square displacement (11) and momentum (9), thereby justifying the existence of the concept of trajectory of a Brownian particle as well as leading the Langevin equation (1) to be interpreted as a genuine differential equation and not as an integral equation. To accomplish this goal, our article takes up two physical situations described by generalized Fokker–Planck equations. First, the case of a free Brownian particle in the *presence* of inertial force described by a non-Markovian Rayleigh equation is examined in Section 2, whereas in Section 3 we treat the case of a free Brownian particle in the *absence* of inertial force described by a non-Markovian Smoluchowski equation. Concluding remarks are presented in Section 4. In addition, five appendices are attached.

2. Free Brownian motion in the presence of inertial force

Let us begin with the inertial Brownian motion of a free particle immersed in a generic environment described by the Ornstein–Uhlenbeck stochastic process (4) that gives rise to the generalized Fokker–Planck equation in momentum space (see Appendices A and B)

$$\frac{\partial \mathcal{F}(p, t)}{\partial t} = \gamma \frac{\partial}{\partial p} [p \mathcal{F}(p, t)] + \mathcal{D}_{pp}(t) \frac{\partial^2 \mathcal{F}(p, t)}{\partial p^2} \tag{13}$$

for the probability distribution function $\mathcal{F}(p, t)$ in terms of the effective momentum

$$p = p' - \frac{b}{\gamma} \langle \Psi(t) \rangle, \tag{14}$$

where the average of $\Psi(t)$ reads

$$\langle \Psi(t) \rangle = \lim_{\varepsilon \rightarrow 0} \frac{1}{\varepsilon} \int_t^{t+\varepsilon} \int_{-\infty}^{\infty} \psi \mathcal{F}_{\Psi}(\psi, t) d\psi dt. \tag{15}$$

In Eq. (13), $\mathcal{D}_{pp}(t)$ denotes the diffusion coefficient in momentum space

$$\mathcal{D}_{pp}(t) = \gamma \varepsilon(t) \tag{16}$$

expressed in terms of the frictional constant γ and the time-dependent function $\varepsilon(t)$ given by

$$\varepsilon(t) \equiv \frac{b^2}{2\gamma m} I(t) = \frac{b^2}{2\gamma m} \lim_{\varepsilon \rightarrow 0} \frac{1}{\varepsilon} \iint_t^{t+\varepsilon} \langle \Psi(t') \Psi(t'') \rangle dt' dt'', \tag{17}$$

which in turn has dimensions of energy, i.e., $[\text{mass} \times \text{length}^2 \times \text{time}^{-2}]$, since

$$I(t) \equiv \lim_{\varepsilon \rightarrow 0} \frac{1}{\varepsilon} \iint_t^{t+\varepsilon} \langle \Psi(t') \Psi(t'') \rangle dt' dt'' \tag{18}$$

is a dimensionless function. Hence, we call $\varepsilon(t)$ the diffusion energy responsible for the Brownian motion of the particle in the presence of a generic environment.

An outstanding feature underlying the concept of diffusion energy (17), which fulfills the validity condition

$$0 < \varepsilon(t) < \infty, \tag{19}$$

is that it bears together both fluctuation and dissipation phenomena through the parameter b and the friction constant γ , respectively. We can then state that Eq. (17) sets up a general fluctuation–dissipation relationship underlying all open systems described by the Langevin equation (4) and its corresponding Fokker–Planck equation (13). Both cases $\mathcal{E}(t) = 0$ and $\mathcal{E}(t) = \infty$ are not concerned, for they may violate the validity condition of the fluctuation–dissipation relation (19): the former case may lead to dissipation without fluctuation, whilst the latter one may give rise to fluctuation without dissipation.

From the physical point of view, the pivotal issue inherent in theory of Brownian motion is to determine some transport coefficients, such as the time-dependent diffusion coefficient (16). At $t = 0$ the answer to this question seems to be fairly straightforward since the diffusion energy is null, i.e., $\mathcal{E}(0) = \gamma \mathcal{D}(0) = 0$, meaning that there is no diffusive motion, $\mathcal{D}(0) = 0$, associated with the initial condition $\mathcal{F}(p, t = 0) = \delta(p)$ to the generalized Fokker–Planck equation (13). On the other hand, if we assume that in the long-time regime the steady diffusion energy could be identified with the thermal energy, $k_B T$, at thermodynamic equilibrium, i.e.,

$$\lim_{t \rightarrow \infty} \mathcal{E}(t) \approx \mathcal{E}(\infty) = \frac{b^2}{2\gamma m} = k_B T, \quad (20)$$

with the dimensionless function (18) having the asymptotic behavior

$$\lim_{t \rightarrow \infty} I(t) \approx 1, \quad (21)$$

then the diffusion constant (16) in thermal equilibrium is evaluated as $\mathcal{D}_{pp}(\infty) = \gamma m k_B T$. Furthermore, from Eq. (20) it follows that the steady dissipation–fluctuation relationship for thermal systems reads

$$b = \sqrt{2\gamma m k_B T}. \quad (22)$$

The physical significance underlying the condition (21) has to do with the fact that environmental fluctuations become Markovian correlations in the long-time limit, i.e., $I(t)$ displays a local (short) range behavior in the steady regime. By contrast, non-Markovian effects show up in the non-equilibrium range $0 < t < \infty$.

So, for thermal open systems the generalized Fokker–Planck equation (13) for the thermal momentum $p = p' - \sqrt{\frac{2mk_B T}{\gamma}} \langle \Psi(t) \rangle$ reads

$$\frac{\partial \mathcal{F}(p, t)}{\partial t} = \gamma \frac{\partial}{\partial p} [p \mathcal{F}(p, t)] + \gamma m k_B T I(t) \frac{\partial^2 \mathcal{F}(p, t)}{\partial p^2} \quad (23)$$

reducing to the Markovian Rayleigh equation (8) as far as $\langle \Psi(t) \rangle = 0$ and $I(t) = 1$ are concerned. For this reason, we dub (23) the non-Markovian Rayleigh equation. (An alternative manner of deriving (23) is presented in Appendix C.)

It is worth pointing out that the non-Markovian Rayleigh equation (23) has been derived without specifying *ab initio* the form of the autocorrelation function of the Langevin force

$$\langle L(t') L(t'') \rangle = 2\mathcal{D}_{pp}(\infty) \langle \Psi(t') \Psi(t'') \rangle = 2\gamma m k_B T \langle \Psi(t') \Psi(t'') \rangle,$$

as well as its average value, i.e.,

$$\langle L(t) \rangle = \sqrt{2\mathcal{D}_{pp}(\infty)} \langle \Psi(t) \rangle = \sqrt{2\gamma m k_B T} \langle \Psi(t) \rangle.$$

The crucial point is the long-time behavior given by Eqs. (20) and (21). This means that the arbitrariness as the form of the statistical properties of the Langevin force $L(t)$ leaves room to incorporate both non-Markovian and averaging effects into the study of Brownian motion through the functions $I(t)$ and $\langle \Psi(t) \rangle$ in the non-Markovian Rayleigh equation (23). In fact, upon considering the correlational function $I(t)$ in (23) as $I(t) = 1 - e^{-t/t_c}$ (see Appendix D), where the correlation time t_c accounts for non-Markovian effects on the Brownian particle, and by starting from the initial condition $\mathcal{F}(p, t = 0) = \delta(p)$, a non-equilibrium solution to (23) reads

$$\mathcal{F}(p, t) = \frac{1}{\sqrt{4\pi g(t)}} e^{-\frac{p^2}{4g(t)}}, \quad (24)$$

where

$$g(t) = \frac{mk_B T}{2} \left[1 - e^{-\frac{t}{t_r}} + \frac{t_c}{t_c - t_r} \left(e^{-\frac{t}{t_r}} - e^{-\frac{t}{t_c}} \right) \right]. \quad (25)$$

The probability distribution function (24) is expressed in terms of the evolution time t , the relaxation time $t_r = (2\gamma)^{-1}$ as well as the correlation time t_c . From the non-equilibrium solution (24) in the steady regime, we can derive the Maxwell–Boltzmann probability distribution function at thermal equilibrium

$$\mathcal{F}(p) = \frac{1}{\sqrt{2\pi m k_B T}} e^{-\frac{p^2}{2m k_B T}} \quad (26)$$

for the effective momentum: $p = p' - \sqrt{2m k_B T / \gamma} \langle \Psi(\infty) \rangle$. Thus, both relaxation and non-Markovian features showing up in (25) are non-equilibrium effects.

Solution (24) gives rise to $\langle P \rangle = 0$ and the variance

$$\langle P^2 \rangle = 2g(t) = mk_B T \left[1 - e^{-\frac{t}{t_r}} + \frac{t_c}{t_c - t_r} \left(e^{-\frac{t}{t_r}} - e^{-\frac{t}{t_c}} \right) \right]. \tag{27}$$

Making use of Eq. (27) the mean mechanical energy associated with the free Brownian particle is given by

$$\langle E(t) \rangle = \frac{\langle P^2 \rangle}{2m} = \frac{k_B T}{2} \left[1 - e^{-\frac{t}{t_r}} + \frac{t_c}{t_c - t_r} \left(e^{-\frac{t}{t_r}} - e^{-\frac{t}{t_c}} \right) \right], \tag{28}$$

whereas the root mean square momentum, $\Delta P(t) = \sqrt{\langle P^2 \rangle - \langle P \rangle^2}$, reads

$$\Delta P(t) = \Delta P'(t) = \sqrt{mk_B T \left[1 - e^{-\frac{t}{t_r}} + \frac{t_c}{t_c - t_r} \left(e^{-\frac{t}{t_r}} - e^{-\frac{t}{t_c}} \right) \right]}, \tag{29}$$

meaning that the average value of $\Psi(t)$ in the thermal momentum (14), $p = p' - \sqrt{(2mk_B T/\gamma)} \langle \Psi(t) \rangle$, has no influence on the physically observable quantity (29). Furthermore, it is readily to check that non-Markovian effects account for diminishing the strength of the momentum fluctuation (29).

Another consequence of Eq. (29) is the existence of the concept of thermal force, $F(t) = d\Delta P(t)/dt$, acting on a free Brownian particle in the presence of inertial force. This sort of non-mechanical force reads

$$F(t) = -\gamma \sqrt{mk_B T} B(t) \tag{30}$$

where the dimensionless function $B(t)$ is given by

$$B(t) = \frac{t_r \left(e^{-\frac{t}{t_r}} - e^{-\frac{t}{t_c}} \right)}{\sqrt{(t_c - t_r) \left[t_r \left(e^{-\frac{t}{t_r}} - 1 \right) + t_c \left(1 - e^{-\frac{t}{t_c}} \right) \right]}}, \tag{31}$$

valid for $t_c \neq t_r$. If $B(t) > 0$, then the force (30) is attractive: $F(t) < 0$. On the contrary, if $B(t) < 0$, then (30) is repulsive, i.e., $F(t) > 0$. Moreover, it is worth stressing that the dimensionless factor $B(t)$ is expressed in terms of the following experimentally accessible time scales: the evolution time t , the correlation time t_c , as well as the relaxation time t_r . So, by way of example, taking into account $|B(t)| \sim 1$, $\gamma \sim 10^{12} \text{ s}^{-1}$, $m \sim 10^{-3} \text{ kg}$, $k_B \sim 10^{-23} \text{ m}^2 \text{ kg s}^{-2} \text{ K}^{-1}$, and $T \sim 300 \text{ K}$, we find $|F(t)| \sim 1 \text{ N}$. That is the magnitude of the thermal force (30) exerted by a heat bath at room temperature 27°C on a Brownian free particle of mass 1 g.

Notice that in the stationary limit $t \rightarrow \infty$, i.e., as $t \gg t_c, t_r$, the dimensionless function (31) vanishes whereas at short times, $t \ll t_c, t_r$, it approximates to $B(t) \sim -(2\gamma t_c)^{-1/2}$. In this case, the non-equilibrium thermal force (30) tends approximately to the following constant repulsive force

$$F(t) \sim \sqrt{\frac{\gamma k_B T}{t_c}}. \tag{32}$$

Such results indicate that the non-Markovian thermal force (30) is a sort of non-equilibrium effect different from zero in the time window $0 \leq t < \infty$. Moreover, because Eq. (32) blows up in the Markovian limit $t_c \rightarrow 0$, non-Markovian effects account for the analyticity property of the root mean square momentum (29), thus justifying the existence of the non-equilibrium thermal force (30) for all time t . Accordingly, for physical reasons the Langevin equation (4) underlying the Fokker–Planck equation (23) should be interpreted as a differential equation and not as an integral equation. In brief, the concept of non-Markovian Brownian trajectory undergone by an inertial particle turns up as a well-defined differentiable quantity.

3. Free Brownian motion in the absence of inertial force

Now we start from the Langevin equation (5) with b given by Eq. (22)

$$\frac{dX}{dt} = \sqrt{\frac{2k_B T}{m\gamma}} \Psi(t), \tag{33}$$

which gives rise to the following Fokker–Planck equation at point x' on configuration space (see Appendix E)

$$\frac{\partial f(x', t)}{\partial t} = -\sqrt{\frac{2k_B T}{m\gamma}} \langle \Psi(t) \rangle \frac{\partial f(x', t)}{\partial x'} + \frac{k_B T}{m\gamma} \left(1 - e^{-\frac{t}{t_c}} \right) \frac{\partial^2 f(x', t)}{\partial x'^2}. \tag{34}$$

For $\langle \Psi(t) \rangle = 0$ and $t_c \rightarrow 0$, the generalized Fokker–Planck equation (34) reduces to the Markovian Smoluchowski equation (10) at $x = x'$. For this reason, we dub (34) the non-Markovian Smoluchowski equation.

Starting from the deterministic initial condition $f(x', t = 0) = \delta(x')$, we obtain the following time solution to Eq. (34)

$$f(x, t) = \sqrt{\frac{m\gamma}{4\pi A(t)}} e^{-\frac{m\gamma x^2}{4A(t)}}, \quad (35)$$

in terms of the thermal position

$$x = x' - \sqrt{\frac{2k_B T}{m\gamma}} \int_0^t \langle \Psi(t) \rangle dt, \quad (36)$$

and the function

$$A(t) = k_B T \int_0^t I(t) dt = k_B T \left[t + t_c \left(e^{-\frac{t}{t_c}} - 1 \right) \right]. \quad (37)$$

Solution (35) yields $\langle X \rangle = 0$ as well as the mean square displacement

$$\langle X^2 \rangle = \frac{2k_B T}{m\gamma} \left[t + t_c \left(e^{-\frac{t}{t_c}} - 1 \right) \right], \quad (38)$$

where the stochastic position $X = X(t)$ corresponds to its realization (36). Accordingly, the root mean square displacement, $\Delta X(t) = \sqrt{\langle X^2 \rangle - \langle X \rangle^2}$, reads

$$\Delta X(t) = \Delta X'(t) = \sqrt{\frac{2k_B T}{m\gamma} \left[t - t_c \left(1 - e^{-\frac{t}{t_c}} \right) \right]}. \quad (39)$$

We notice that averaging effects in Eq. (34) are unobservable as they do have no influence on the physically measurable quantity (39), albeit they can bring about a shift in the position (36). Furthermore, the root mean square displacement (39) leads to the non-Markovian thermal velocity

$$\mathcal{V}(t) = \sqrt{\frac{k_B T}{2m\gamma}} \frac{\left(1 - e^{-\frac{t}{t_c}} \right)}{\sqrt{t - t_c \left(1 - e^{-\frac{t}{t_c}} \right)}}, \quad (40)$$

which is a non-equilibrium effect since it vanishes in the steady limit, i.e., $\mathcal{V}(\infty) = 0$. On the other hand, at short times Eq. (40) reduces to

$$\mathcal{V}(t) = \sqrt{\frac{k_B T}{m\gamma t_c}}, \quad (41)$$

which in turn does diverge in the Markovian limit $t_c \rightarrow 0$. In consequence, non-Markovian effects account for the existence of the concept of thermal velocity (40) of a free Brownian particle in the absence of inertial force for all time. From the mathematical viewpoint, the upshot (41) implies that the Langevin equation (33) should be interpreted as a differential equation and not as an integral one.

Summing up, using our approach to Brownian motion we have come up with a detailed evaluation of non-Markovian effects on the differentiability of the position fluctuation (39), thus justifying the existence of the concept of differentiable path of a non-inertial Brownian particle for all $t \geq 0$.

4. Summary and discussion

Our paper does feature the following novel findings:

(i) For the case of a free Brownian particle in the *presence* of inertial force, the derivation of the generalized Fokker–Planck equation (23) from the Langevin equation (4) reveals that a simple non-Markovian addition to the Fokker–Planck equation by means of a correlational function $I(t)$ may diminish the values of the root mean square momentum (29). Further, in contrast to the Markovian case (9), non-Markovian effects render the quantity (29) differentiable at $t = 0$ (see Eq. (32)), thereby leading to the concept of non-equilibrium thermal force (30). Mathematically, this upshot implies that the corresponding Langevin equation (4) should be interpreted not as an integral equation but as a genuine differential equation.

(ii) For the case of a free Brownian particle in the *absence* of inertial force, the non-Markovian Smoluchowski equation (34) has been derived from the Langevin equation (33). Again, the novelty is the presence of the time-dependent function $I(t)$ in the diffusion coefficient. Accordingly, the root mean square displacement (39) becomes differentiable at $t = 0$ in view of the presence of the correlation time t_c (see Eq. (41)), thus assuring the existence of the concept of non-equilibrium thermal velocity (40) for all time t . As well as diminishing the strength of the root mean square displacement (39), non-Markovian effects allow for interpreting the Langevin equation (33) as a differential equation and not as an integral one.

Summing up, the differentiability property of both the root mean square momentum (29) and the root mean square displacement (39) for all times imply that the Langevin equation (1) should be interpreted as a differential equation. We

thus eschew some inconsistencies related to such physically measurable quantities in the Markovian approximation as well as the conundrum existing among different rules for interpreting a stochastic differential equation: Stratonovich, Doob, and Itô interpretations. Therefore, our article upholds the view that the trajectory of a Brownian particle actually does exist as a physical concept being mathematically well defined by differentiable functions in the non-Markovian regime. By contrast, Markovian Brownian motion does not exist at all in the physical world, albeit non-differentiable paths make up a prolific abstraction in the mathematical realm.

By way of discussion, we would like to clarify some points concerning the differences between the present study and van Kampen's, Adelman's, and Mori's approaches [5,31].

In his authoritative monograph [5] van Kampen states: “*The Fokker–Planck equation is a special type of master equation, which is often used as an approximation to the actual equation or as a model for more general Markov process*”. Later, he called attention to the pivotal relevance of non-Markovian stochastic processes in nature: “*Non-Markov is the rule, Markov is the exception*” [14]. Yet, he had provided us no clue in this direction. Indeed, such van Kampen's statements can raise doubts concerning to the correctness of dealing with non-Markovian phenomena under the framework of Fokker–Planck equations. By contrast, our present article can shed some light on the non-Markovian character of Brownian motion within a structure of generalized Fokker–Planck equations.

On the other hand, on the basis of the memory Mori–Langevin equation (12) and assuming a Gaussian solution Adelman [31] derived Fokker–Planck equations in which the friction coefficient relies on time (see his Eq. (2.17) in Ref. [31], for instance). Moreover, he claimed that his Fokker–Planck equation is an *exact* one. Yet this statement is untrue. His Gaussian solution is in fact an approximation hypothesis! By contrast, in our approach the friction γ appears as a time-independent quantity. In addition, we have shown that non-Markovian features show up in the non-steady regime, whereas Markov property arises at long time alone.

Lastly, it is worth stressing that our upshots (29) and (39) have been reached without making use of the generalized Langevin equation (12). In contrast to Mori's approach, our present study has predicted that *memory-independent non-Markovian effects* can be physically gauged for a free Brownian particle immersed in a heat bath by measuring its root mean square displacement in the absence of inertial force as well as its root mean square momentum in the presence of inertial force. Surprisingly, this feature coming out in the Einstein–Langevin framework seems to have been overlooked in the centennial literature on Brownian motion [1–46], as well as in graduate student books on Brownian motion while treated as a topic in statistical mechanics (see Refs. [47,48], for instance).

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Appendix A. Kolmogorov equations

In this Appendix we wish to show how a given stochastic differential equation gives rise to a Kolmogorov equation which in turn reduces to a Fokker–Planck equation in the Gaussian approximation. First, we take up the case of one random variable and then the case of two variables.

We suppose the dynamics of the stochastic process $\Phi = \Phi(t)$ to be governed by the ordinary differential equation

$$\frac{d\Phi(t)}{dt} = \mathcal{K}(\Phi, t). \quad (\text{A.1})$$

To find out the time evolution of the probability distribution function $\mathcal{F}(\varphi, t)$ expressed in terms of the realizations φ of the random variable Φ , we closely follow Stratonovich's procedure [3,42–44]. We resort to the definition of conditional probability density given by

$$W(\varphi', t'|\varphi, t) = \frac{f(\varphi', t'; \varphi, t)}{\mathcal{F}(\varphi, t)}, \quad (\text{A.2})$$

where $f(\varphi, t; \varphi', t')$ is the joint probability density function of Φ at different times t and t' with $\varphi' \equiv \varphi(t')$ and $\varphi \equiv \varphi(t)$.

From (A.2), we arrive at the Einstein identity [17]

$$\mathcal{F}(\varphi', t') = \int_{-\infty}^{\infty} W(\varphi', t'|\varphi, t) \mathcal{F}(\varphi, t) d\varphi, \quad (\text{A.3})$$

after using the Kolmogorov compatibility condition [42]

$$\int_{-\infty}^{\infty} f(\varphi', t'; \varphi, t) d\varphi = \mathcal{F}(\varphi', t'). \quad (\text{A.4})$$

The characteristic function for the increment $\Delta\Phi \equiv \Phi(t') - \Phi(t)$ is expressed in terms of the conditional probability density (A.2) as

$$\langle e^{iu\Delta\Phi} \rangle = \int_{-\infty}^{\infty} e^{iu\Delta\varphi} W(\varphi', t'|\varphi, t) d\varphi, \quad (\text{A.5})$$

the inverse of which is

$$W(\varphi', t'|\varphi, t) = \frac{1}{2\pi} \int_{-\infty}^{\infty} e^{-iu\Delta\varphi} \langle e^{iu\Delta\Phi} \rangle du. \quad (\text{A.6})$$

Now, using the expansion

$$\langle e^{iu\Delta\Phi} \rangle = \sum_{s=0}^{\infty} \frac{(iu)^s}{s!} \langle (\Delta\Phi)^s \rangle, \quad (\text{A.7})$$

Eq. (A.6) turns out to be expressed as

$$W(\varphi', t'|\varphi, t) = \sum_{s=0}^{\infty} \frac{(-1)^s}{s!} \langle (\Delta\Phi)^s \rangle \frac{\partial^s}{\partial \varphi^s} \delta(\varphi - \varphi'). \quad (\text{A.8})$$

Inserting (A.8) into (A.3) and dividing the resulting equation by ε , we arrive at

$$\frac{\mathcal{F}(\varphi', t') - \mathcal{F}(\varphi', t)}{\varepsilon} = \sum_{s=1}^{\infty} \frac{(-1)^s}{s!} \frac{\partial^s}{\partial \varphi^s} \left\{ \frac{\langle (\Delta\Phi)^s \rangle}{\varepsilon} \mathcal{F}(\varphi, t) \right\}, \quad (\text{A.9})$$

with $t' = t + \varepsilon$ and $\varphi' \equiv \varphi(t + \varepsilon)$. The procedure of taking the limit $\varepsilon \rightarrow 0$ in both sides of (A.9) leads to the Kolmogorov equation

$$\frac{\partial \mathcal{F}(\varphi, t)}{\partial t} = \mathbb{K} \mathcal{F}(\varphi, t), \quad (\text{A.10})$$

where the Kolmogorovian operator \mathbb{K} acts on the probability distribution function $\mathcal{F}(\varphi, t)$ according to

$$\mathbb{K} \mathcal{F}(\varphi, t) = \sum_{s=1}^{\infty} \frac{(-1)^s}{s!} \frac{\partial^s}{\partial \varphi^s} [B^{(s)}(\varphi, t) \mathcal{F}(\varphi, t)], \quad (\text{A.11})$$

with the coefficients $B^{(s)}(\varphi, t)$, given by

$$B^{(s)}(\varphi, t) = \lim_{\varepsilon \rightarrow 0} \frac{\langle (\Delta\Phi)^s \rangle}{\varepsilon}, \quad (\text{A.12})$$

calculated from the stochastic differential equation (A.1) in the following integral form

$$\Delta\Phi \equiv \Phi(t + \varepsilon) - \Phi(t) = \int_t^{t+\varepsilon} \mathcal{K}(\Phi, t') dt', \quad (\text{A.13})$$

after averaging $\langle (\Delta\Phi)^s \rangle$ over a given conditional probability density $W(\varphi', t'|\varphi, t)$ according to Eq. (A.5).

Summing up, we have set out a general scheme for deriving from the stochastic differential equation (A.1) the Kolmogorov equation (A.10) which reckons with non-Gaussian features on account of the presence of the s th moment of the increment $\Delta\Phi$, i.e., $\langle (\Delta\Phi)^s \rangle$.

According to Pawula's theorem [44] there exists no non-Gaussian approximation to the non-Gaussian Kolmogorov equation (A.10) in compliance with the positivity of $\mathcal{F}(\varphi, t)$. Hence, in the Gaussian approximation Eq. (A.10) reads

$$\frac{\partial \mathcal{F}_\Phi(\varphi, t)}{\partial t} = -\frac{\partial}{\partial \varphi} [B^{(1)}(\varphi, t) \mathcal{F}_\Phi(\varphi, t)] + \frac{1}{2} \frac{\partial^2}{\partial \varphi^2} [B^{(2)}(\varphi, t) \mathcal{F}_\Phi(\varphi, t)]. \quad (\text{A.14})$$

In the physics literature, the class of Gaussian stochastic differential equation (A.1) in the Gaussian approximation is known as Langevin equation (Eqs. (4) and (5), for instance), whereas the corresponding Gaussian Kolmogorov equation (A.14) is dubbed Fokker–Planck equation in configuration space.

For the case of two random variables Π and Φ , the set of stochastic differential equations, given by

$$\frac{d\Pi(t)}{dt} = \mathcal{K}_1(\Pi, \Phi, t), \quad (\text{A.15})$$

$$\frac{d\Phi(t)}{dt} = \mathcal{K}_2(\Pi, \Phi, t), \quad (\text{A.16})$$

generates the following phase space Kolmogorov equation for the joint probability distribution $\mathcal{F}_{\Pi\Phi}(\pi, \varphi, t)$

$$\frac{\partial \mathcal{F}_{\Pi\Phi}(\pi, \varphi, t)}{\partial t} = \sum_{s=1}^{\infty} \sum_{r=0}^s \frac{(-1)^s}{r!(s-r)!} \frac{\partial^s}{\partial \pi^{s-r} \partial \varphi^r} [B^{(s-r,r)}(\pi, \varphi, t) \mathcal{F}_{\Pi\Phi}(\pi, \varphi, t)], \tag{A.17}$$

with

$$B^{(s-r,r)}(\pi, \varphi, t) = \lim_{\varepsilon \rightarrow 0} \left[\frac{\langle (\Delta \Pi)^{s-r} (\Delta \Phi)^r \rangle}{\varepsilon} \right]. \tag{A.18}$$

The increments $\Delta \Pi$ and $\Delta \Phi$ are evaluated from (A.15) and (A.16) in the integral form

$$\Delta \Phi \equiv \Phi(t + \varepsilon) - \Phi(t) = \int_t^{t+\varepsilon} \mathcal{K}_1(\Pi, \Phi, t) dt \tag{A.19}$$

and

$$\Delta \Pi \equiv \Pi(t + \varepsilon) - \Pi(t) = \int_t^{t+\varepsilon} \mathcal{K}_2(\Pi, \Phi, t) dt. \tag{A.20}$$

In the Gaussian approximation the phase space Kolmogorov equation (A.17) reduces to the following Fokker–Planck equation in phase space

$$\frac{\partial \mathcal{F}_{\Pi\Phi}(\pi, \varphi, t)}{\partial t} = \sum_{s=1}^2 \sum_{r=0}^s \frac{(-1)^s}{r!(s-r)!} \frac{\partial^s}{\partial \pi^{s-r} \partial \varphi^r} [B^{(s-r,r)}(\pi, \varphi, t) \mathcal{F}_{\Pi\Phi}(\pi, \varphi, t)]. \tag{A.21}$$

The Langevin equations (C.1) and (C.2) in phase space (see Appendix C) are physical examples of (A.15) and (A.16) while the non-Markovian Klein–Kramers equation (C.15) is a special case of phase space Fokker–Planck equation (A.21).

In Appendices B and E we evaluate explicitly the coefficients $B^{(s)}(\varphi, t)$ in (A.14) for the cases of the non-Markovian Rayleigh equation and the non-Markovian Smoluchowski equation, respectively. Appendix C in turn yields the coefficients $B^{(s-r,r)}(\pi, \varphi, t)$ concerning the non-Markovian Klein–Kramers equation.

Appendix B. The non-Markovian Rayleigh equation

The stochastic differential equation (A.1) for $\Phi(t) \equiv P(t)$ and $\mathcal{K}(P, t) \equiv -\gamma P + b\Psi(t)$ becomes the Uhlenbeck–Ornstein process (4) that generates the Gaussian Kolmogorov equation (A.14) for $\mathcal{F}(\varphi, t) \equiv f(p', t)$, i.e.,

$$\frac{\partial f(p', t)}{\partial t} = -\frac{\partial}{\partial p'} [A_1(p', t)f(p', t)] + \frac{1}{2} \frac{\partial^2}{\partial p'^2} [A_2(p', t)f(p', t)], \tag{B.1}$$

where the drift coefficient $B^{(1)}(\varphi, t) \equiv A_1(p', t)$ and the diffusion coefficient $B^{(2)}(\varphi, t) \equiv A_2(p', t)$ are, respectively, given by

$$A_1(p', t) = \lim_{\varepsilon \rightarrow 0} \left[\frac{\langle \Delta P \rangle}{\varepsilon} \right] = -\gamma p' + b \langle \Psi(t) \rangle \tag{B.2}$$

and

$$A_2(p', t) = \lim_{\varepsilon \rightarrow 0} \left[\frac{\langle (\Delta P)^2 \rangle}{\varepsilon} \right] = b^2 \lim_{\varepsilon \rightarrow 0} \frac{1}{\varepsilon} \int_t^{t+\varepsilon} \int_t^{t+\varepsilon} \langle \Psi(t') \Psi(t'') \rangle dt' dt''. \tag{B.3}$$

Our aim is to show how such coefficients (B.2) and (B.3) are built up from the Uhlenbeck–Ornstein process (4) in the integral form

$$\Delta P = P(t + \varepsilon) - P(t) = -\gamma \int_t^{t+\varepsilon} P(t) dt + b \int_t^{t+\varepsilon} \Psi(t) dt. \tag{B.4}$$

First, in order to calculate the drift coefficient $A_1(p', t)$ we average the stochastic increment ΔP over the probability distribution function $\delta(p - p') \mathcal{F}(\psi, t)$, such that

$$\langle P(t) \rangle = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} p \delta(p - p') \mathcal{F}(\psi, t) dp d\psi = p' \tag{B.5}$$

and

$$\langle \Psi(t) \rangle = \int_{-\infty}^{\infty} \psi \mathcal{F}(\psi, t) d\psi. \tag{B.6}$$

Next, we divide the resulting $\langle \Delta P \rangle$ by ε and then take the limit $\varepsilon \rightarrow 0$. Thus, the coefficient (B.2) is obtained after noticing that

$$\lim_{\varepsilon \rightarrow 0} \frac{1}{\varepsilon} \int_t^{t+\varepsilon} \langle \Psi(t) \rangle dt = \langle \Psi(t) \rangle \quad (\text{B.7})$$

and

$$\lim_{\varepsilon \rightarrow 0} \int_t^{t+\varepsilon} \langle \Psi(t) \rangle dt = 0. \quad (\text{B.8})$$

To evaluate the diffusion coefficient (B.3) we start with Eq. (B.4) in the form

$$(\Delta P)^2 = \left[-\gamma \int_t^{t+\varepsilon} P(t) dt + b \int_t^{t+\varepsilon} \Psi(t) dt \right] \left[-\gamma \int_t^{t+\varepsilon} P(t') dt' + b \int_t^{t+\varepsilon} \Psi(t') dt' \right]. \quad (\text{B.9})$$

Then, we average $(\Delta P)^2$ over the distribution $\delta(p - p') \mathcal{F}_\psi(\psi, t)$ and calculate the limiting process $\lim_{\varepsilon \rightarrow 0} [\frac{(\Delta P)^2}{\varepsilon}]$. So we readily arrive at the result (B.3).

By performing the variable change $p = p' - \frac{b}{\gamma} \langle \Psi(t) \rangle$, such that $f(p', t) \mapsto \mathcal{F}(p, t)$, from the Gaussian Kolmogorov equation (B.1) we derive the generalized Fokker–Planck equation (13) or the non-Markovian Rayleigh equation (23).

Appendix C. Another derivation for the non-Markovian Rayleigh equation

The Langevin equation (1) may be written in phase space (X, P) as

$$\frac{dP}{dt} = -\frac{dV(X)}{dX} - \gamma P + b\Psi(t), \quad (\text{C.1})$$

$$\frac{dX}{dt} = \frac{P}{m}, \quad (\text{C.2})$$

which in turn give rise to the Kolmogorov equation in phase space (x, p')

$$\frac{\partial \mathcal{F}(x, p', t)}{\partial t} = \mathbb{K} \mathcal{F}(x, p', t), \quad (\text{C.3})$$

where the Kolmogorovian operator \mathbb{K} acts on the probability distribution function $\mathcal{F}(x, p', t)$ according to

$$\mathbb{K} \mathcal{F}(x, p', t) = \sum_{k=1}^{\infty} \sum_{r=0}^k \frac{(-1)^k}{r!(k-r)!} \frac{\partial^k}{\partial x^{k-r} \partial p'^r} [A^{(k-r,r)}(x, p', t) \mathcal{F}(x, p', t)] \quad (\text{C.4})$$

with

$$A^{(k-r,r)}(x, p', t) = \lim_{\varepsilon \rightarrow 0} \left[\frac{\langle (\Delta X)^{k-r} \rangle \langle (\Delta P)^r \rangle}{\varepsilon} \right]. \quad (\text{C.5})$$

Both the increments $\Delta X \equiv X(t + \varepsilon) - X(t)$ and $\Delta P \equiv P(t + \varepsilon) - P(t)$ in the coefficients (C.5) are calculated from Eqs. (C.1) and (C.2) in the form

$$\Delta X = \frac{1}{m} \int_t^{t+\varepsilon} P(t) dt, \quad (\text{C.6})$$

$$\Delta P = -\frac{dV(X)}{dX} \varepsilon - \gamma \int_t^{t+\varepsilon} P(t) dt + b \int_t^{t+\varepsilon} \Psi(t) dt. \quad (\text{C.7})$$

The average values $\langle (\Delta X)^{k-r} \rangle \langle (\Delta P)^r \rangle$, in turn are to be calculated about the sharp values q and \bar{p} , i.e.,

$$\mathcal{F}_{XP\psi}(x, p', \psi, t) = \delta(x - q) \delta(p' - \bar{p}) \mathcal{F}_\psi(\psi, t). \quad (\text{C.8})$$

The phase space Kolmogorov equation (C.3) describes the time evolution of a Brownian particle immersed in a general non-Gaussian environment. Yet in the Gaussian approximation Eq. (C.3) changes into the generalized Fokker–Planck equation

$$\frac{\partial \mathcal{F}}{\partial t} = -\frac{\partial}{\partial x} [A^{(1,0)} \mathcal{F}] - \frac{\partial}{\partial p} [A^{(0,1)} \mathcal{F}] + \frac{1}{2} \frac{\partial^2}{\partial x^2} [A^{(2,0)} \mathcal{F}] + \frac{\partial^2}{\partial x \partial p'} [A^{(1,1)} \mathcal{F}] + \frac{1}{2} \frac{\partial^2}{\partial p'^2} [A^{(0,2)} \mathcal{F}], \quad (\text{C.9})$$

whereby $\mathcal{F} \equiv \mathcal{F}(x, p', t)$. The drift coefficients are given by

$$A^{(1,0)}(x, p', t) = \lim_{\varepsilon \rightarrow 0} \left[\frac{\langle \Delta X \rangle}{\varepsilon} \right] = \frac{p}{m}, \tag{C.10}$$

and

$$A^{(0,1)}(x, p', t) = \lim_{\varepsilon \rightarrow 0} \left[\frac{\langle \Delta P \rangle}{\varepsilon} \right] = -\gamma p' - \frac{dV(x)}{dx} + b\langle \Psi(t) \rangle, \tag{C.11}$$

whereas the diffusion coefficients are

$$A^{(2,0)}(x, p', t) = \lim_{\varepsilon \rightarrow 0} \left[\frac{\langle (\Delta X)^2 \rangle}{\varepsilon} \right] = 0, \tag{C.12}$$

$$A^{(1,1)}(x, p', t) = \lim_{\varepsilon \rightarrow 0} \left[\frac{\langle \Delta X \rangle \langle \Delta P \rangle}{\varepsilon} \right] = 0, \tag{C.13}$$

$$A^{(0,2)}(x, p', t) = \lim_{\varepsilon \rightarrow 0} \left[\frac{\langle (\Delta P)^2 \rangle}{\varepsilon} \right] = b^2 I(t). \tag{C.14}$$

Taking into account Eqs. (C.10)–(C.14), the Fokker–Planck equation (C.9) turns out to be written as

$$\frac{\partial \mathcal{F}}{\partial t} = -\frac{p'}{m} \frac{\partial}{\partial x} \mathcal{F} + \frac{\partial}{\partial p'} \left[\frac{dV(x)}{dx} + \gamma p' - b\langle \Psi(t) \rangle \right] \mathcal{F} + \frac{b^2}{2} I(t) \frac{\partial^2}{\partial p'^2} \mathcal{F}, \tag{C.15}$$

with $\langle \Psi(t) \rangle$ and $I(t)$ given by Eqs. (15) and (17), respectively.

As far as $\langle \Psi(t) \rangle = 0$, $I(t) = 1$, and Eq. (22) are concerned, our non-thermal Fokker–Planck equation (C.15) reduces to the Markovian Klein–Kramers equation at $p' = p$ [45,46]

$$\frac{\partial \mathcal{F}}{\partial t} = -\frac{p'}{m} \frac{\partial}{\partial x} \mathcal{F} + \frac{\partial}{\partial p'} \left[\frac{dV(x)}{dx} + \gamma p' \right] \mathcal{F} + \gamma m k_B T \frac{\partial^2}{\partial p'^2} \mathcal{F}. \tag{C.16}$$

Hence, we dub Eq. (C.15) the non-Markovian Klein–Kramers equation.

For $V(x) = 0$, the non-Markovian Klein–Kramers equation (C.15) reads

$$\frac{\partial \mathcal{F}}{\partial t} = -\frac{p'}{m} \frac{\partial}{\partial x} \mathcal{F} + \gamma \frac{\partial}{\partial p'} \left[p' - \sqrt{\frac{2mk_B T}{\gamma}} \langle \Psi(t) \rangle \right] \mathcal{F} + \gamma m \varepsilon(t) \frac{\partial^2}{\partial p'^2} \mathcal{F}. \tag{C.17}$$

Then, upon performing the variable change given by

$$p = p' - \sqrt{\frac{2mk_B T}{\gamma}} \langle \Psi(t) \rangle \tag{C.18}$$

so that $\mathcal{F}(x, p', t) \mapsto \mathcal{F}(x, p, t)$, and taking $\mathcal{F}(p, t) = \int_{-\infty}^{\infty} \mathcal{F}(x, p, t) dx$, Eq. (C.17) turns out to be rewritten as the non-Markovian Rayleigh equation (23).

Upon employing Kramers' technique [46], it is also possible to derive the non-Markovian Smoluchowski equation (34) directly from the non-Markovian Klein–Kramers equation (C.17).

Appendix D. The correlational function

Both generalized Fokker–Planck equations (23) and (34) present the correlational function

$$I(t) = \lim_{\varepsilon \rightarrow 0} \frac{1}{\varepsilon} \iint_t^{t+\varepsilon} \langle \Psi(t') \Psi(t'') \rangle dt' dt''. \tag{D.1}$$

On the condition that $I(\infty) = 1$, the autocorrelation function $\langle \Psi(t') \Psi(t'') \rangle$ can be built up as

$$\langle \Psi(t') \Psi(t'') \rangle = \left(1 - e^{-\frac{(t'+t'')}{2t_c}} \right) \delta(t' - t''), \tag{D.2}$$

where t_c is the correlation time of $\Psi(t)$ at times t' and t'' . Then, it follows that

$$I(t) = 1 - e^{-\frac{t}{t_c}}, \tag{D.3}$$

reducing to $I(t) = 1$ in the Markovian regime, $t_c \rightarrow 0$.

The autocorrelation function (D.2) defines a sort of nonwhite noise which in the Markovian limit, $t_c \rightarrow 0$, changes into the so-called white noise

$$\langle \Psi(t')\Psi(t'') \rangle = \delta(t' - t''), \quad (\text{D.4})$$

meaning that the stochastic function $\Psi(t)$ is delta-correlated.

It has been argued that the Markov property (D.4) is a highly idealized feature [3,6,13,14,19] as the physical interaction between the Brownian particle and the thermal bath actually takes places for a finite correlation time $t_c \neq 0$. Hence, our autocorrelation function (D.2) seems to be a more realistic feature underlying the statistical behavior of the Langevin stochastic force.

In addition, it is worth highlighting that the functional form of $\langle \Psi(t')\Psi(t'') \rangle$ is not experimentally determined. Analytically, the functional form $I(t) = 1 - e^{-t/t_c}$ is suitable to solving the non-Markovian Rayleigh equation (23) and examining non-Markovian effects on the physically measurable quantity (29), for instance.

Appendix E. The non-Markovian Smoluchowski equation

Eq. (A.1) for $\Phi(t) \equiv X(t)$ and $\mathcal{K}(X, t) \equiv \sqrt{\frac{2k_B T}{m\gamma}} \Psi(t)$ changes to Eq. (33) that in turn gives rise to the following Gaussian Kolmogorov equation (A.14) in position space $\mathcal{F}(\varphi, t) \equiv g(x', t)$

$$\frac{\partial g(x', t)}{\partial t} = -\frac{\partial}{\partial x'} [B^{(1)}(x', t)g(x', t)] + \frac{1}{2} \frac{\partial^2}{\partial x'^2} [B^{(2)}(x', t)g(x', t)]. \quad (\text{E.1})$$

Both the drift coefficient $B^{(1)}(x', t)$ and the diffusion coefficient $B^{(2)}(x', t)$ are built up as follows. First, we start from the stochastic differential equation (33) in the integral form

$$\Delta X \equiv X(t + \varepsilon) - X(t) = \sqrt{\frac{2k_B T}{m\gamma}} \int_t^{t+\varepsilon} \Psi(t) dt \quad (\text{E.2})$$

that in turn leads to

$$(\Delta X)^2 = \frac{2k_B T}{m\gamma} \left[\int_t^{t+\varepsilon} \Psi(t) dt \right] \left[\int_t^{t+\varepsilon} \Psi(t') dt' \right] = \frac{2k_B T}{m\gamma} \iint_t^{t+\varepsilon} \Psi(t)\Psi(t') dt dt'. \quad (\text{E.3})$$

Next, we average both ΔX and $(\Delta X)^2$ over $\delta(x - x')\mathcal{F}(\psi, t)$, i.e.,

$$\langle \Delta X \rangle = \sqrt{\frac{2k_B T}{m\gamma}} \int_t^{t+\varepsilon} \langle \Psi(t) \rangle dt, \quad (\text{E.4})$$

$$\langle (\Delta X)^2 \rangle = \frac{2k_B T}{m\gamma} \iint_t^{t+\varepsilon} \langle \Psi(t)\Psi(t') \rangle dt dt'. \quad (\text{E.5})$$

Thereby, making use of the result $\lim_{\varepsilon \rightarrow 0} \frac{1}{\varepsilon} \int_t^{t+\varepsilon} \langle \Psi(t) \rangle dt = \langle \Psi(t) \rangle$ the drift coefficient reads

$$B^{(1)}(x', t) \equiv \lim_{\varepsilon \rightarrow 0} \left[\frac{\langle \Delta X \rangle}{\varepsilon} \right] = \sqrt{\frac{2k_B T}{m\gamma}} \langle \Psi(t) \rangle \quad (\text{E.6})$$

whereas the diffusion coefficient becomes

$$B^{(2)}(x', t) \equiv \lim_{\varepsilon \rightarrow 0} \left[\frac{\langle (\Delta X)^2 \rangle}{\varepsilon} \right] = \frac{2k_B T}{m\gamma} \lim_{\varepsilon \rightarrow 0} \frac{1}{\varepsilon} \iint_t^{t+\varepsilon} \langle \Psi(t)\Psi(t') \rangle dt dt'. \quad (\text{E.7})$$

Inserting (E.6) and (E.7) into the equation of motion (E.1) and reckoning with (D.3) yields to the non-Markovian Smoluchowski equation (34).

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