Stochastic hydrodynamic velocity field and the representation of Langevin equations

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Abstract

The fluctuation-dissipation theorem, in the Kubo original formulation, is based on the decomposition of the thermal agitation forces into a dissipative contribution and a stochastically fluctuating term. This decomposition can be avoided by introducing a stochastic velocity field, with correlation properties deriving from linear response theory. Here, we adopt this field as the comprehensive hydrodynamic/fluctuational driver of the kinematic equations of motion. With this description, we show that the Langevin equations for a Brownian particle interacting with a solvent fluid become particularly simple and can be applied even in those cases in which the classical approach, based on the concept of a stochastic thermal force, displays intrinsic difficulties e.g., in the presence of the Basset force. We show that a convenient way for describing hydrodynamic/thermal fluctuations is by expressing them in the form of Extended Poisson-Kac Processes possessing prescribed correlation properties and a continuous velocity density function. We further highlight the importance of higher-order correlation functions in the description of the stochastic hydrodynamic velocity field with special reference to short-time properties of Brownian motion. We conclude by outlining some practical implications in connection with the statistical description of particle motion in confined geometries.

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I. INTRODUCTION

One of the major results of statistical physics for systems at thermal equilibrium is the fluctuation-dissipation theorem, formulated by Einstein in 1905 to 1906 in the analysis of Brownian motion [1], generalized by Callen and Welton using a quantum formalism [2] and extended by Kubo to generalized Langevin processes [3]. In its essence, the fluctuation-dissipation theorem is based on the decomposition of the thermal fluctuations into two main contributions: a dissipative force proportional to velocity that, in the Einstein-Langevin theory, coincides with the Stokesian friction [1, 4], and a stochastic force accounting for the thermal fluctuations. The same setting characterizes the Kubo approach that involves a generalized Langevin equation of the form

$$\dot{v}(t) = -\int_0^t h(t-\tau)\, v(\tau)\, d\tau + \frac{R(t)}{m} \tag{1}$$

for a particle of mass m and velocity $v(t)$ in a still liquid at thermal equilibrium, where $\dot{v}(t) = dv(t)/dt$. In eq. (1), $h(t)$ is the dissipative response kernel and $R(t)$ the stochastic fluctuating contribution. Starting from this formulation, Kubo generalized Einstein's result, connecting the velocity autocorrelation function $\langle v(t)v(0)\rangle$ and the autocorrelation function of the stochastic forcing term $\langle R(t)R(0)\rangle$, $t \geq 0$, to the dissipative response kernel $h(t)$, namely

$$\frac{1}{\mathrm{i}\omega + h[\omega]} = \frac{1}{k_B\, T} \int_0^\infty \langle v(t)v(0)\rangle\, e^{-\mathrm{i}\omega t}\, dt \;, \tag{2}$$

$\mathrm{i} = \sqrt{-1}$, and

$$\langle R(t)R(0)\rangle = m\, k_B\, T\, h(t) \;, \tag{3}$$

where T is the temperature, k_B the Boltzmann constant and $h[\omega]$ the Fourier-Laplace transform of $h(t)$, i.e., the Laplace transform of $h(t)$ when the Laplace variable s is set equal to $\mathrm{i}\omega$. Equation (3) is valid if $h(t)$ is not impulsive. Equation (2) is customarily referred to as the first Fluctuation-Dissipation (FD) theorem, while eq. (3) holds for the second FD theorem [5].

As observed by Kubo et al. [5]: "... the random force appearing in the fluctuation-dissipation theorem is not simple, because the separation of the force into frictional and random forces is itself a complex problem of statistical physics." This observation indicates that some level of arbitrariness resides in this decomposition [6]. A similar remark is also stressed by Tothova and Lisy [7], who correctly observe that a "physical" thermal force, namely $R(t)$ in eq. (1), should in principle be measurable independently of the hydrodynamic interactions, while in experiments it is just a byproduct of the reprocessing of particle trajectory time series [8–10].

The problem is even more complex for particles moving in a fluid whenever the fluid inertia is accounted for, e.g., by considering the time-dependent Stokes regime, so that eq. (1) is replaced by [11, 12]

$$\dot{v}(t) = -\int_0^t h(t-\tau)\, v(\tau) d\tau - \int_0^t k(t-\tau)\, \dot{v}(\tau)\, d\tau + \frac{R(t)}{m}\,. \tag{4}$$

This equation is valid assuming $v(0) = 0$, otherwise the additional term $-k(t)\, v(0)$ should be added, as addressed in the remainder (Section III). In eq. (4), the inertial effects appear as a memory integral defined by the kernel $k(t)$ acting on the time derivative of the velocity. This inertial term in an incompressible fluid is expressed by the dynamic added mass, corresponding to the superposition of the impulsive contribution of the added mass m_a and of the Basset force [13]

$$k(t) = m_a\, \delta(t) + \beta\, \frac{1}{\sqrt{t}}\,. \tag{5}$$

For a spherical particle of radius R_p it is $\beta = 6\,\sqrt{\pi\rho\mu}\, R_p^2$, where ρ and μ are the density and the viscosity of the fluid, respectively, and $m_a = m\rho/(2\rho_p)$, where ρ_p is the particle density. Note that in this case, the Kubo formulation of the FD theorem does not strictly apply [5]. Nevertheless, enforcing Linear Response Theory (LRT) it is possible to recover the velocity autocorrelation function, and out of it, the autocorrelation function of the stochastic forcing $R(t)$ [5]. Phenomenologically, the above mentioned inertial effects correspond to the back-action to the particle of the correlated motion of the fluid elements in its neighbourhood [14]. Viewed in this perspective, it is even more difficult to separate conceptually their influence from that of the stochastic forcing $R(t)$.

The importance of the FD theorem is essentially two-fold: (i) to provide a unified framework for fluctuations and dissipation, at least for systems at thermal equilibrium. This has been originally expressed by the Stokes-Einstein relation, connecting the main physical quantity accounting for the intensity of thermal fluctuations, namely the particle diffusivity D, to the strength of the dissipative action, given by the Stokesian friction factor η, and to the thermodynamic state of the system (characterised by the temperature T), $D = k_B T/\eta$; (ii) to make possible direct stochastic simulations of particle trajectories, via the concept of Wiener-Langevin equations (with the caveat of breaking Galilei invariance which, however, can be cured by suitably adapted methods [15]). Thanks to the introduction of Wiener processes, compactly expressing the long-term properties of Brownian fluctuations [16, 17], and to the seminal work by Ito on stochastic analysis and integration [18], this powerful numerical tool was made available, and soon became a cornerstone in the investigation of large-scale molecular systems starting from the analysis of individual trajectories. The possibility of making stochastic simulations of particle motion paved the way to the last 70 years of research and elaborations, not only in classical statistical physics, but also in quantum physics (path-integrals on Wiener trajectories [19]), condensed-matter physics and quantum field theory [20]. Accordingly, new fields of physics have been opened, for instance, the fractal theory for processes and objects controlled by fluctuational dynamics (diffusion-limited processes, random fractals) [21], where numerical simulations played a central role for the development of the theory of disordered and fractal systems.

Back to FD theory, out of the two main conceptual and practical implications pointed out above, the second one is probably the most delicate, as, within this framework, a generalized Stokes-Einstein relation is essentially a very general and elementary property of equilibrium and dissipation (see the discussion in Section III). The goal of this article is to show that this classical decomposition into dissipative and fluctuational forces can be overcome, as a comprehensive description of the hydrodynamic and thermal fluctuations is embedded in the concept of a stochastic hydrodynamic/thermal velocity field $\mathbf{v}_s(t;\mathbf{x})$, introduced in this article. Consequently, the main physical problem is the determination and the representation of the statistical properties of the hydrodynamic fluctuations for generic fluids and flow devices. This approach answers the doubts and reflections of Kubo et al. [5] on the real physical meaning of dissecting the complex interaction of a particle with a fluid continuum into dissipative/inertial/fluctuating contributions. In the unitary and compact description of the stochastic velocity field $\mathbf{v}_s(t;\mathbf{x})$ lies the essence of the indecomposable relation between fluctuation, dissipation and fluid medium inertia.

In this respect, the formulation that we put forward in this paper provides a completely different point of view compared to FD theory, in the sense that in this approach the concept of a stochastic thermal force is redundant. This simplifies, in turn, the application to hydrodynamic problems, involving, e.g., fluid inertial effects. The practical implementation of this approach for the simulation of stochastic particle motion finally implies the reduction of the comprehensive stochastic velocity field into generic stochastic processes. Using

Poisson-Kac processes [23, 24], Generalized Poisson-Kac Processes [25–27] or Lévy Walks [28] is a simple and computionally efficient way of expressing the stochastic velocity field for this purpose. All these processes can be unified into the class of Extended Poisson-Kac processes as recently formulated in Ref. [29].

The article is organized as follows. Section II introduces the velocity splitting for generic stochastic dynamics described by equations of the form of eq. (4) into a "deterministic" velocity term, accounting for the deterministic interactions, and a stochastic velocity field for the hydrodynamical/thermal fluctuating contributions. In Section III, we derive the velocity autocorrelation function of the fluctuating part using LRT. In Section III, we describe the simple problem of a Brownian particle influenced by inertial fluid effects. Section V compares and contrasts the present formulation of the hydrodynamic Langevin equation based on the definition of a stochastic hydrodynamic velocity field with the classical approach based on eq. (4). We also discuss the experimental determination of this field and how this approach leads to a different and alternative interpretation of FD theory. The comprehensive description of hydrodynamic/thermal interactions within a unique stochastic velocity field shifts the modeling focus from the classical Wiener-based description of thermal forces to the use of stochastic processes possessing either exponentially or power-law decaying correlation functions of time, corresponding, in their more general setting, to the class of Extended Poisson-Kac processes analyzed in Ref. [29]. In Section VI, we explore this aspect by explictly addressing how these extended processes can be equipped, in a simple way, with a generic velocity density function, still keeping unchanged their correlation properties. Furthermore, we also show how the present formulation hinges on a more detailed description of the statistical properties of thermal velocity fluctuations, beyond the analysis of second-order correlation functions. Experimental validation of the theoretical predictions may be obtained from the analysis of Brownian motion at short time scales, using the techniques addressed in Refs. [8–10, 30, 31]. We discuss the application of this approach to microfluidic problems in Appendix C. Apart from the practical relevance to microfluidics, this analysis opens interesting theoretical perspectives connected with the extension of LRT to the nonlinear case.

II. DETERMINISTIC-STOCHASTIC VELOCITY SPLITTING

Consider eq. (4) in its more general setting,

$$\dot{\mathbf{v}} = L_d[\mathbf{v};\mathbf{x}] + L_i[\dot{\mathbf{v}};\mathbf{x}] + \mathbf{a}(\mathbf{x}) + \frac{\mathbf{R}(t)}{m} \ , \tag{6}$$

where L_d and L_i are linear functionals of the particle velocity, $\{\mathbf{v}(\tau)\}_{\tau=0}^t$, acceleration, $\{\dot{\mathbf{v}}(\tau)\}_{\tau=0}^t$, and position, $\{\mathbf{x}(\tau)\}_{\tau=0}^t$. These operators are associated with dissipative and inertial effects. The dependence on the position history is relevant for studying particle motion in confined geometries, wherein both frictional and inertial effects depend on the particle position and are described by tensor-valued quantities [32, 33]. In eq. (6), $\mathbf{a}(\mathbf{x})$ is the acceleration deriving either from external potentials or from externally-driven hydrodynamic flows. Equation (6) is completed by the kinematic equation $\dot{\mathbf{x}} = \mathbf{v}$.

We postulate the decomposition of the particle velocity into a term accounting for all deterministic perturbations, $\mathbf{v}_d$, and a term accounting for all stochastic perturbations, $\mathbf{v}_s$. Accordingly, we adopt the notation with suffix "d" denoting deterministic terms and "s" stochastic ones. We remark that this velocity splitting technique has been proposed in Ref. [34] to study the Langevin equation of a particle moving in a tilted potential. We set therefore

$$\mathbf{v} = \mathbf{v}_d + \mathbf{v}_s \ . \tag{7}$$

Substituting this decomposition into eq. (6) and invoking the linearity of the operators, we obtain the following two dynamical evolution equations,

$$\dot{\mathbf{v}}_d = L_d[\mathbf{v}_d;\mathbf{x}] + L_i[\dot{\mathbf{v}}_d;\mathbf{x}] + \mathbf{a}(\mathbf{x}) \tag{8a}$$

$$\dot{\mathbf{v}}_s = L_d[\mathbf{v}_s;\mathbf{x}] + L_i[\dot{\mathbf{v}}_s;\mathbf{x}] + \frac{\mathbf{R}(t)}{m} \tag{8b}$$

The two velocity contributions are coupled via the kinematic equation,

$$\dot{\mathbf{x}}(t) = \mathbf{v}_d(t;\mathbf{x}(t)) + \mathbf{v}_s(t;\mathbf{x}(t)) \ . \tag{9}$$

We note that $\mathbf{v}_d(t;\mathbf{x})$ should be regarded as a "weakly perturbed" stochastic process. Albeit its evolution equation is deterministic, it is coupled to the stochastic term $\mathbf{v}_s(t,\mathbf{x})$ via eq. (9).

Equations (8a), (8b), (9) deserve further discussion and explanation, as they represent the core of this, otherwise elementary, transformation of variables. In the transformed system of variables, the phase-space independent coordinates of the particle are its position vector $\mathbf{x}(t)$ and its "deterministic" velocity component $\mathbf{v}_d(t;\mathbf{x}(t))$. These are stochastic processes dependent on time t. The interpretation of the term $\mathbf{v}_s(t;\mathbf{x})$ is altogether different. In essence, it is a stochastic field of fluctuations providing the overall description of the hydrodynamic/thermal fluctuations exerted by the fluid/molecular environment on the particle. In practice, it is a stochastic process, governed by eqs. (8a),(8b), whose statistical properties are modulated by the position $\mathbf{x}$.

The statistical description of this field involves the characterization of its properties conditional to a fixed value of the position. This approach is identical to the determination of the position-dependent friction tensor in microfluidic channels for fixed $\mathbf{x}$, and the equations of motion follow by superimposing to the so-determined friction forces the influence of external perturbations, while assuming that the latter do not modify the former [32]. The validity of this method relies on the ansatz that the statistical structure of $\mathbf{v}_s(t;\mathbf{x})$ does not depend on the externally forcing deterministic perturbations, gathered in the term $\mathbf{a}(\mathbf{x})$.

Conversely, if both L_d and L_i do not depend on the particle position $\mathbf{x}$, a situation that occurs in free space or in microfluidic channels far away from solid boundaries, $\mathbf{v}_s(t)$ is independent on $\mathbf{v}_d$. In consequence, it is simply prescribed as a stochastic process in time.

III. LINEAR RESPONSE THEORY

In this section, we show how to derive the statistical properties of $\mathbf{v}_s(t;\mathbf{x})$. We consider the statistics of this field conditional to a given value of the position vector $\mathbf{x}$. Henceforth, to simplify the notation, we omit any explicit dependence on $\mathbf{x}$. From LRT [3, 5, 6, 35, 36], the correlation function of $\mathbf{v}_s$ can be derived by considering the linear response of the dynamics to an initial condition $\mathbf{v}_s^0$, averaging it with respect to the equilibrium measure of $\mathbf{v}_s^0$ by assuming independence between $\mathbf{R}(t)$ and $\mathbf{v}_s^0$. As observed by Tothova et al. [37, 38] the original formulation of this approach is due to V. Vladimirsky [39] in a scarsely known paper from 1942 written in Russian. Equation (8b) can thus be rewritten in operatorial form as

$$(I - L_i)\dot{\mathbf{v}}_s(t) = L_d \mathbf{v}_s(t) + \frac{\mathbf{R}(t)}{m} \;, \tag{10}$$

where I is the identity operator, equipped with the initial condition $\mathbf{v}_s(t=0) = \mathbf{v}_s^0$. The solution of eq. (10) is

$$\mathbf{v}_s(t) = e^{(I-L_i)^{-1}L_d t}\mathbf{v}_s^0 + \frac{1}{m} e^{(I-L_i)^{-1}L_d t} * \mathbf{R}(t) \;, \tag{11}$$

where "$*$" denotes the convolution operation. Indicating with $v_{s,h}(t)$ the h-entry of $\mathbf{v}_s(t)$, it follows that $v_{s,h}(t)v_{s,k}(0)$ can be expressed as $v_{s,h}(t)v_{s,k}(0) = \sum_j \left(e^{(I-L_i)^{-1}L_d t}\right)_{h,j} v_{s,j}(0)v_{s,k}(0) + \sum_j \left(e^{(I-L_i)^{-1}L_d t}\right)_{h,j} * R_j(t)v_{s,k}(0)$. If $\langle \ldots \rangle$ holds for the average with respect to the equilibrium probability measure for the velocities and random fluctuations, from the independence of $\mathbf{R}(t)$ and $\mathbf{v}_0$ it follows that $\langle R_j(t)v_{s,k}(0)\rangle = 0$. Moreover,

$$\langle v^0_{s,j} v^0_{s,k}\rangle = \frac{k_B T}{m}\, \delta_{j,k} \; . \tag{12}$$

We remark that added mass effects can be relevant in experimental settings [41]. These can be accounted for in the formalism by adding the added mass term to the particle mass m. In any case, the entries of the velocity autocorrelation tensor can be expressed as

$$\langle v_{s,h}(t)v_{s,k}(0)\rangle = \frac{k_B T}{m} \cdot \left(e^{(I-L_i)^{-1}L_d t}\right)_{h,k} \tag{13}$$

In tensorial form, therefore, the velocity autocorrelation function attains the expression

$$\mathbf{C}^{v_s}(t) = \langle \mathbf{v}_s(t) \otimes \mathbf{v}_s(0)\rangle = \frac{k_B T}{m}\, e^{(I-L_i)^{-1}L_d t} \; . \tag{14}$$

We now restore all the functional dependencies on the position $\mathbf{x}$. From eq. (14), it follows that the entries $C^{v_s}_{h,k}(t\,|\,\mathbf{x})$, $h, k = 1, 2, 3$, of the conditional correlation tensor $\mathbf{C}^{v_s}(t\,|\,\mathbf{x})$ for $\mathbf{v}_s(t;\mathbf{x})$ at a given position $\mathbf{x}$ can be expressed as

$$C^{v_s}_{h,k}(t\,|\,\mathbf{x}) = \frac{k_B T}{m}\, c^{(k)}_h(t\,|\mathbf{x}) \tag{15}$$

where $c^{(k)}_h(t\,|\,\mathbf{x})$, for fixed k, are the entries of the vector-valued function $\mathbf{c}^{(k)}(t\,|\,\mathbf{x})$ satisfying the initial value problem

$$\dot{\mathbf{c}}^{(k)}(t\,|\,\mathbf{x}) = L_d[\mathbf{c}^{(k)}(t\,|\,\mathbf{x});\mathbf{x}] + L_i[\mathbf{c}^{(k)}(t\,|\,\mathbf{x});\mathbf{x}] \tag{16a}$$

$$c^{(k)}_h(0\,|\,\mathbf{x}) = \delta_{h,k} \; . \tag{16b}$$

The solution of eqs. (16) determines the spatio-temporal correlation properties of the stochastic velocity field $\mathbf{v}_s(t;\mathbf{x})$. Equations (16) are analogous to the evolution equation for the correlation function deriving from the original Vladimirsky approach that considers, instead of $c(t\,|\mathbf{x})$, the integral of the correlation function $V(t\,|\,\mathbf{x}) = \langle v^2\rangle \int_0^t c(\tau\,|\,\mathbf{x})d\tau$ (see eq. (6) in [7] and the related discussion).

A final comment concerns generalized Stokes-Einstein FD relations. Consider eq. (4) for a Brownian particle in a still fluid in the scalar approximation, motivated by the isotropy of the problem. In this case $v_s(t) = v(t)$ and $v_d = 0$. Let $\widehat{h}(s)$, $\widehat{k}(s)$ be the Laplace transforms of the dissipative and inertial memory kernels, respectively. These two functions satisfy the following properties: (i) $\widehat{h}(0) = \int_0^\infty h(t)\, dt = \eta_\infty > 0$, where η_∞ is the effective friction factor of the model that we assume it is bounded. (ii) $\lim_{s\to 0} s\widehat{k}(s) = 0$, i.e., there are no dissipative contributions. We also assume that $k(t)$ does not contain any impulsive contribution (i.e., no added mass) corresponding to the condition $\lim_{\varepsilon\to 0} \int_0^\varepsilon k(t)\, dt = 0$. This latter condition can be easily removed, because it does not alter the final result. From eqs. (16), the normalized correlation function $c(t)$ admits a Laplace transform $\widehat{c}(s)$ as a solution of the equation

$$m\, s\, \widehat{c}(s) - m = -\widehat{h}(s)\, \widehat{c}(s) - \widehat{k}(s)\, s\, \widehat{c}(s) \tag{17}$$

and thus

$$\widehat{c}(s) = \frac{m}{m\, s + \widehat{h}(s) + s\, \widehat{k}(s)} \, . \tag{18}$$

The diffusion coefficient D is the time integral of the correlation function $\langle v(t)\, v(0)\rangle = \langle v_s(t)\, v_s(0)\rangle$ and is thus equal to the value $\widehat{c}(s = 0)$, namely

$$D = \int_0^\infty \langle v_s(t) v_s(0)\rangle\, dt = \langle v^2\rangle \int_0^\infty c(t)\, dt = \langle v^2\rangle\, \widehat{c}(0) = \frac{\langle v^2\rangle\, m}{\eta_\infty} \, . \tag{19}$$

Further assuming $\langle v^2\rangle = k_B T/m$, we recover the generalized Stokes-Einstein relation $D = \frac{k_B T}{\eta_\infty}$. In our theory, this follows as a consequence of bounded friction.

IV. A SIMPLE EXAMPLE: BROWNIAN MOTION IN AN INERTIAL FLUID

We consider a Brownian particle in free space subjected to hydrodynamic interactions including fluid inertia. This example is not only interesting in itself, but is also instructive to clarify a common source of misunderstanding [12]. From time-dependent Stokes hydrodynamics, the Laplace transform of the force $\widehat{\mathbf{F}}_{f\to p}(s)$ exerted by a Newtonian fluid on a spherical particle of radius R_p (s is the Laplace variable) is given by [22]

$$\widehat{\mathbf{F}}_{f\to p}(s) = -6\pi\mu R_p\, \widehat{\mathbf{v}}(s) - 6\pi\sqrt{\rho\mu} R_p^2 \frac{1}{\sqrt{s}} \left(s\, \widehat{\mathbf{v}}(s)\right) - \frac{2}{3}\rho\pi R_p^3 \left(s\, \widehat{\mathbf{v}}(s)\right) . \tag{20}$$

Transforming this equation back to the time domain and neglecting the fluctuations $\mathbf{R}(t)$, we obtain the evolution equation

$$m\, \dot{\mathbf{v}}(t) = -\eta\, \mathbf{v}(t) - \beta \frac{1}{\sqrt{t}} * \left[\dot{\mathbf{v}}(t) + \mathbf{v}(0)\delta(t)\right] \, , \tag{21}$$

where $\eta = 6\pi\mu R_p$, $\beta = 6\sqrt{\pi\rho\mu}R_p^2$ and $\mathbf{v}(0) = \mathbf{v}(t=0)$. For the sake of simplicity, we neglect in this equation the added mass term. While this is of physical relevance in other contexts, it is not important for our current discussion.

The dissipative and inertial functionals, L_d and L_i, are defined as

$$L_d[\mathbf{v}] = -\eta\,\mathbf{v} \tag{22a}$$

$$L_i[\dot{\mathbf{v}}] = -\beta\frac{1}{\sqrt{t}} * [\dot{\mathbf{v}}(t) + \mathbf{v}(0)\delta(t)]\,. \tag{22b}$$

For the following discussion it suffices to restrict ourselves to the one-dimensional case. Let $c(t) = \langle v(t)v(0)\rangle/\langle v^2\rangle$ so that $c(0) = 1$ and eq. (21) becomes

$$m\,\dot{c}(t) = -\eta\,c(t) - \beta\int_0^t \frac{1}{\sqrt{t-\tau}}\frac{dc(\tau)}{d\tau}d\tau - \frac{\beta}{\sqrt{t}}\,. \tag{23}$$

Equation (23) coincides with the analogous relation obtained by Widom, eq. (9) in [12], Observe that LRT does not provide the estimate for $\langle v^2\rangle$, which should be derived from kinetic/hydrodynamic arguments.

Introducing a dimensionless time $t' = t/t_{\mathrm{diss}}$, rescaled with respect to the dissipation time $t_{\mathrm{diss}} = m/\eta$, eq. (23) becomes

$$\dot{c}(t') = -c(t') - \gamma\int_0^{t'} \frac{1}{\sqrt{t'-\tau}}\frac{dc(\tau)}{d\tau}d\tau - \frac{\gamma}{\sqrt{t'}} \qquad \text{where} \qquad \gamma = \left(\frac{9}{2\,\pi}\frac{\rho}{\rho_p}\right)^{1/2} = \frac{t_{\mathrm{diss}}}{t_{\mathrm{inert}}} \tag{24}$$

expresses the ratio of the dissipation time t_{diss} to the characteristic time t_{inert} for the occurrence of inertial effects. For $\rho = 10^3\rho_p$ (such as for gas bubbles in water), $\gamma = 38$, while for $\rho_p = 5\cdot 10^3\rho$ (heavy solid particles in air), $\gamma = 1.7\cdot 10^{-2}$, so that the physical range of values of γ is $(10^{-2}, 10^2)$. Figure 1 shows the behaviour of $c(t)$, obtained by solving eq. (23), in non-dimensional form for several values of γ.

Apart from the well-known $t^{-3/2}$ long-term scaling induced by the effect of the Basset force, and typical for Brownian motion in liquids [42–49], Fig. 1 indicates that the influence of γ is significant in modulating the short-time behaviour of the velocity autocorrelation function. This observation will be further addressed in Sec. VI.

V. FLUCTUATION-DISSIPATION THEORY: A CHANGE OF PERSPECTIVE

In the previous sections, we have developed the formalism leading to the formulation of the hydrodynamic Langevin equations for particle motion in a fluid medium in terms of the stochastic velocity field $\mathbf{v}_s(t)$. The formal simplicity of the approach, based on the velocity

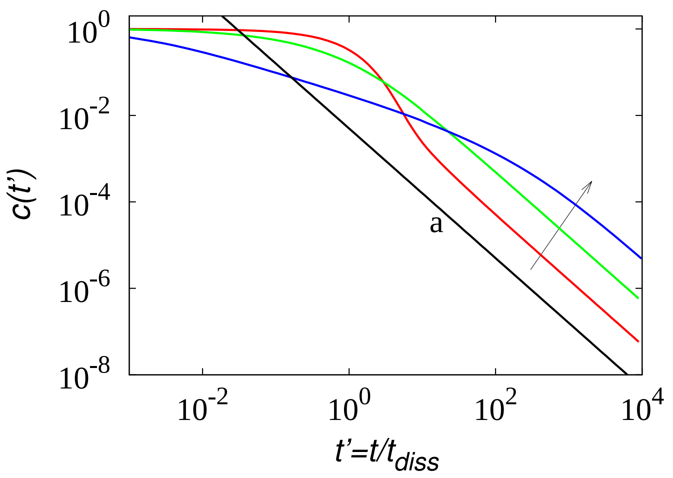

FIG. 1. Normalized autocorrelation function $c(t')$ vs $t' = t/t_{\rm diss}$ for a spherical particle in a still fluid, the dynamics of which is defined by eqs. (21). The arrows indicate increasing values of the nondimensional parameter $\gamma = 10^{-1}, 1, 10$. Line (a) represents the long-term scaling $c(t') \sim (t')^{-3/2}$.

splitting and on LRT, may suggest that it would represent an alternative "reshuffling" of known concepts without any major novelty, nor significant physical meaning. Here, we compare and contrast the proposed approach against the existing FD theory, showing not only its relevance for cases of physical importance, but also how to modify the perception of known FD relations.

The FD analysis of Langevin equations has been developed by considering dissipative memory effects, i.e., with reference to eq. (1). The hydrodynamic approach to Brownian motion in fluids made clear lately that fluid inertia (deriving from a time-dependent Stokesian analysis of fluid-particle interactions) represents an important correction at short times to the long-term picture based on instantaneous dissipation [11, 12]. Consider eq. (4) for a spherical particle of radius R_p in a Newtonian fluid with viscosity μ and density ρ. In this case, $h(t) = \eta\delta(t)$, whereas $k(t)$ is given by eq. (5). This problem has been investigated extensively in experiments, which have shown the importance of the fluid inertia in determining the short-time dynamics of these Brownian particles [8–10, 30, 31]. The presence of memory terms, depending on the history of the particle acceleration, makes the traditional approach based on Kubo FD theory [3] technically impossible, as correctly observed in [5].

Nevertheless, several authors [51, 52] have analyzed the representation of the thermal force $R(t)$ in the presence of fluid-inertial effects. Specifically, Bedeau and Mazur [52] derived for the Fourier transform $\widehat{R}(\omega)$ of the thermal force the following result:

$$\langle \widehat{R}(\omega)\, \widehat{R}(\omega') = 2\, k_B\, T Re[\widehat{\zeta}(\omega)]\, \delta(\omega - \omega') = \widehat{\zeta}_R(\omega)\, \delta(\omega - \omega')\ , \tag{25}$$

where

$$\widehat{\zeta}(\omega) = 6\, \pi\, \mu\, R_p \left[1 + (1 - \mathrm{i}) R_p (\omega\, \rho/(2\, \mu))^{1/2} - (\mathrm{i}\, \omega\, \rho\, R_p^2/9\, \mu)\right]\ . \tag{26}$$

Equations (25),(26) seem to suggest that Kubo fluctuation-dissipation theory can be applied also in this setting. However, we show below that this is ultimately problematic.

From eqs. (25),(26) it follows that the correlation function $\langle R(t)\, R(0)\rangle$ is the inverse Fourier transform $\zeta_R(t)$ of $\widehat{\zeta}_R(\omega)$ entering eq. (25). $\widehat{\zeta}(\omega)$ can be further simplified by renormalizing the term $\propto \omega$ into the added mass. The equation then reduces to

$$\widehat{\zeta}(\omega) = 6\, \pi\, \mu\, R_p \left[1 + (1 - \mathrm{i}) R_p (\omega\, \rho/(2\, \mu)^{1/2}\right]\ . \tag{27}$$

This relation can be used to derive scaling results for the thermal forces, such as

$$\langle R(t)\, R(0)\rangle \sim t^{-3/2} \tag{28}$$

near $t = 0$, as discussed in [9], which are consistent with experimental results for the power spectral density of the thermal noise [8–10]. However, it is important to observe that the power spectral density of $R(t)$ is a result of the post-processing of the experimental measurements of position and velocity of a Brownian particle. For this post-processing, a hydrodynamic model must be assumed. Therefore, $\langle R(t)\, R(0)\rangle$, or its Fourier transform, is not a directly measurable quantity.

In any case, due to the power-law singularity of eq. (28) near $t = 0$, we can show that there is no stochastic process possessing eqs. (25)-(26) as the Fourier transform of its autocorrelation function (see Appendix B). As such, these equations should be viewed as purely formal. In addition, we can also show that the fluid-inertial interactions expressed by the Basset term, leading to eq. (28), renders the representation of the thermal force unphysical and difficult to handle in practical Langevin simulations (Appendix B). This is not the case in the presence of only purely dissipative hydrodynamic effects (Appendix A). These observations altogether supports our view that standard FD theory is only an approximate description for Brownian dynamics in more general settings.

The velocity splitting approach in which the characterization of the thermal fluctuations is described by means of the stochastic velocity field $v_s(t)$ does not suffer these limitations. For any hydrodynamic model considered, its correlation function can be obtained using the LRT (see Sec. III). The resulting function is well behaved both at $t = 0$, at which it attains a constant value, and for $t \to \infty$, where it vanishes in an integrable way (as its integral is finite and corresponds to the particle diffusivity).

Most importantly, the stochastic velocity field $v_s(t)$ has a clear physical meaning, as it corresponds to the stochastic velocity of the particle in the absence of any external forcing or perturbations. Consequently, $v_s(t)$ is amenable, in principle, to a direct experimental measurement from the trajectory data of a free Brownian particle (which is not the case of the thermal force $R(t)$). However, it should be observed that in real experiments the use of an external forcing, in the form of a potential, e.g., deriving from the effects of an optical trap, is always needed in order to localize particle motion in a given region of the fluid domain, enabling the measurement of its position with optical techniques. In this case, the correlation functions of $v_s(t)$ can be directly obtained experimentally by reducing progressively the spring constant of the trap (see Sec. VI).

It follows from the above discussion that introducing $v_s(t)$ as the descriptor of thermal fluctuations yields a different approach to FD theory. In classical statistical mechanics FD relations, expressed by eqs. (2)-(3), are aimed at (i) connecting velocity fluctuations to the properties of the hydrodynamic response function (eq. (2)); and (ii) characterizing the thermal force $R(t)$ in terms of the hydrodynamic response function (eq. (3)). The action of hydrodynamic/thermal fluctuations has thus been split into two contributions: dissipation (i.e., $h(t)$), and fluctuations (i.e., $R(t)$), and eq. (3) is representative of the link connecting them. While this approach works extremely well for eq. (1) (see also Appendix A), problems arise in the presence of fluid inertial contributions (Appendix B). The presence of hydrodynamic inertia makes this classical dichotomic description blurred, in the meaning that fluid-inertial contributions cannot be ascribed neither to pure dissipation nor to fluctuations. The present theory based on the stochastic hydrodynamic velocity field formalizes this concept, indicating that the separation between fluctuation and dissipation in the description of thermal motion is, essentially, an epistemic approach grounded on a model perspective of splitting the various forces acting on the particle. Such a splitting derives from comprehensible reasons, as it stems from the results on linear hydrodynamics in the

Stokes or in the time-dependent Stokes regime [22] to which the thermal fluctuations are added as a further contribution. The theory of Stochastic Hydrodynamics [53] provides a further interpretation of this result. Nonetheless, this is still a model-based assuption in which the fluctuational term $\mathbf{R}(t)$ is added by physical necessity (as in the original paper by Langevin [4]). The stochastic velocity approach not only resolves practical problems, such as those discussed above in connection with the Basset force and further in the next section, but treats the fluctuations in a unitary perspective, separating the role of thermal fluctuations from the hydrodynamic effects. As a consequence, given $v_s(t)$, no additional FD relations are needed for describing thermal effects. It puts stochasticity at the center of the physical focus, and asks for a physical interpretation of it. In the next two sections, a variety of examples are discussed in which different models for the stochastic fluctuations, still possessing the same velocity autocorrelation function, provide different, and experimentally measurable, predictions. Therein the formulation of the stochastic velocity approach is finalized for catalyzing new interest in this direction.

VI. STOCHASTIC REPRESENTATION OF HYDRODYNAMIC/THERMAL VELOCITY FLUCTUATIONS

The velocity splitting approach shifts the stochastic description of particle motion from the characterization of the fluctuational force $\mathbf{R}(t)$ to the representation of the comprehensive hydrodynamic/thermal stochastic velocity field $\mathbf{v}_s(t;\mathbf{x})$. This naturally provides a different setting for representing the stochastic velocity field in terms of elementary stochastic processes. This aspect is fundamental in order to develop accurate stochastic Lagrangian descriptions (Langevin equations) for particle motion.

In the early days of Einstein-Langevin investigations of Brownian motion, the description of thermal fluctuations made use of stochastic processes possessing no memory, i.e., characterized by an impulsive correlation function. The use of Wiener processes $w(t)$, and of their distributional derivatives $\xi(t) = dw(t)/dt$ (white noise), is the natural and most convenient choice related to this level of approximation. This led to equations of motion of the form

$$\dot{x}(t) = v(t) \tag{29a}$$

$$m\dot{v}(t) = -\eta v(t) + \sqrt{2k_B T\eta}\,\xi(t)\,, \tag{29b}$$

where $\langle\xi(t')\xi(t)\rangle = \delta(t-t')$. The use of δ-correlated stochastic processes was adequate to this level of approximation representing the physical phenomenology of Brownian motion [50], essentially because the hydrodynamic interactions were considered to be instantaneous, as expressed exclusively by the Stokesian drag. Reinterpreted in the light of the velocity decomposition developed above, the statistical properties of particle motion defined by eqs. (29) are equally well predicted by the kinematic model

$$\dot{x}(t) = v_s(t) \; , \tag{30}$$

where $v_s(t)$ is any stochastic process possessing zero mean and exponential correlation function $\langle v_s(t)v_s(0)\rangle = \langle v^2\rangle e^{-\eta t/m}$. We remark that $v_s(t)$ represents the equilibrium velocity fluctuations, hence eq. (30) should not be confused with the classical overdamped approximation, valid in the limit $m \to 0$, as $v_s(t)$ in the present case is not δ-correlated. For instance, one could choose for $v_s(t)$ the Poisson-Kac process [23]

$$v_s(t) = b_0(-1)^{\chi(t,\lambda)} \; , \tag{31}$$

where $\chi(t,\lambda)$ is a Poisson counting process characterized by the transition rate $\lambda > 0$. Since $\left\langle(-1)^{\chi(t,\lambda)}(-1)^{\chi(0,\lambda)}\right\rangle = e^{-2\lambda t}$, the Poisson-Kac process fits the physical requirements, provided that $b_0^2 = \langle v^2\rangle = k_B T/m$ and $\lambda = \eta/2m$. This approach has been applied successfully even in the presence of potentials [34]. There is, however, an important caveat. While the choice eq. (31), applied to the kinematics eq. (30), reproduces correctly all the statistical properties of particle diffusional dynamics defined by eqs. (29), it fails for describing the equilibrium velocity probability density function. This is so, because eq. (31) represents a one-velocity model, characterized by a single velocity value b_0, which determines an impulsive probability density function for the velocity v_s, $p_v(v_s) = [\delta(v_s + b_0) + \delta(v_s - b_0)]/2$.

But in point of fact, this problem has a simple solution, as in all the cases where an accurate reproduction of the velocity statistics is required a Generalized Poisson-Kac process $\Xi_g(t,\lambda;v)$, possessing velocity as a continuous transitional variable [26, 29], can be used instead of the conventional dichotomous Poisson-Kac process (31),

$$v_s(t) = \Xi_g(t,\lambda;v) \; . \tag{32}$$

The process $\Xi_g(t,\lambda;v)$ is continuously parametrized with respect to the velocity $v \in \mathbb{R}$, possesses an exponential statistics of transition times specified by the transition rate λ, and is such that the probability density function for v would be any equilibrium function $g(v)$, for instance, the Maxwellian distribution $g(v) = Ae^{-mv_s^2/2k_BT}$, where A is the normalization constant. The statistical characterization of $\Xi_g(t,\lambda;v)$ involves the probability density $P_\Xi(v,t)$, $v \in \mathbb{R}$,

$$P_\Xi(v',t)dv' = \text{Prob}[\,\Xi_g(t,\lambda;v) \in (v', v'+dv')\,]\,, \tag{33}$$

that satisfies the balance equation

$$\frac{\partial P_\Xi(v,t)}{\partial t} = -\lambda\, P_\Xi(v,t) + \lambda\, g(v) \int_{-\infty}^{\infty} P_\Xi(v',t)dv'\,. \tag{34}$$

Consequently, the statististical properties of the process $X(t)$ (we use the notation $X(t)$ for the process and $x(t)$ for a realization of it), defined by the kinematic eq. (30), are described by the probability density $p(x,v,t)$ satisfying the linear Boltzmann equation

$$\frac{\partial p(x,v,t)}{\partial t} = -v\frac{\partial p(x,v,t)}{\partial x} - \lambda\, p(x,v,t) + \lambda\, g(v) \int_{-\infty}^{\infty} p(x,v',t)\,.\,dv'\,. \tag{35}$$

It is important to observe that the simulation of the process $\Xi_g(t,\lambda;v)$ is as simple as the Poisson-Kac process $(-1)^{\chi(t,\lambda)}$ since, at any transition time τ, whose statistics is defined by the exponential density $p_\tau(\tau) = \lambda e^{-\lambda\tau}$, a new velocity variable is selected, independently of the previous one, from the equilibrium distribution $g(v)$. Because of this property, the correlation function $c_\Xi(t) = \langle\, \Xi_g(t,\lambda;v)\,\Xi_g(0,\lambda;v)\,\rangle$ is given by

$$C_\Xi(t) = \sigma_v^2 e^{-\lambda t}\,, \quad \sigma_v^2 = \int_{-\infty}^{\infty} v^2\, g(v)\, dv \tag{36}$$

independently of the functional form of the velocity probability density $g(v)$. Figure 2 compares the velocity probability density function and the correlation function obtained from stochastic simulations of the process $\Xi_g(t,\lambda;v)$ for the case where $g(v)$ is the normal distribution for different values of λ. Simulations involve an ensemble of 10^8 realizations. The above example requires a further comment. Generalized Poisson-Kac processes have been developed in order to generate stochastic dynamics characterized by bounded propagation velocity [25]. The choice of a Gaussian probability density $g(v)$ for the statistics of $\Xi_g(t,\lambda;v)$ is conceptually in contradiction with this founding principle. The mathematical occurrence of the Gaussian distribution can be justified by invoking the Central Limit Theorem, and therefore it represents a long-term asymptotics. As regards particle velocities, the application of the Central Limit Theorem is physically limited by relativistic constraints, due to the fact that for large velocities the assumption of independence among the velocity entries fails.

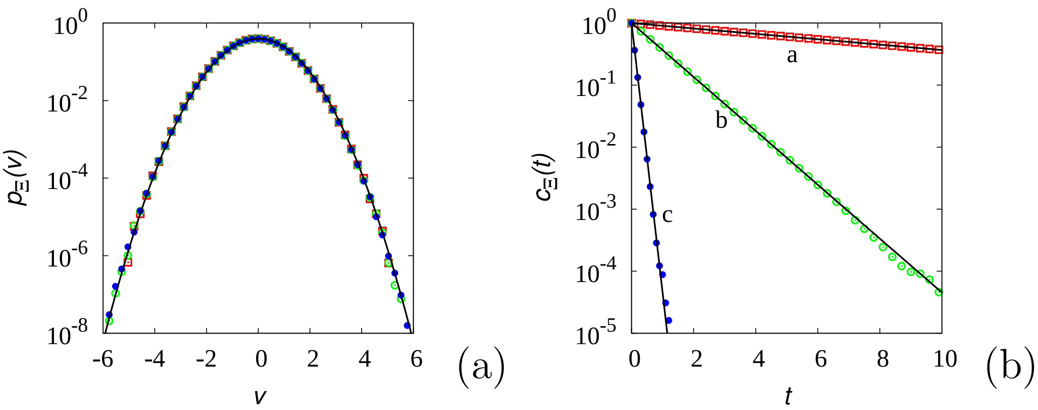

FIG. 2. Statistical properties of the process $\Xi_g(t,\lambda;v)$ with $g(v)=e^{-v^2/2}/\sqrt{2\pi}$. Panel (a): Equilibrium probability density function $p(v)$ for $\Xi_g(t,\lambda;v)$. Symbols corresponds to the results of stochastic simulations at different values of $\lambda = 10^{-1}$, $1, 1, 10$. The solid line represents the normal probability density $g(v)$. Panel (b): Correlation function $c_\Xi(t)$ vs. t. Symbols corresponds to the results of stochastic simulations, lines to the exponential functions $c_\Xi(t) = e^{-\lambda t}$, eq. (36) since $\sigma_v^2 = 1$. Line(a) and ($\square$): $\lambda = 10^{-1}$; line (b) and ($\circ$): $\lambda = 1$, line (c) and ($\bullet$): $\lambda = 10$.

The Maxwellian distribution thus represents an excellent approximation of the relativistic Jüttner distribution [54, 55] that, in the low-velocity/low-temperature limit (with respect to the speed of light in vacuo), is practically indistinguishable from its relativistic counterpart. It follows from the above reasoning that if we require the process $X(t)$ (i.e. the particle position) to possess a bounded propagation velocity, the process $\Xi_g(t,\lambda;v)$ defining $v_s(t)$ via eq. (32) should be constructed, e.g., by using a truncated Maxwellian distribution, i.e.,

$$g(v) = \begin{cases} A e^{-mv^2/2k_BT} & v \in (-v_{\max}, v_{\max}) \\ 0 & \text{otherwise} \end{cases} \tag{37}$$

with $v_{\max} \gg \sqrt{k_BT/m}$ but still bounded, in order to fulfill the Maxwellian equilibrium distribution in its physical range of validity, but still satisfying the requirement of bounded propagation.

The above construction applies a fortiori for particle dynamics accounting for inertial hydrodynamic effects. Considering the case analyzed in Sec. III, due to the occurrence of asymptotic power-law tails in the velocity correlation function, the natural candidates for modeling $v_s(t)$ are Lévy Walks [28, 56]. The classical Lévy Walk model is characterized by a transition rate $\lambda(\tau)$ depending on the transition age τ (the transition age corresponds to the time elapsed from the latest transition) in the form of [56]

$$\lambda(\tau) = \frac{\xi}{1 + \tau/\tau_0} \tag{38}$$

with $\xi > 1$ and $\tau_0 > 0$. The equilibrium correlation function of this process fulfills the long-term scaling [28]

$$\langle v_s(t) v_s(0) \rangle \sim t^{-(\xi-1)} \, . \tag{39}$$

For the problem considered in Sec. III, the $t^{-3/2}$-hydrodynamic scaling is matched by taking $\xi = 5/2$. The same approach, applied above in the case of Poisson-Kac processes to obtain a continuous distribution of velocities, can be verbatim enforced in order to define a Lévy Walk $\Lambda_g(t, \xi, \tau_0; v)$, characterized by the transition rate eq. (38), by a continuous probability density function $g(v)$ and by the correlation properties pertaining to the corresponding one-velocity Lévy Walk model.

The above generalizations, either for Poisson-Kac, Generalized Poisson-Kac processes or Lévy Walks, have been analyzed in a broader stochastic framework in [29], introducing the class of Extended Poisson-Kac (EPK) processes that subsume the family of stochastic processes possessing Markov and semi-Markov transition mechanisms and arbitrary parametrization with respect to the transitional variables. It follows from the above analysis that the velocity splitting approach finds in EPK processes the natural and computationally simple candidates for expressing $v_s(t)$ (or $v_s(t, x)$). This means that $v_s(t)$ can be expressed with arbitrary accuracy by means of a linear combination of independent EPK processes,

$$v_s(t) = \sum_{h=1}^{N_\Lambda} a_h \, \Lambda_g(t, \xi_h, \tau_{0,h}; v) + \sum_{k=1}^{N_\Xi} b_k \, \Xi_g(t, \lambda_k; v) \, , \tag{40}$$

where the number of processes N_Λ, N_Ξ, the process parameters ξ_h, $\tau_{0,h}$, λ_k, and the expansion coefficients a_h, and b_k should be optimized with respect to the correlation function $\langle v_s(t) v(0) \rangle$. The details of the expansion eq. (40) , and of parameter optimization, are of little interest in the present analysis, and they will be developed elsewhere in the light of specific hydrodynamic applications. But the application of eq. (40) to Brownian motion and to problems deriving from transport in microfluidic systems opens up interesting and new research directions as briefly outlined in Sec. C.

A. Correlations in continuous vs. dichotomous models

In order to highlight the importance of the transition from dichotomous Poisson-Kac processes to EPK processes possessing a continuous parametrization with respect to velocity in the characterization of the $v_s(t)$, let us consider the case of a particle in a thermostated fluid environment at constant temperature T, subjected to an external potential $U(x)$. In the one-dimensional case, the particle dynamics reads

$$\begin{aligned} dx &= v\,dt \\ m\,dv &= -\eta\, v\,dt - \partial_x U(x)\,dt + \sqrt{2k_B T\eta}\,dw(t)\,. \end{aligned} \tag{41}$$

Introducing $t = T_c\,t'$, $x = L_c\,y$, $v = V_c\,u$, where T_c, L_c, V_c are the characteristic time, length and velocity scales, letting $U(x) = U_0\,\overline{U}(y)|_{y=x/L_c}$, and assuming $T_c = m/\eta = t_{\text{diss}}$, $V_c = \sqrt{k_B T/m}$, $L_c = V_c T_c$, eqs. (41) take the non-dimensional form

$$\begin{aligned} dy &= u\,dt' \\ du &= -u\,dt' - \alpha\,\partial_y \overline{U}(y)\,dt' + \sqrt{2}\,dw(t')\,, \end{aligned} \tag{42}$$

where $\alpha = U_0/mV_c^2$. Consider two cases. The first case is represented by a harmonic potential $U(x) = k_s x^2/2$, so that $U_0 = k_S L_c^2$. In this case, $\alpha = U_0/mV_c^2 = k_s L_c^2/mV_c^2 = k_s T_c^2/m$, i.e.,

$$\alpha = \frac{k_s\,m}{\eta^2} = \frac{t_{\text{diss}}}{t_k}\,, \tag{43}$$

where $t_k = \eta/k_s$ is the characteristic time associated with the coupling of the harmonic potential and frictional dissipation, and $\partial_y \overline{U}(y) = y$. This case represents the dynamics of a Brownian particle (neglecting fluid inertial effects) in an optical trap, and physically reasonable values for α range from 10^{-2} to 10^{-1} [30]. Specifically, fluid-inertial effects are negligible if the fluid is a gas [30]. Figure 3 panel (a) depicts the comparison of the velocity autocorrelation function obtained from the direct simulation of eq. (41), for $\alpha = 0.1$, using $N = 10^6$ realizations of the process, with the one from the velocity split representation

$$\begin{aligned} dy &= (u_d + u_s(t))\,dt' \\ du_d &= -u_d\,dt' - \alpha\,\partial_y \overline{U}(y)\,dt'\,, \end{aligned} \tag{44}$$

where the nondimensional stochastic velocity $u_s(t)$ is represented by a Poisson-Kac dichotonous stochastic process. In this case, the dichotomous model correctly reproduces the behaviour of the velocity autocorrelation function, as predicted by LRT.

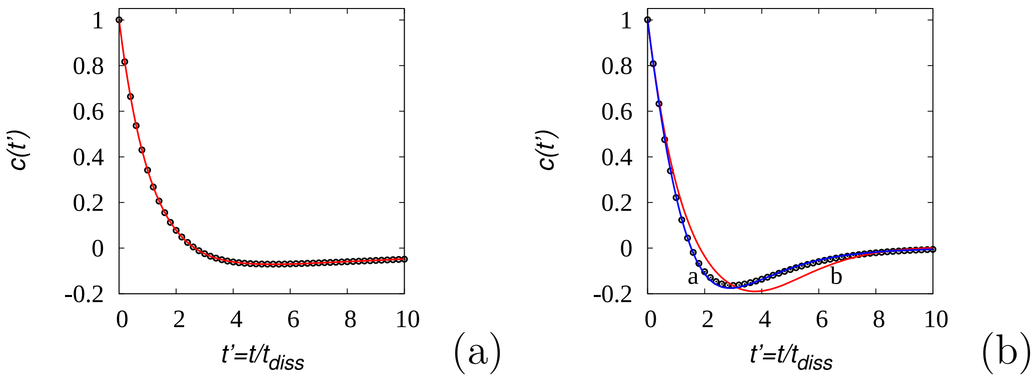

FIG. 3. Velocity autocorrelation function $c(t')$ vs $t' = t/t_{\rm diss}$ associated with the stochastic dynamics eq. (42). Symbols (•) corresponds to the results of the stochastic simulation of eq. (42) Panel (a) refers to a harmonic potential at $\alpha = 0.1$. The solid line represents the velocity autocorrelation function obtained from the velocity split model eq. (44) describing $u_s(t)$ by means of a dichotomous Poisson-Kac process. Panel (b) refers to the case of a bistable potential described in the main text. Line (a) represents the velocity autocorrelation function obtained using a continuously parametrized EPK process $\Xi(t', 1; v)$ possessing a Maxwellian velocity density function, line (b) the corresponding result adopting for $u_s(t')$ a dichotomous Poisson-Kac process.

This model provides clear evidence for what was stated in Sec. V, and specifically that the velocity autocorrelation function $\langle v_s(t)\, v_s(0)\rangle$ can be measured in physically realizable experiments. Indeed, still keeping the Brownian particle confined in a trap (and therefore under the influence of a harmonic potential), by decreasing the trap spring constant and measuring the corresponding particle velocity autocorrelation function $\langle v(t)\, v(0)\rangle$, one obtains direct and accurate experimental measurements of the free-particle velocity autocorrelation function, i.e., of $\langle v_s(t) v_s(0)\rangle$ (see Fig. 4). For values of the nondimensional parameter α less or equal to 10^{-2}, the velocity autocorrelation function of the trapped particle represents a sufficiently accurate representation for that of the free particle, and thus of the autocorrelation function of $u_s(t)$. In experiments involving trapped micrometric particles, the values of α are usually small enough to operate in this limit [30]. Next, consider a nonlinear potential, such as the bistable potential expressed in nondimensional form by $\overline{U}(y) = y^4/4 - y^2/2$. Figure 3 panel (b) depicts the temporal behaviour of the velocity autocorrelation function obtained from the stochastic simulation of eq. (42) at $\alpha = 0.1$, and for the velocity split model eq. (44)

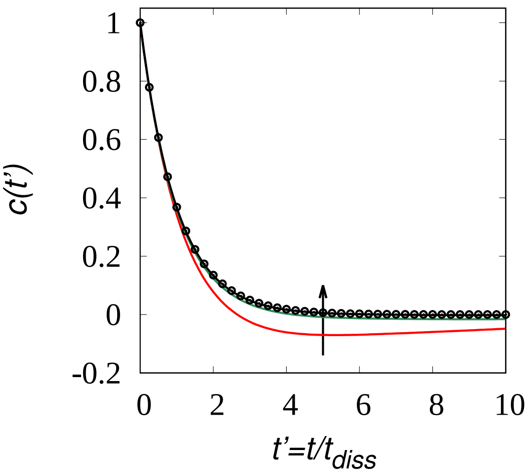

FIG. 4. Velocity autocorrelation function $c(t')$ vs. $t' = t/t_{\rm diss}$ associated with the stochastic dynamics eq. (42) in the presence of a harmonic potential for different values of α, i.e., of the nondimensional spring constant, $\alpha = 10^{-1}$, 10^{-2}, 10^{-3}. The arrow indicates decreasing values of α. Symbols ($\circ$) correspond to the free-particle velocity autocorrelation function, $c(t') = e^{-t'}$.

in the case $u_s(t)$ is an EPK process continuously parametrized with respect to the velocity and possessing a Maxwellian distribution (line a) and a dichotomous Poisson-Kac process (line b). Owing to the nonlinearity of the model, the importance of a continuous distribution of velocity is evident. While the EPK process $\Xi(t, \lambda; v)$ accurately reproduces the velocity autocorrelation function, this is not the case for the dichotomous Poisson-Kac counterpart. This indicates that when moving towards nonlinear models, in the present case expressed by a non-quadratic potential, the fine structure of the velocity fluctuation becomes important.

B. On the fine structure of the thermal fluctuations

In Kubo FD theory, and in transport modeling, the velocity autocorrelation function is the statistical quantity used to characterize thermal fluctuations. This is inherent to transport theory, as the integral over time of the velocity autocorrelation function returns the diffusion coefficient (Green-Kubo theorem). More generally, the whole architecture of statistical physics of non-equilibrium phenomena is grounded on the autocorrelation functions of fluctuating fluxes and forces [57, 58].

The recent advances in the experimental analysis of Brownian motion at short time scales have revealed the possibility of characterizing Brownian fluctuations beyond the limit that "Einstein deemed possible" [50]. The experimental investigation of Brownian motion can nowadays provide a deeper understanding of the physical meaning of thermal noise, of the interaction between hydrodynamic and thermodynamic properties, and possibly of the macroscopic dissipative effects of quantum fluctuations, either at the level of radiative interactions with molecules and particles (involving photon exchange) or at the field level involving zero-point fluctuations. In this perspective, the analysis of the fine properties of the stochastic velocity field $v_s(t)$ and of its influence on macroscopic and experimentally measurable quantities becomes central.

Consider again the case of a Brownian particle in a trap (i.e., where $U(x)$ is a harmonic potential) described by means of eq. (42). Experimentally, eq. (42) corresponds to particle motion in a gaseous environment at thermal equilibrium for which the fluid-inertial interactions are negligible. Consider the other second-order correlation functions, namely $C_{yy}(t') = \langle y(t')\, y(0)\rangle$, $C_{yu}(t') = \langle y(t')\, u(0)\rangle$, and $C_{uy}(t') = \langle u(t')\, y(0)\rangle$. Due to the linearity of the dynamics, any stochastic representation of $u_s(t')$, entering eq. (44) and possessing the correct exponential decay of the velocity autocorrelation function, would provide the same temporal behavior for all the second-order correlation functions. This phenomenon is depicted in Fig. 5, where the correlation function obtained from the solutions of eq. (42) are compared with the corresponding ones obtained from eq. (44) adopting for $u_s(t')$ an EPK model possessing a Maxwellian probability density function as described and used in the previous paragraph. An analogous result could be obtained adopting for $u_s(t')$ a dichotomous one-velocity Poisson-Kac model.

Recent experimental studies suggest that velocity fluctuations of Brownian particles both in gases and liquids could be characterized by a much more regular (smooth) behavior than that predicted by the classical Einsteinian theory eq. (42), based on Wiener processes as a model of thermal fluctuations [10, 50]. The experimental assessment of the regularity of thermal fluctuations represents an important issue in statistical physics, as it would suggest the necessity of considering more regular stochastic processes for the modeling of particle transport. Beside a regularity analysis of Brownian trajectories, clear experimental evidence of these properties may come from the estimate of higher-order correlation functions. Figure 6 depicts the comparison of the third and fourth-order velocity autocorrelation functions $C_{u2u}(t') = \langle u^2(t')\, u(0)\rangle$, $C_{u3u}(t') = \langle u^3(t')\, u(0)\rangle$, obtained from the classical Wiener-based

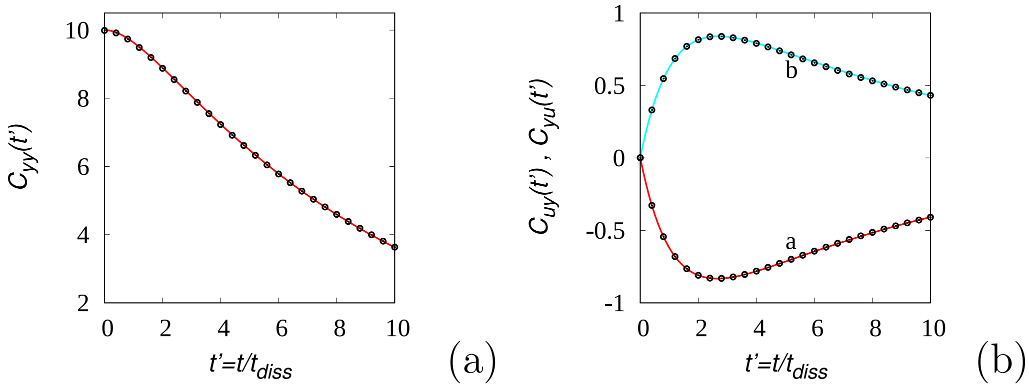

FIG. 5. Second-order correlation functions associated with the stochastic dynamics eq. (42) in the presence of a harmonic potential with $\alpha = 0.1$. Solid lines represent the correlation functions obtained from the velocity split model eq. (44), using for $u_s(t')$ a continuously parametrized EPK process $\Xi(t', 1; v)$ possessing a Maxwellian velocity probability density function. Panel (a): $C_{yy}(t')$ vs. t'. Panel (b): $C_{uy}(t')$ (line a) and $C_{yu}(t')$ (line b) vs. t'. Dots correspond to correlation functions obtained from solving eq. (42).

model eq. (42), and from eq. (44) in the presence of a piecewise smooth description of the stochastic velocity field $u_s(t')$ via the EPK model described in the previous section, possessing a Maxwellian velocity distribution function. While $C_{u2u}(t') = 0$ in both cases, a slight difference between the two descriptions of the thermal fluctuations is observed in the fourth-order autocorrelation function $C_{u3u}(t')$, although this difference is too small to be an object of experimental scrutiny. Conversely, a significant, and experimentally verifiable discrepancy between the Wiener description and and a smoother regular descriptions of thermal velocity fluctuations characterizes higher-order (fourth-order) mixed correlation functions, such as $C_{y2v2}(t') = \langle y^2(t')\, u^2(0)\rangle$ or $C_{v2y2}(t') = \langle u^2(t')\, y^2(0)\rangle$, depicted in Fig. 7.

We may therefore conclude from these qualitative observations that the experimental analysis of higher-order correlation functions of position and velocity variables may provide a way, not only for obtaining a finer statistical characterization of the stochastic velocity field $v_s(t)$, but also for verifying quantitatively, via stable and accurate experimental measurements, the validity of the Einsteinian paradigm for thermal fluctuations based on almost everywhere singular stochastic processes (Wiener process), and to assess experimentally the regularity of thermal fluctuations. This would close the circle on the properties of Brownian

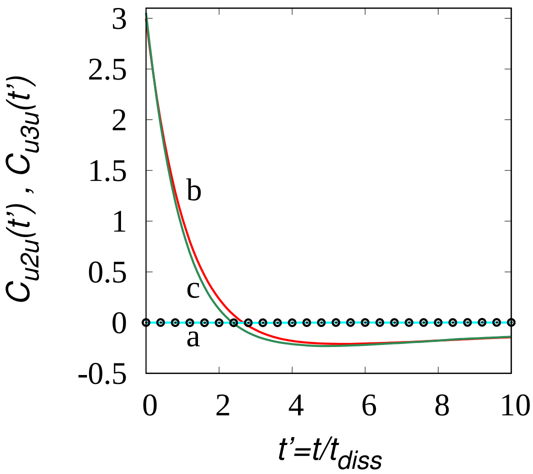

FIG. 6. Higher-order velocity autocorrelation functions associated with the stochastic dynamics eq. (42) in the presence of a harmonic potential with parameter $\alpha = 0.1$. Line (a) and line (b) refer to $C_{u2u}(t')$ and $C_{u3u}(t')$, respectively, obtained from the solutions of eq. (42). Symbols ($\circ$) and line (c) refer to $C_{u2u}(t')$ and $C_{u3u}(t')$, respectively, obtained from the solutions of eq. (44) using for $u_s(t')$ a continuously parametrized EPK process $\Xi(t', 1; v)$ possessing a Maxwellian velocity probability density function.

motion that at times of Einstein would be unthinkable to assess experimentally [50].

VII. CONCLUDING REMARKS

The classical FD formalism involves a decomposition of hydrodynamic/thermal effects into a dissipative contribution and a purely stochastic fluctuational term, including eventually fluid-inertial back action. The separation of fluid inertial effects (the Basset force) from the thermal contribution $\mathbf{R}(t)$ is essentially a technical simplification not related to the physics of the problem, which conversely indicates a highly correlated motion of the particle and of its nearby fluid elements [14]. An alternative to this approach is to consider all these hydrodynamic/thermal effects as a single fluctuating field possessing prescribed statistical and correlation properties. This is the key idea of the velocity splitting approach that we developed here, which provides a comprehensive description of these interactions in terms of the stochastic process $\mathbf{v}_s(t)$, or the stochastic field $\mathbf{v}_s(t; \mathbf{x})$. The correlation properties of $\mathbf{v}_s(t, \mathbf{x})$ can be derived from linear response theory even in those cases - as for particle

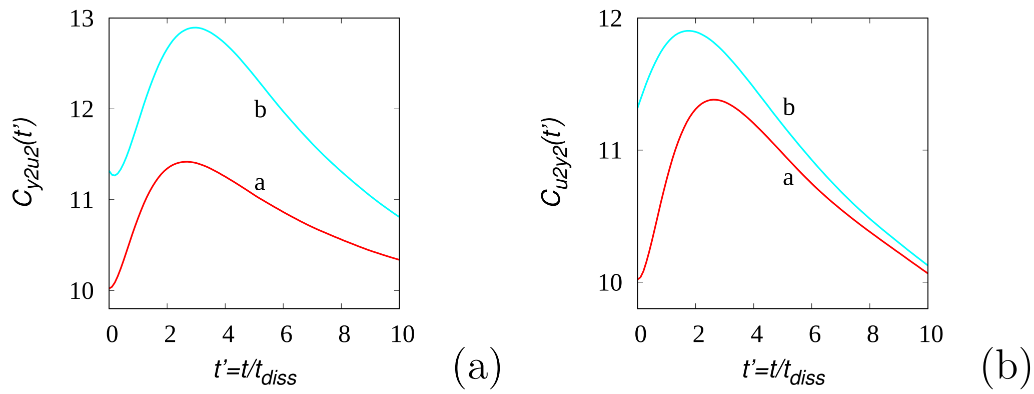

FIG. 7. Higher-order mixed correlation functions for a particle in a harmonic potential. Panel (a) depicts $C_{y2u2}(t')$ vs. t', panel (b) $C_{u2y2}(t')$ vs. t'. Lines (a) refer to the solutions of eq. (42), lines (b) to the solutions of eq. (44) using for $u_s(t')$ a continuously parametrized EPK process $\Xi(t', 1; v)$ possessing a Maxwellian velocity probability density function.

transport in confined systems - where both dissipative and inertial effects depend on the particle position, by considering the conditional correlation functions $\mathbf{c}^{(k)}(t \,|\, \mathbf{x})$.

Within this framework, the stochatic force $\mathbf{R}(t)$ is no longer necessary, as it is encompassed, together with all the (dissipative/inertial) hydrodynamic thermal interations, in the stochastic velocity field $\mathbf{v}_s(t; \mathbf{x})$. This field is a directly measurable quantity in short-time Brownian motion experiments, which is not the case for the thermal force $\mathbf{R}(t)$ in hydrodynamic problems involving not only friction but also fluid inertial effects. In the latter case we have shown that, apart from formal results, it is difficult to obtain a computationally operative definition of $\mathbf{R}(t)$ in terms of elementary stochastic processes to be used in the numerical simulation of the corresponding Langevin equations.

For modeling $\mathbf{v}_s(t; \mathbf{x})$, the class of EPK processes emerges as a natural choice, due to their simplicity in the implementation and due to the typical exponential/power-law behaviour of their correlation functions. The decomposition of a generic stochastic velocity field $\mathbf{v}_s(t; \mathbf{x})$ into a family of EPK process is an interesting computational problem that will be addressed in forthcoming works.

Pursuing this alternative approach, and comparing it with the results derived from classical Wiener-based Langevin equations, in several examples we have shown that

- the sole velocity autocorrelation function is not sufficient to describe correctly Brownian motion and micrometric particle dynamics in the presence of nonlinear potentials, which points at a more detailed characterization (both theoretically and experimentally) of thermal fluctuations;

- higher-order correlation functions, or correlation functionals, can be used as probes for determining the fine structure of the stochastic fluctuations.

In this manuscript, we have mainly considered the description of $\mathbf{v}_s(t)$ in the free space, where no external deterministic accelerations $\mathbf{a}(\mathbf{x})$ or position-dependent hydrodynamic effects are present. Whenever present, these determine the explicit dependence of the linear functionals L_d and L_i on $\mathbf{x}$. The presence of external position-dependent fields generating $\mathbf{a}(\mathbf{x})$ does not present any conceptual issue, because the statistical structure of the hydrodynamic/thermal stochastic velocity $\mathbf{v}_s(t)$ is independent of $\mathbf{a}(\mathbf{x})$ (see Subsec. VI A). In the case of position-dependent hydrodynamic interactions, which occurs for particles in confined geometries, such as in microchannels of transversal length scale comparable to the particle diameter, the statistical properties of the stochastic velocity field $\mathbf{v}_s(t;\mathbf{x})$ can be derived, for fixed $\mathbf{x}$, from the conditional normalized correlation functions $\mathbf{c}^{(k)}(t\,|\mathbf{x})$ satisfying eqs. (16) (see Sec. III). In this setting, it is no longer possible to derive a priori the velocity correlation function, or the statistical characterisation of the spatial particle distribution, even in the long-term limit (if an asymptotic equilibrium distribution emerges in the system). At present, these can be obtained solely from the direct numerical simulations of the equations. The theoretical prediction of these properties represents a major technical issue, whose solution would necessarily rely on the extension of LR theory to the nonlinear case. The outline of a specific setup to be studied is further described in Appendix C.

The experimental characterization of the fine structure of the stochastic velocity fields $\mathbf{v}_s(t;\mathbf{x})$ defines another important topic for future investigations that can be addressed by measuring higher-order correlation functions. This analysis could potentially be used to check for the reliability of the singular Wiener-based approach to characterize thermal fluctuations at short time scales, which is in contrast to a more smooth and piecewise regular description of equilibrium fluctuations that seems to emerge from experiments [50].

We conclude by observing that while the classical decomposition of hydrodynamic/thermal interactions into dissipative, inertial, and fluctuational forces follows naturally from a reductionistic Newtonian approach of accounting for all the "distinct" forces acting on a material body, our formulation of the statistical physical properties of a particle in a thermalized fluid, based on the stochastic velocity field $\mathbf{v}_s(t;\mathbf{x})$, shifts the focus of the description on the kinematic equation for the particle motion driven by the "almost deterministic" velocities $\mathbf{v}_d(t)$, and by the stochastic velocities $\mathbf{v}_s(t;\mathbf{x})$. This alternative decomposition bears some analogies with the Aristotelian description of motion, where $\mathbf{v}_d(t)$ expresses the "violent motion" and $\mathbf{v}_s(t;\mathbf{x})$ the "natural motion" in thermal systems at equilibrium [64].

Appendix A: Modal decomposition of the thermal force - The case of purely dissipative hydrodynamics

Consider the dissipative Langevin equation (1). In this case, the Kubo FD theory predicts for the correlation function of the stochastic forcing

$$\langle R(t)\, R(0)\rangle = m\, k_B\, T h(t)\,, \quad t \geq 0\,, \tag{A1}$$

where we have assumed $\langle \xi(t)\, \xi(0)\rangle = \delta(t)$ for the distributional derivative $\xi = dw(t)/dt$ of a Wiener process. The friction kernel $h(t)$ satisfies the property

$$h(t) \geq 0\,, \qquad t \geq 0\,. \tag{A2}$$

Let us assume that it admit a modal representation of the form

$$h(t) = \frac{1}{m} \sum_{i=1}^{\infty} a_i\, e^{-\lambda_i\, t}\,, \tag{A3}$$

where a_i, $\lambda_i > 0$, $\sum_{i=1}^{\infty} a_i < \infty$. The stochastic forcing $R(t)$ can be expressed in the following form,

$$R(t) = \sum_{i=1}^{\infty} \sqrt{k_B\, T\, b_i}\, \psi_{\lambda_i}(t)\,, \tag{A4}$$

where $\psi_{\lambda_i}(t)$, $i = 1, \ldots, n$ are stochastic processes, independent of each other and possessing exponential correlation functions

$$\langle \psi_{\lambda_i}(t)\, \psi_{\lambda_j}(t')\rangle = \delta_{i,j}\, e^{-\lambda_i\, |t-t'|}\,, \tag{A5}$$

where the positive constants $b_i > 0$ need to be determined. The correlation function of $R(t)$ is given by

$$\langle R(t)\, R(0)\rangle = k_B\, T \sum_{i=1}^{\infty} \sqrt{b_i} \sum_{j=1}^{\infty} \sqrt{b_j}\, \langle \psi_{\lambda_i}(t)\, \psi_{\lambda_j}(0)\rangle = k_B\, T \sum_{i=1}^{\infty} b_i\, e^{-\lambda_i\, t}\,, \tag{A6}$$

and from eqs. (A1), (A3) it follows that the expansion coefficients b_i are simply given by

$$b_i = a_i \,, \quad i = 1, \ldots \,. \tag{A7}$$

The processes $\psi_{\lambda_i}(t)$ characterized by the property eq. (A5) can be defined in many ways. For instance, they can be chosen in the form of filtered Wiener processes,

$$d\psi_{\lambda_i}(t) = -\lambda_i \, \psi_{\lambda_i}(t) \, dt + \sqrt{2\lambda_i} \, dw_i(t) \,, \tag{A8}$$

where $dw_i(t)$ are the increments of independent one-dimensional Wiener processes so that $\langle dw_i(t) \, dw_j(t) \rangle = \delta_{i,j} \, dt$. Or they can be represented in terms of Poisson-Kac processes,

$$\psi_{\lambda_i}(t) = (-1)^{\chi_i(t,\lambda_i/2)} \,, \tag{A9}$$

where $\chi_i(t, \lambda_i/2)$ are independent Poisson processes characterized by the transition rate $\lambda_i/2$, $\left\langle (-1)^{\chi_i(t,\lambda_i/2)} \, (-1)^{\chi_j(t,\lambda_j/2)} \right\rangle = \delta_{i,j}$. Or one may use independent EPK processes, possessing exponential correlation functions and arbitrary velocity probability density functions with zero mean and unit variance.

The same approach can be extended to a continuous representation. In that case $h(t)$ can be represented in the form

$$h(t) = \frac{1}{m} \int_0^\infty a(\lambda) \, e^{-\lambda \, t} \, dt \tag{A10}$$

with $\alpha(\lambda) \geq 0$. Here the thermal force can be expressed as

$$R(t) = \sqrt{k_B \, T} \int_0^\infty \sqrt{a(\lambda)} \, \psi_\lambda(t) \, d\lambda \,, \tag{A11}$$

where

$$\langle \psi_\lambda(t) \, \psi_\mu(t') \rangle = \delta(\lambda - \mu) \, e^{-\lambda \, |t-t'|} \,. \tag{A12}$$

It follows from this analysis that the thermal forces admit simple and compact representations in the presence of purely dissipative hydrodynamic contributions, and that there are in principle infinitely many elementary systems of stochastic processes $\psi_\lambda(t)$ that can be used to represent $R(t)$ in a way consistent with the FD theorem.

Appendix B: Representation of thermal forces in the presence of fluid inertia

In this Appendix the application of the classical FD theorem in the presence of fluid inertia is discussed. In this case, for large ω (see Sec. V),

$$\mathrm{Re}[\zeta(\omega)] \sim \omega^{1/2} , \tag{B1}$$

which implies that $\langle R(t)\, R(0) \rangle \sim t^{-3/2}$ at short timescales. Therefore, without loss of generality let us assume that

$$\langle R(t)\, R(0) \rangle = \frac{C}{t^{3/2}} , \tag{B2}$$

where C is a constant, for fixed T, the actual value of which is inessential in the present analysis. Also, in this case $R(t)$ admits a modal representation in the form of eq. (A11), say $R(t) = \int_0^\infty \sqrt{b(\lambda)}\, \psi_\lambda(t)\, d\lambda$, where $b(\lambda)$ is the solution of the functional equation

$$\int_0^\infty b(\lambda)\, e^{-\lambda\, t}\, d\lambda = \frac{C}{t^{3/2}} \tag{B3}$$

indicating that $b(\lambda)$ is the inverse Laplace transform of the correlation function $\langle R(t)\, R(0) \rangle$ in which time t plays the role of the Laplace variable. Using known results of the theory of Laplace transforms, eq. (B3) admits the solution

$$b(\lambda) = \frac{2\, C}{\sqrt{\pi}} \sqrt{\lambda} \, . \tag{B4}$$

Consider $\langle R^2(t) \rangle$. From the above relations, it follows that

$$\langle R^2(t) \rangle = \frac{4\, C^2}{\pi} \int_0^\infty \lambda^{1/4}\, d\lambda \int_0^\infty \mu^{1/4}\, \langle \psi_\lambda(t)\, \psi_\mu(t) \rangle\, d\mu = \frac{4\, C^2}{\pi} \int_0^\infty \lambda^{1/2}\, d\lambda = \infty \, . \tag{B5}$$

Therefore, the second-order moment of the thermal force is unbounded, which is indeed a highly singular and unpleasent property. One could argue that this is also the case for the distributional derivative $\xi(t) = dw(t)/dt$ of a Wiener process, modelling the thermal forces in the presence of a purely instantaneous Stokesian friction. In the latter case, however, the infinitesimal increments

$$dF(t) = R(t)\, dt \tag{B6}$$

over time dt possess bounded second-order moments $\langle dF^2(t) \rangle \sim dt$, proportional to dt. It is therefore interesting to evaluate this quantity, in the case of the process defined by eq. (B4). Introduce the new process $q_\lambda(t)$, defined by the relation

$$dq_\lambda(t) = \psi_\lambda(t)\, dt \quad \Rightarrow \quad q_\lambda(t) = \int_0^t \psi_\lambda(\tau)\, d\tau \tag{B7}$$

where, conventionally, $q_\lambda(0) = 0$. Thus

$$dF(t) = \int_0^\infty \sqrt{b(\lambda)}\, dq_\lambda(t)\, d\lambda = \int_0^\infty \sqrt{b(\lambda)} \int_t^{t+dt} \psi_\lambda(\tau)\, d\tau\,. \tag{B8}$$

Therefore $\langle dF^2(t)\rangle$ can be evaluated to

$$\begin{aligned}
\langle dF^2(t)\rangle &= \int_0^\infty \sqrt{b(\lambda)}\, d\lambda \int_0^\infty \sqrt{b(\mu)}\, d\mu \int_t^{t+dt} d\tau \int_t^{t+dt} \langle \psi_\lambda(\tau)\, \psi_\mu(\theta)\, d\theta \\
&= \int_0^\infty b(\lambda)\, d\lambda \int_t^{t+dt} d\tau \int_t^{t+dt} e^{-\lambda|\tau-\theta|}\, d\theta = 2\int_0^\infty b(\lambda)\, d\lambda \int_t^{t+dt} d\tau \int_t^{\tau} e^{-\lambda(\tau-\theta)} d\theta \\
&= 2\int_0^\infty \frac{b(\lambda)}{\lambda}\, d\lambda \left[\int_t^{t+dt} d\tau - e^{\lambda t} \int_t^{t+dt} e^{-\lambda \tau}\, d\tau \right] \\
&= 2\int_0^\infty \frac{b(\lambda)}{\lambda}\, d\lambda\, dt + 2\int_0^\infty \frac{b(\lambda)}{\lambda^2} \left(e^{-\lambda\, dt} - 1\right) d\lambda = 2\int_0^\infty b(\lambda) d\lambda\, dt^2\,.
\end{aligned} \tag{B9}$$

Consequently, due to the singular behavior of $b(\lambda)$ at infinity, see eq. (B4), $\langle dF^2(t)\rangle = \infty$ for any dt. This essentially implies that either $R(t)$ or $dF(t)$ admit a purely formal modal representation in terms of elementary stochastic processes which, hovewer, does not correspond to any physically realizable stochastic evolution.

Appendix C: Application to microfluidics and transport in confined geometries

In this paper we have mainly considered the description of $\mathbf{v}_s(t)$ in the free space in the absence either of external deterministic accelerations $\mathbf{a}(\mathbf{x})$ or of position-dependent hydrodynamic effects, determining the explicit dependence of the linear functionals L_d and L_i on $\mathbf{x}$. The presence of external position-dependent fields generating $\mathbf{a}(\mathbf{x})$ does not present any further problem, as the statistical structure of the hydrodynamic/thermal stochastic velocity $\mathbf{v}_s(t)$ is independent of $\mathbf{a}(\mathbf{x})$. This case has been briefly addressed in Subsec. VI A, in order to highlight the importance of a continuous velocity distribution for $\mathbf{v}_s(t)$. In the case of position-dependent hydrodynamic interactions, which occurs for particles in microchannels of transversal lengthscale comparable to the particle diameter, the statistical properties of the stochastic velocity field $\mathbf{v}_s(t;\mathbf{x})$ can be derived, for fixed $\mathbf{x}$, from the conditional normalized correlation functions $\mathbf{c}^{(k)}(t\,|\mathbf{x})$ defined in Sec. III and satisfying eqs. (16).

What makes these problems peculiar with respect to the free space case is that it is no longer possible, due to the generic nonlinear dependence of $\mathbf{a}(\mathbf{x})$, L_d and L_i on $\mathbf{x}$, to derive a priori the velocity correlation function, nor the statistical characterization of the spatial particle distribution, even in the long-term limit (if an asymptotic equilibrium distribution emerges in the system). This is the case whenever hydrodynamic effects (convective fluxes) cope with potential contributions (conservative forces) so that the resulting acceleration field $\mathbf{a}(\mathbf{x})$ possesses a full Helmholtz decomposition,

$$\mathbf{a}(\mathbf{x}) = \nabla \phi_a(\mathbf{x}) + \nabla \times \mathbf{K}_a(\mathbf{x}) \; , \tag{C1}$$

where the scalar, $\phi_a(\mathbf{x})$, and vector, $\mathbf{K}_a(\mathbf{x})$, potential are both different from zero. In these cases, the understanding of the emergent statistical properties relies on the direct simulation of particle dynamics. In the velocity split approach, this implies the simulation of the stochastic differential equations

$$\begin{aligned} \dot{\mathbf{x}}(t) &= \mathbf{v}_d(t) + \mathbf{v}_s(t, \mathbf{x}(t)) \\ \dot{\mathbf{v}}_d(t) &= L_d[\mathbf{v}_d(t); \mathbf{x}(t)] + L_i[\dot{\mathbf{v}}_d(t); \mathbf{x}(t)] + \mathbf{a}(\mathbf{x}(t)) \; , \end{aligned} \tag{C2}$$

where the choice of the simplest and most efficient representation of the stochastic field $\mathbf{v}_s(t, \mathbf{x})$, e.g., via a decomposition of it into elementary EPK processes, see eq. (40), becomes essential. Due to hydrodynamic confinement, the parameters a_h, b_k, ξ_h, $\tau_{0,h}$ and λ_k entering eq. (40) do depend on the position $\mathbf{x}$. Substituting the expansion eq. (40) into eq. (C2) leads to stochastic differential equations that are formally similar to nonlinear Langevin equations [59]. With respect to the Wiener-Langevin counterparts, they do not suffer all the troubles of the Wiener singularity (lack of bounded variation) in the definition of the stochastic integrals, forcing the choice of a stochastic calculus (Ito, Stratonovich, Hänggi-Klimontovich) by causing the so-called Ito-Stratonovich dilemma [60]. Nevertheless, their emergent properties could be far from trivial and could lead to interesting new phenomena. To clarify this issue, consider the simplest but physically meaningful example of a micrometric particle moving in a still liquid at constant temperature T close to an infinite wall, subjected to gravity and to a repulsive double layer Debye potential from the wall, comprehensively described by the potential $\phi(\mathbf{x})$. This problem has been analyzed experimentally in [61, 62]. Indicate with x the distance of the particle from the wall so that $\phi = \phi(x)$ and, according to eqs. (7), (8a),(40),

$$\begin{aligned} \dot{x} &= v_d + \sum_{h=1}^{N_\Lambda} a_h(x)\, \Lambda_g(t, \xi_h(x), \tau_{0,h}(x); v) + \sum_{k=1}^{N_\Xi} b_k(x)\, \Xi_g(t, \lambda_k(x); v) \\ \dot{v}_d &= L_d[v_d; x] + L_i[\dot{v}_d; x] - \frac{1}{m} \partial_x \phi_x(x) \; . \end{aligned} \tag{C3}$$

Equation (C3) represents the prototype of a stochastic problem, i.e., the formulation of a detailed hydrodynamic description of fluid/particle interactions, here by using the velocity splitting approach. In the overdamped case, it is known that the equilibrium probability distribution $p^*(x)$ is the classical Boltzmannian density, $p^*(x) = Ae^{-\phi(x)/k_BT}$. But is this still true if the fluid inertial contributions expressed by the Basset long-range force are accounted for? Albeit it is likely to be the case, a conclusive answer to this problem has not yet been given. A violation of the Boltzmannian behaviour controlled by the potential $\phi(x)$, i.e., a stationary density $p^*(x) = Ae^{-\psi(x)/k_BT}$ with $\psi(x) \neq \phi(x)$, would imply the emergence of fluctuational forces defined by the potential $\psi(x) - \phi(x)$ deriving from confinement and hydrodynamic effects, conceptually analogous to the Casimir forces between metallic plates [63]. This, and even more interesting problems (arising from coupling effects whenever the tensorial structure of the hydrodynamic resistance and inertial term is accounted for [32, 33]) are posed by microfluidic applications to fundamental statistical physics, once the hydrodynamic/thermal fluctuations are included in detail. The use of the velocity splitting approach, leading to stochastic models of the form of eq. (C2), represents a feasible way to analyze them, at least via direct stochastic simulations.

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[1] A. Einstein, Investigations on the theory of the Brownian movement (Dover, New York, 1956).

[2] H. B. Callen and T. A. Welton, Phys. Rev. **83** 34 (1951).

[3] R. Kubo, Rep. Prog. Phys. **29** 235 (1966).

[4] P. Langevin, C. R. Acad. Sci. (Paris) **146** 530 (1908).

[5] R. Kubo, M. Toda and N. Hashitsume, Statistical Physics II - Nonequilibrium Statistical Mechanics (Springer-Verlag, Berlin, 1991).

[6] R. Zwanzig, Nonequilibrium Statistical Mechanics (Oxford Univ. Press, Oxford, 2001).

[7] J. Tothova and V. Lisy, Phys. Lett A **380** 2561 (2016).

[8] A. Jannasch, M. Mahamdeh and E. Schäffer, Phys. Rev. Lett. **107** 228301 (2011).

[9] T. Franosch, M. Grimm, M. Belushkin, F. M. Mor, G. Foffi, L. Forró and S. Jeney, Nature 478 85 (2011).

[10] S. Kheifets, A. Simha, K. Melin, T. Li and M. G. Raizen, Science **343** 1493 (2014).

[11] L. D. Landau and E. M. Lifshitz, Statistical Physics (Pergamon Press, Oxford, 1980).

[12] A. Widom, Phys. Rev. A **3** 1394 (1971).

[13] M. R. Maxey and J. J. Riley, Phys. Fluids **26** 883 (1983).

[14] C. Darwin, Math. Proc. Cambridge Phil. Soc. **49** 342 (1953).

[15] A. Cairoli, R. Klages, and A. Baule, PNAS **22** 5714 (2018).

[16] N. Wiener, Proc. Natl. Acad. Sci. **7** 253 (1921).

[17] N. Wiener, Proc. Natl. Acad. Sci. **7** 294 (1921).

[18] K. Ito, Stochastic Processes (Springer Verlag, New York, 2004).

[19] R. Feynman and A. R. Hibbs, Quantum Mechanics and Path Integrals (McGraw-Hill Comp., New York, 1965).

[20] J. Zinn-Justin, Quantum Field Theory and Critical Phenomena (Clarendon Press, Oxford, 2002).

[21] T. Vicsek Fractal Growth Phenomena (World Scientific, Singapore, 1992).

[22] S. Kim and S. J. Karrila Microhydrodynamics (Dover Publ. New York, 2005).

[23] M. Kac, Rocky Mountain J. Math. **4** 497 (1974).

[24] I. Bena, Int. J. Mod. Phys. B **20** 2825 (2006).

[25] M. Giona, A. Brasiello and S. Crescitelli, J. Phys. A **50**, 335002 (2017).

[26] M. Giona, A. Brasiello and S. Crescitelli, J. Phys. A **50**, 335003 (2017).

[27] M. Giona, A. Brasiello and S. Crescitelli, J. Phys. A **50**, 335004 (2017).

[28] V. Zaburdaev, S. Denisov and J. Klafter, Rev. Mod. Phys. **87** 483 (2015).

[29] M. Giona, A. Cairoli and R. Klages, Phys. Rev. X **12** 021004 (2022). (2020).

[30] T. Li and M. G. Raizen, Ann. Phys. (Berlin) **525** 281 (2013).

[31] J. Mo and M. G. Raizen, Annu. Rev. Fluid Mech. **51** 403 (2019).

[32] J. Happel and H. Brenner, Low Reynolds number hydrodynamics: with special applications to particulate media (Springer Science & Business Media, New York, 2012).

[33] G. Procopio and M. Giona, Fluids **7** 105 (2022).

[34] C. Venditti, A. Adrover and M. Giona, Physica A **585** 126407 (2022).

[35] R. Livi and P. Politi, Nonequilibrium Statistical Physics (Cambridge Univ. Press, Cambridge, 2017).

[36] L. E. Reichl, A Modern Course in Statistical Physics (Wiley-VCH, Weinheim, 2009).

[37] J. Tothova, G. Vaszlova, L. Glod and v. Lisy, Eur. J. Phys. **32** 645 (2011).

[38] J. Tothova, G. Vaszlova, L. Glod and v. Lisy, Eur. J. Phys. **32** L47 (2011).

[39] V. Vladimirsky, Z. Eksp. Teor. Fiz. (russian) **12** 199 (1942).

[40] R. Huang, I. Chavez, K. M. Taute, B. Lukic, S. Jeney, M. G. Raizen and E.-L. Florin, Nature Physics **7** 576 (2011).

[41] J. Mo, A. Simha, S. Kheifets and M. G. Raizen, Optics Express **23** 1888 (2015).

[42] V. Vladimirsky and Y. A. Terletzky, Zh. Eksp. Theor. Fiz. **15** 258 (1945).

[43] B. J. Alder and T. E. Wainwright, Phys. Rev. Lett. **18** 988 (1967).

[44] B. J. Alder and T. E. Wainwright, Phys. Rev. A **1** 18 (1970).

[45] E. J. Hinch, J. Fluid Mech. **72** 499 (1975).

[46] H. J. H. Clercx and P. P. J. M. Schram, Phys. Rev. A **46** 1942 (1992).

[47] R. Huang, I. Chavez, K. M. Taute, B. Lukic, S. Jeney, M. G. Raizen and E.-L. Florin, Nature Physics **7** 576 (2011).

[48] M. Grimm, S. Jeney, S. and T. Franosch, Soft Matter **7** 2076 (2011).

[49] M. Grimm, T. Franosch, and Jeney, Phys. Rev. E **86** 021912 (2012).

[50] M. G. Raizen and T. Li, Physics Today **68** 56 (2015).

[51] E. H. Hauge and A. Martin-Löf, J. Stat. Phys. **7** 259 (1973).

[52] D. Bedeaux and P. Mazur, Physica **76** 247 (1974).

[53] J. M. Ortiz de Zárate and J. V. Sengers, Hydrodynamic Fluctuations in Fluids and Fluid Mixtures (Elsevier, Amsterdam, 2006).

[54] J. Dunkel and P. Hänggi, Phys. Rep. **471** 1 (2009).

[55] M. Giona, EPL **126** 50001 (2019).

[56] S. Fedotov, Phys. Rev. E **93** 020101 (2016).

[57] R. Zwanzig, Annu. Rev. Phys. Chem. **102** 16 (1965).

[58] J. G. Kirkwood, Selected Topics in Statistical Mechanics (Gordon and Breach, New York, 1967).

[59] W. T. Coffey and Y. P. Kalmykov, The Langevin Equation (World Scientific, Singapore, 2012).

[60] N. G. van Kampen, J. Stat. Phys. **24** 175 (1981).

[61] G. Volpe, L. Helden, T. Brettschneider, J. Wehr and C. Bechinger, Phys. Rev. Lett. **104** 170602 (2010).

[62] T. Brettschneider, G. Volpe, L. Helden, J. Wehr and C. Bechinger, Phys. Rev. E **83** 041113 (2011).

[63] W. M. R. Simpson and U. Leonhardt, Forces of the Quantum Vacuum (World Sci., Singapore,

2015).

[64] C. Rovelli, J. Am. Philosoph. Ass. **1** 23 (2015).