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Spherical particle Brownian motion in viscous medium as non-Markovian random process

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ABSTRACT

The Brownian motion of a spherical particle in an infinite medium is described by the conventional methods and integral transforms considering the entrainment of surrounding particles of the medium by the Brownian particle. It is demonstrated that fluctuations of the Brownian particle velocity represent a non-Markovian random process. The features of Brownian motion in short time intervals and in small displacements are considered.

1. Introduction

To describe the random physical processes, the stochastic differential equations are conventionally used. This approach leads to Markovian character of the physical values fluctuations. Such method allows the researchers to take advantage of well-developed theory of stochastic differential systems [1–3] which determines all necessary statistical characteristics of the random changes of the physical values. However the Markovian model is approximate. The actual processes proceeding in physical and technical systems are often non-Markovian ones [4]. In particular, when a Markovian random process acts on a dynamic system, its response represents a non-Markovian random process. The processes observed after integration of a Markovian process or finding of a sliding average of the process with independent values are also non-Markovian ones. For example, the coordinate of the Brownian particle calculated as an integral of its velocity is not always described by the model of a random Markovian process, and the Wiener approximation is correct for the Brownian particles only for sufficiently long time intervals much longer than the particle relaxation time.

Typical non-Markovian process observed in various physical, technical, social and biological systems is so-called $1/f$ noise (or flicker noise). For example, fluctuations of the kinetic coefficients, such as the electric conductivity, noise of the electronic devices limiting their sensitivity, changes of a human brain activity have character of $1/f$ noise [5–8]. Impulse fluctuations of one-dimensional Brownian motion in an infinite medium and temperature fluctuations of one-dimensional thermal distribution are $1/f$ noise [9].

The above-indicated reasons demonstrate that we practically always use non-Markovian random processes to describe actual physical processes and technical devices. Models of the Markovian process can be considered only as a first approximation.

Non-Markovian processes description is suggested to realize with help of the integral transforms instead the differential operators. The problem of linear integral transform of arbitrary random processes was solved using the characteristic functionals which is more general characteristics than multidimensional characteristic functions [10,11]. The nonlinear transform of random processes was regarded in works [12,13]. The characteristic functional of generalized Wiener process was found in [14,15] based on the theory of infinitely divisible distributions [16]. The generalized Langevin equation is described in [17,18].

In this work the description of Brownian motion specified by linear integral transforms is given. Movement of Brownian particle in small displacements is considered. General expressions for one-dimensional and multidimensional characteristic functions are found.

2. Brownian motion of spherical particle

Let us consider Brownian motion of the spherical particle of mass M and radius R in the medium with kinematic viscosity ν and density ρ. The dynamical equation of motion has the form

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$$M\frac{dV(t)}{dt} = F_0(t) + F_r(t) + \xi_V(t), \tag{1}$$

where $V(t)$ is the particle velocity, $F_r(t)$ is the resistance force, $F_0(t)$ is the sum of other external forces, and $\xi_V(t)$ is a random force. The resistance force $F_r(t) = F_{r0}(t)$ for the linear viscous medium is conventionally written as

$$F_{r0}(t) = -\gamma V(t), \tag{2}$$

where γ is the viscous friction coefficient which for the spherical particle can be written as $\gamma = 6\pi\rho\nu R$.

A mean square of the Brownian particle displacement $\langle X^2(t)\rangle$ is defined by the Einstein formula

$$\langle X^2(t)\rangle = 2D_p t, \tag{3}$$

where $D_p = \frac{kT}{\gamma}$ is a diffusion coefficient.

To describe the motion of a spherical Brownian particle with allowance for the entrainment of particles of the medium, expression (see problem 7 in Section 24 of [19])

$$F_r(t) = -2\pi\rho R^3\left[\frac{3\nu}{R^2}V(t) + \frac{1}{3}\frac{dV(t)}{dt} + \frac{3}{R}\sqrt{\frac{\nu}{\pi}}\int_0^t \frac{dV(\tau)}{d\tau}\frac{d\tau}{\sqrt{t-\tau}}\right] \tag{4}$$

should be used instead of (2). The particle motion in Eq. (4) started at time $t = 0$. Formula (4) is used in [20–22] for the description of Brownian motion of a particle in the viscous medium.

Substituting Eq. (4) into Eq. (1), we derive the equation of the motion of the spherical particle:

$$\left(M + \frac{2}{3}\pi\rho R^3\right)\frac{dV(t)}{dt} + 6\pi\rho\nu R V(t) + 6\rho R^2\sqrt{\pi\nu}\int_0^t \frac{dV(\tau)}{d\tau}\frac{d\tau}{\sqrt{t-\tau}} = F_0(t) + \xi_V(t), \tag{5}$$

or

$$Z(t) + A\int_0^t \left(1 + \frac{B}{\sqrt{t-\tau}}\right)Z(\tau)\,d\tau = \tilde{\xi}(t), \tag{6}$$

where

$$Z(t) = \frac{dV(t)}{dt}, \tag{7}$$

$$A = \frac{6\pi\rho\nu R}{M + \frac{2}{3}\pi\rho R^3}, \qquad B = R\sqrt{\frac{1}{\pi\nu}},$$

$$\tilde{\xi}(t) = \frac{\xi(t)}{M + \frac{2}{3}\pi\rho R^3}, \tag{8}$$

and we consider that $F_0(t) = 0$.

Integral equation (6) presents the specific case of hereditary stochastic systems. General methods for these equations are described in Section 7.8.3 of [2].

3. Brownian motion in short time intervals

After Laplace transform of expression (6) we obtain

$$Mp^2\hat{X}(p) + \chi(p)p^2\hat{X}(p) = \hat{\xi}_V(p), \tag{9}$$

where

$$\chi(p) = \frac{4}{3}\pi\rho R^3 + \frac{6\pi\rho R^2\sqrt{\nu}}{\sqrt{p}} + \frac{6\pi\rho\nu R}{p}, \tag{10}$$

and p is a transform parameter, $\hat{X}(p) = \widehat{V}(p)/p$ and $\hat{\xi}_V(p)$ are Laplace transforms of the Brownian particle coordinate $X(t)$ and random force $\xi_V(t)$. From Eq. (9) we find

$$\hat{X}(p) = \frac{\hat{\xi}_V(p)}{p^2(M + \chi(p))}. \tag{11}$$

This expression allows writing the formula for coordinate $X(t)$ on form of convolution

$$X(t) = \int_0^t G_X(t-\tau)\xi_V(\tau)\,d\tau, \tag{12}$$

where for Laplace transform $G_X(t)$ we have

$$\hat{G}_X(p) = \frac{1}{p^2(M + \chi(p))}. \tag{13}$$

One-dimensional $g_1(\lambda; t)$ and L-dimensional $g_L(\lambda_1, \ldots, \lambda_L; t_1, \ldots, t_L)$ characteristic functions which correspond to the transform (13) have the form of [23]

$$g_1(\lambda; t) = \exp\left[-\frac{1}{2}\nu_w\lambda^2\int_0^t G_X^2(t-\tau)\,d\tau\right], \tag{14}$$

$$g_L(\lambda_1, \ldots, \lambda_L; t_1, \ldots, t_L) = \exp\left[-\frac{1}{2}\nu_w\sum_{l,k=1}^{L}\lambda_l\lambda_k\int_0^{\min(t_l,t_k)} G_X(t_l-\tau)G_X(t_k-\tau)\,d\tau\right]. \tag{15}$$

Here the intensity of random force ν_w is defined by formula

$$\nu_w = 2\gamma kT. \tag{16}$$

From (11)–(13) we find

$$\hat{G}(p) = \frac{1}{ap(p + b\sqrt{p} + c)}, \tag{17}$$

where

$$a = M + \frac{4}{3}\pi\rho R^3, \qquad b = \frac{6\pi\rho R^2\sqrt{\nu}}{a}, \qquad c = \frac{6\pi\rho\nu R}{a}. \tag{18}$$

Eq. (17) generally does not allow finding the Laplace inversion. However, if $b > 2\sqrt{c}$ expression (17) is presented by formula

$$\frac{1}{ap(p + b\sqrt{p} + c)} = \frac{1}{a}\left(\frac{A}{p} + \frac{B}{\sqrt{p}} + \frac{C}{\sqrt{p} - x_1} + \frac{D}{\sqrt{p} - x_2}\right), \tag{19}$$

where

$$x_1 = \frac{1}{2}\left(-b + \sqrt{b^2 - 4c}\right), \qquad x_2 = \frac{1}{2}\left(-b - \sqrt{b^2 - 4c}\right),$$

$$A = \frac{1}{c}, \qquad B = -\frac{b}{c^2}, \qquad C = \frac{b^2 + b\sqrt{b^2 - 4c} - 2c}{2c^2\sqrt{b^2 - 4c}},$$

$$D = \frac{-b^2 + b\sqrt{b^2 - 4c} + 2c}{2c^2\sqrt{b^2 - 4c}}.$$

Taking notice that necessary condition $b > 2\sqrt{c}$ is occurred for the wide range of particles and media corresponding to the actual conditions. Particularly, for the Brownian particle with radius $R = 1$ μm moving in water (density $\rho = 1000$ kg/m^3, kinematic viscosity $\nu = 10^{-6}$ m^2/s) the inequality $b > 2\sqrt{c}$ is occurred if a mass of the particle $M < 5.2 \cdot 10^{-16}$ kg (light porous particles). In the sequel in the capacity of a test particle (meaning following comparison with experiments) we will consider the motion in water of the Brownian particle with density $\rho_0 = 100$ kg/m^3 and radius $R = 0.1$ μm.

Laplace inversion of function (17) considering (19) leads to an analytical formula for the transform kernel $G_X(t-\tau)$:

$$G_X(t-\tau) = \frac{1}{a}\left[A + \frac{B+C+D}{\sqrt{\pi(t-\tau)}} + x_1 \exp\bigl(x_1^2(t-\tau)\bigr)\,\mathrm{erfc}(-x_1\sqrt{t-\tau}) + x_2 D \exp\bigl(x_2^2(t-\tau)\bigr)\,\mathrm{erfc}(-x_2\sqrt{t-\tau})\right]. \tag{20}$$

Here $\mathrm{erfc}(x)$ is an additional error integral. From definition of variables B, C and D the sum $B + C + D = 0$ follows. Then

$$G_X(t-\tau) = \frac{1}{a}\bigl[A + x_1 \exp\bigl(x_1^2(t-\tau)\bigr)\,\mathrm{erfc}(-x_1\sqrt{t-\tau}) + x_2 D \exp\bigl(x_2^2(t-\tau)\bigr)\,\mathrm{erfc}(-x_2\sqrt{t-\tau})\bigr]. \tag{21}$$

Expression (21) principally makes it possible to find a mean square of the Brownian particle displacement $\langle X^2(t)\rangle$ by formula

$$\bigl\langle X^2(t)\bigr\rangle = \left.\frac{\partial^2 g_1(\lambda;t)}{\partial(i\lambda)^2}\right|_{\lambda=0}, \tag{22}$$

where one-dimensional characteristic function $g_1(\lambda;t)$ defined by Eq. (14).

If $t - \tau \gg 0$ the function $\mathrm{erfc}(x)$ decreases faster increasing the function $\exp(x^2)$, and the kernel $G_X(t-\tau)$is equal to constant (see Eq. (21)):

$$G_X(t-\tau)|_{t-\tau\to\infty} = \frac{A}{a}. \tag{23}$$

In this case from expression (22) we have

$$\bigl\langle X^2(t)\bigr\rangle\big|_{t\to\infty} = \nu_w \frac{A^2}{a^2} t = \nu_w \frac{1}{a^2 c^2} t = \frac{2\gamma k T}{\gamma^2} t = \frac{2kT}{\gamma} t = 2D_p t, \tag{24}$$

which is well-known Einstein formula (3).

For approximate finding of the value $\langle X^2(t)\rangle$ in short time intervals (about $10^{-3} - 10^{-2}$ s) in expression (21) functions $\exp(x)$ and $\mathrm{erfc}(x)$ are developed in series. In view of the fact that values of x_1 and x_2 are generally large (for the testing particle $\sim 10^4$ s$^{-1/2}$) substantial number of series terms of functions $\exp(x)$ and $\mathrm{erfc}(x)$ it is necessary to keep. Keeping 30 terms we find that for $t - \tau < 10^{-3}$ s the function $G_X(t-\tau)$ is approximated by formula

$$G_X(t-\tau)|_{t-\tau\to 0} = \frac{3.5 \cdot 10^{-8}(t-\tau)^{0.5}}{a}. \tag{25}$$

The use of the finding approximating formula for the kernel $G_X(t-\tau)$ allows after substitution Eq. (25) to Eq. (14) and application of Eq. (22) to find for small t mean square of Brownian particle displacement:

$$\bigl\langle X^2(t)\bigr\rangle\big|_{t\to 0} = 2D\alpha t^2, \tag{26}$$

where a parameter $\alpha \approx 210$ s^{-1}.

4. Comparison with experiment

Experimental research [24,25] of the movement of the Brownian particle (with parameters similar to testing particle) in viscous medium in short time intervals and in small displacements displays that for time intervals about 10^{-3}–10^{-2} s the mean square of displacement is good described by derived above formula (26) (in experiments with particle moving in water parameter $\alpha = 180$ s^{-1}), and in long time intervals (more than 10^{-1} s) classical formula (3) is satisfactory that from considered above theory also follows. In addition, a computational modeling of Brownian motion with the resistance force (4) [26] also leads to a ballistic character of movement in the field of short time intervals that validates calculus in Section 3.

Good coordination theoretical results based on Eq. (4) for the resisting force with experiments in short and long intervals indicates to legality of the use of expression (4) considering interaction of the Brownian particle and surrounding it medium particles that leads to their entrainment by the Brownian particle.

5. Resume

The above description of Brownian motion of the spherical particle in the infinite viscous medium by random non-Markovian process demonstrates its significant difference from the process investigated by the classical method, especially in the field of short time intervals of the Brownian particle movement.

The results obtained can be important for consideration of various random processes for which the model of Brownian motion is applicable.

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