FOKKER–PLANCK EQUATIONS WITH MEMORY: THE CROSS OVER FROM BALLISTIC TO DIFFUSIVE PROCESSES

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PACS 05.40.-a

The unified description of diffusion processes that crosses over from a ballistic behavior at short times to a fractional diffusion (sub- or superdiffusion), as well as to the ordinary diffusion at longer times, is proposed on the basis of a non-Markovian generalization of the Fokker–Planck equation. The relations between the non-Markovian kinetic coefficients and observable quantities (mean- and mean square displacements) are established. The problem of calculations of the kinetic coefficients using the Langevin equations is discussed. Solutions of the non-Markovian equation describing diffusive processes in the real (coordinate) space are obtained. For long times, such a solution agrees with results obtained within the continuous random walk theory but is much superior to this solution at shorter times, where the effect of the ballistic region is crucial.

1. Introduction

Molecular diffusion is the process of molecular spreading from regions of high concentrations to regions with low concentrations. The history of the investigation of the particle diffusion process covers two centuries (details can be found in [1]). The quantitative description of the diffusion was developed by Fick (in 1855) and is based on two points. The diffusive flux of a substance through the small area during a short time interval was defined by an empirical relation to the gradient of the particle number density in a similar way as in the case of thermo-conductivity (Fourier, in 1822) and electro-conductivity (Ohm, in 1827). This relation is known as the Fick's first law and reads

$$\mathbf{\Gamma}(\mathbf{r},t) = -D\nabla n(\mathbf{r},t), \tag{1}$$

where $n(\mathbf{r},t)$ is the particle number density, $\mathbf{\Gamma}(\mathbf{r},t)$ is the flux, D is the diffusion coefficient which depends on a substance, t is the time variable, $\mathbf{r}$ is the spatial variable, and ∇ is the nabla-operator. As the next step, the continuity equation (which reflects the particle number conservation) relating the flux and the concentration was used:

$$\frac{\partial n(\mathbf{r},t)}{\partial t} = -\nabla\mathbf{\Gamma}(\mathbf{r},t), \tag{2}$$

Substitution of Eq. (1) to Eq. (2) yields the diffusion equation known also as the Fick's second law:

$$\frac{\partial n(\mathbf{r},t)}{\partial t} = D\nabla^2 n(\mathbf{r},t). \tag{3}$$

This approach to solving the diffusion problem is a phenomenological one. The microscopic theory of the diffusion was developed in [2]. There were used the assumptions that the movement of a particle can be treated as a random walk independent of the motions of all other particles. In this case, the change of the particle number density in a one-dimensional system in the time interval τ is defined by;

$$n(x,t+\tau) = \int\limits_{-\infty}^{\infty} P_\tau(\Delta x) n(x-\Delta x,t)\mathrm{d}\Delta x, \tag{4}$$

where Δx is the particle position displacement in a time interval τ, and $P_\tau(\Delta x)$ is the probability distribution function for this displacement. Particle movements can be considered in a time interval large enough to neglect time self-correlations (in fact, the definition of the Markov process), but small in comparison with the time of the observation. Actually, the following double inequality should be satisfied:

$$\tau_{\mathrm{cor}} \ll \tau \ll \tau_{\mathrm{rel}}, \tag{5}$$

where τ_{cor} is the correlation time of the forces producing the random particle motion, and τ_{rel} is the characteristic time of a change of the concentration field. Assuming that the correlation time is vary small and that the significant displacement probability exists only for small Δx, one can expand the particle number density in Eq. (4) in powers of τ (left side) and Δx (right side), so that this equation is reduced to the following differential form:

$$\frac{\partial n(x,t)}{\partial t} = -u\frac{\partial n(x,t)}{\partial x} + D\frac{\partial^2 n(x,t)}{\partial x^2}, \quad (6)$$

where

$$u = \frac{1}{\tau}\int_{-\infty}^{\infty} \Delta x P_\tau(\Delta x) \mathrm{d}\Delta x,$$

$$D = \frac{1}{2\tau}\int_{-\infty}^{\infty} \Delta x^2 P_\tau(\Delta x) \mathrm{d}\Delta x. \quad (7)$$

Notice that the inequality $\tau \ll \tau_{\mathrm{rel}}$ makes it possible to treat Eqs. (7) in the limit $\tau \to 0$ which should be considered in the "macroscopic" (kinetic), but not "microscopic" sense, since τ could not be less than the physically infinitesimal time τ_{ph}, with respect to which the time averaging of microscopic quantities is performed.

In the absence of external fields, $P(\Delta x) = P(-\Delta x)$ and $u \equiv 0$. In this case, Eq. (6) recovers the diffusion equation (3) in the one-dimensional space, and, as follows from Eq. (7), the diffusion coefficient is defined by the mean square displacement of a particle:

$$\langle \Delta x^2 \rangle = 2D\tau. \quad (8)$$

The solution of Eq. (3) for systems with spherical symmetry and the initial condition defined by the Dirac delta-function $n(\mathbf{r}, t=0) = N\delta(\mathbf{r}-\mathbf{r}')$, where N is the particle number, is given by

$$\frac{n(\mathbf{r},t)}{N} = \frac{2}{\Gamma(d/2)(4Dt)^{d/2}}\exp[-(\mathbf{r}-\mathbf{r}')^2/(4Dt)], \quad (9)$$

where d is the space dimension, and $\Gamma(x)$ is the gamma-function. It follows from this solution that it is suitable to introduce the probability function of the particle distribution (pdf) at a given time t:

$$f(\mathbf{r},t) = \frac{n(\mathbf{r},t)}{N}. \quad (10)$$

The variance of this distribution is defined by

$$\langle (\mathbf{r}-r')^2 \rangle = At, \quad (11)$$

with $A = 2Dd$ (for one-dimensional systems, $A = 2D$, and the variance is the same as the mean square displacement in Eq. (8)).

It is easy to see from Eqs. (7) and (8) that the finite value of the diffusion coefficient implies that the displacement probability function should depend also on the time interval τ. The product of two Gaussians is the Gauss function, therefore, it follows from Eq. (9) that Eq. (4) is satisfied for the displacement probability distribution

$$P_\tau(\Delta x, \tau) = \frac{1}{2\sqrt{\pi}}\frac{\exp(-\Delta x^2/(4D\tau))}{\sqrt{D\tau}}. \quad (12)$$

Nevertheless, as was emphasized in [3], relation (11) is not correct for arbitrary short time intervals, and the left inequality in (5) should be valid.

Assumptions used in the development of the Brownian diffusion are very often not satisfied in real flows. There are many experimental evidences that the growth of the variance is described by a more general power law

$$\langle \Delta x^2 \rangle \sim D_\alpha t^\alpha \quad \text{for } t \gg \tau_{\mathrm{cor}}\ , \quad (13)$$

where $\alpha \neq 1$, the angular brackets mean the average over repeated experiments, and D_α is a coefficient with the appropriate dimensionality. It seems that the first example of such anomalous behavior is the Richardson relative diffusion of particles in the turbulent media [4] ($\alpha = 3$). Since the classical work of Hurst [5] on the stochastic discharge of reservoirs and rivers, it is clear that that this long-time non-classical behavior is generic, when the diffusion steps are correlated, with persistence for $\alpha > 1$ and antipersistence for $\alpha < 1$ [6]. The modern list of systems displaying such a behavior is extensive and growing. Some subdiffusive examples ($\alpha < 1$): charge carrier transport in amorphous semiconductors [7], NMR diffusiometry in percolative [8] and porous systems [9], reptation dynamics in polymeric systems [10], transport on fractal geometries [11], diffusion of a scalar tracer under convection [12], macroparticle diffusion in complex liquids [13], *etc.* Superdiffusive ($\alpha > 1$) examples include special domains of a rotating flow [14], collective slip diffusion on solid surfaces [15], bulk-surface exchange controlled dynamics in porous glasses [16], quantum optics [17], turbulent plasmas [18–22] *etc.*

These examples created an urgent need to formulate novel stochastic theories to compute the probability distribution function that is associated with the anomalous diffusion processes of this type. The possible ways to solve this problem and the extension of the pdf behavior to short times are discussed in this paper.

2. Non-Fickian Diffusion

In general, Fick's assumption that the constant diffusion coefficient in Eq. (1) is only a property of a substance is not adequate. For example, in the case of the relative particles diffusion, it was shown from observations that the rate of diffusion increases with the separation of neighbors [4]. The coefficient of diffusion on fractal objects depends on the spatial variables [23]. Therefore, a possible generalization of the first Fick's law employs a spatial dependence of the diffusion coefficient in Eq. (1). Alternatively, it was proposed in [24] that the relative particle diffusion coefficient is time dependent. The same result for the effective diffusion coefficient in the case of the Brownian motion with long-time correlation of the randomly fluctuating force was obtained in [25]. Therefore, Eq. (1) can be rewritten in the general case, by taking Eq. (10) into account as follows:

$$\Gamma(\mathbf{r},t) = -D(\mathbf{r},t)\nabla f(\mathbf{r},t), \tag{14}$$

where the diffusion coefficient is a function of both the spatial and temporal variables. Application of the continuity equation yields the following diffusion equation for the pdf:

$$\frac{\partial f(\mathbf{r},t)}{\partial t} = \nabla D(\mathbf{r},t)\nabla f(\mathbf{r},t). \tag{15}$$

For a particular case [26, 27] where

$$D(\mathbf{r},t) = D_0 t^{\theta_1} r^{\theta_2}, \tag{16}$$

the three-dimensional spherically symmetric solution for a point-source at the center of the coordinate system at $t = 0$ is defined by:

$$f(\mathbf{r},t) = \frac{2-\theta_2}{4\pi^2\Gamma[3/(2-\theta_2)]}\left[\frac{1+\theta_1}{D_0(2-\theta_2)^2 t^{1+\theta_1}}\right]^{\frac{3}{2-\theta_2}} \times$$

$$\times \exp\left[-\frac{(1+\theta_1)r^{2-\theta_2}}{D_0(2-\theta_2)t^{1+\theta_1}}\right]. \tag{17}$$

The variance of the distribution given by Eq. (17) reads:

$$\langle r^2\rangle = \frac{\Gamma[5/(2-\theta_2)]}{\Gamma[3/(2-\theta_2)]}\left[\frac{D_0(2-\theta_2)^2}{1+\theta_1}\right] t^{2(1+\theta_1)/(2-\theta_2)}. \tag{18}$$

One moment can not define a distribution, and one can see from the comparison of Eq. (13) and Eq. (18) that any fixed value of the parameter $\alpha = 2(1+\theta_1)/(2-\theta_2)$ can be obtained in different diffusion models defined by Eq. (16).

The generalization of Eq. (4) to the case of the phase variable $X \equiv (\mathbf{r},\mathbf{v})$ is defined by the following equation for the pdf [28, 29]:

$$f(X,t+\tau) = \int P(X-\Delta X,\Delta X;t;\tau)f(X-\Delta X,t)\mathrm{d}\Delta X, \tag{19}$$

where $P(X,\Delta X;t,\tau)$ is the probability of the particle displacement by ΔX from the phase point X at time t during the time interval τ.

The differential equation corresponding to Eq. (6) in this case is known as the Fokker–Planck equation [28–30] and is defined by:

$$\frac{\partial f(X,t)}{\partial t} = \sum_{i=1}^{2d}\frac{\partial}{\partial X_i}\Bigg[-A_i(X)f(X,t)+$$

$$+\frac{1}{2}\sum_{j=1}^{2d}\frac{\partial}{\partial X_j}B_{i,j}(X)f(X,t)\Bigg], \tag{20}$$

where the right-hand side consists of two contributions from the advective term and the diffusive term, d is the spatial dimension, and the coefficients are given by:

$$A_i(X,t) = \frac{1}{\tau}\int \Delta X\, P(X,\Delta X;t,\tau)\mathrm{d}\Delta X,$$

$$B_{i,j}(X,t) = \frac{1}{\tau}\int \Delta X_i\Delta X_j\, P(X,\Delta X;t,\tau)\mathrm{d}\Delta X. \tag{21}$$

For the isotropic diffusion reduced to the coordinate space, Eq. (20) corresponds to the following flux:

$$\mathbf{\Gamma}(\mathbf{r},t) = \mathbf{u}(\mathbf{r},t)f(\mathbf{r},t) - \nabla D(\mathbf{r},t)f(\mathbf{r},t). \tag{22}$$

If the advection doesn't contribute to the Fokker–Planck generalization of the Fick's law, Eq. (22) is different from the phenomenological one given by Eq. (14) for the space-dependent diffusion coefficient. Respectively, for the spatially inhomogeneous diffusion, these two definitions of a flux yield different diffusion equations (the discussion can be found, e.g., in [31–33]). The two fluxes (and diffusion equations) are the same only for constant or time-dependent diffusion coefficients or under the condition $\mathbf{u}(\mathbf{r},t) = \nabla D(\mathbf{r},t)$. Unfortunately, this assumption is incorrect for the spatial dimension larger than one [33, 34].

The flux defined by Eq. (14) or Eq. (22) corresponds to inhomogeneous media and is local as the Fick's law given by Eq. (1). The merit of these laws consists in the possibility to describe the anomalous time behavior of the mean square displacement and to obtain a simple analytic solution. Unfortunately, the local approach which neglects time correlations suffers from the instantaneous action propagation. In order to overcome this problem, a phenomenological equation for the flux relaxation was proposed in [35]. For a one-dimensional system, it reads

$$\frac{\partial \mathbf{\Gamma}}{\partial t} = \frac{\mathbf{\Gamma}_0 - \mathbf{\Gamma}}{\tau}, \tag{23}$$

where τ is the scale of time, and the flux $\mathbf{\Gamma}_0$ is given by the Fick's law. The solution of this equation is defined by

$$\mathbf{\Gamma} = -\frac{D}{\tau}\int_0^t \exp\left[-\frac{t-t'}{\tau}\right]\frac{\partial f(x,t')}{\partial x}\,\mathrm{d}t'. \tag{24}$$

Application of the continuity equation (2) yields the time nonlocal diffusion integro-differential equation:

$$\frac{\partial f(x,t)}{\partial t} = \frac{D}{\tau}\int_0^t \exp\left[-\frac{t-t'}{\tau}\right]\frac{\partial^2 f(x,t')}{\partial x^2}\,\mathrm{d}t'. \tag{25}$$

The exponential kernel $K(t) = exp(-t/\tau)$ allows us to reduce Eq. (25) by the time differentiation to the telegraph equation:

$$\tau\frac{\partial^2 f(x,t)}{\partial t^2} + \frac{\partial f(x,t)}{\partial t} = D\frac{\partial^2 f(x,t)}{\partial x^2}. \tag{26}$$

In contrast to the parabolic type of local diffusion equations, the telegraph equation is the equation of the hyperbolic type, which opens the possibility to describe the propagation with a finite velocity. In the frame of the heat conduction theory, the equation for a heat flux in the form of Eq. (23) was proposed in [36]. It is known as the Maxwell–Cattaneo model of heat convection (see, e.g., [37]). The telegraph equation follows also from the *persistent* random walk approach to the diffusion problem [38–41].

The solution of the telegraph equation is given in terms of modified Bessel functions of the first kind (see, e.g., [41]) and is defined in a finite domain with boundary expanding with a constant velocity. Details of this solution will be discussed below in Section 5 5.2.

3. Time-Nonlocal Fokker–Planck Equation

A natural generalization of Eq. (23) in the spirit of the local Fokker–Planck flux given by Eq. (22) in the one-dimensional case can be suggested in the following form:

$$\mathbf{\Gamma}(x,t) = \int_0^t \left[-a(t-t')f(x,t') + \frac{1}{2}b(t-t')\frac{\partial f(x,t')}{\partial x}\right]\mathrm{d}t', \tag{27}$$

where kernels $a(t-t')$ and $b(t-t')$ reflect the time-nonlocality of advective and diffusive contributions to the flux. The substitution of Eq. (27) into Eq. (2) which is the conservation law and is valid also in the case under consideration yields a version of the time-nonlocal Fokker–Planck equation [42, 45]:

$$\frac{\partial f(x,t)}{\partial t} = \int_0^t \frac{\partial}{\partial x}\left[-a(t-t')f(x,t') + \frac{1}{2}b(t-t')\frac{\partial f(x,t')}{\partial x}\right]\mathrm{d}t'. \tag{28}$$

The key point here is to find the explicit form of the time-nonlocal kernels in Eq. (28). It is quite natural to express them in terms of the observable quantities, for example, in terms of mean and mean square displacements similar to those in the case of the ordinary Fokker–Planck equation (21).

Supplementing Eq. (28) with the initial condition $f(x,t)|_{t=t'} = \delta(x-x')$ and multiplying Eq. (28) by Δx and Δx^2 ($\Delta x = x - x'$), we obtain, after the integration, the following system of equations for the first and second displacement moments:

$$\frac{\partial \langle\Delta x\rangle_{t-t'}}{\partial t} = \int_{t'}^t a(t-t'')\mathrm{d}t'', \tag{29}$$

$$\frac{\partial \langle\Delta x^2\rangle_{t-t'}}{\partial t} = 2\int_{t'}^t a(t-t'')\langle\Delta x\rangle_{t-t''}\mathrm{d}t'' + \int_{t'}^t b(t-t'')\mathrm{d}t''. \tag{30}$$

Since $a(t,t') = a_0\delta(t-t')$ and $b(t,t') = b_0\delta(t-t')$ in the particular case of the time-local approximation, we have to look for the solutions of Eqs. (29), (30) in the class of generalized functions. The result is

$$a(t-t') = \frac{\partial\langle\Delta x\rangle_{t-t'}}{\partial t}\Big|_{t'\to t}\delta(t-t') + \frac{\partial^2\langle\Delta x\rangle_{t-t'}}{\partial t^2}, \tag{31}$$

$$b(t-t') = \frac{\partial\langle\Delta x^2\rangle_{t-t'}}{\partial t}\Big|_{t'\to t}\delta(t-t') + \frac{\partial^2\langle\Delta x^2\rangle_{t-t'}}{\partial t^2} -$$

$$-2\frac{\partial}{\partial t}\int_{t'}^{t} dt''\frac{\partial\langle\Delta x\rangle_{t-t''}}{\partial t''}\frac{\partial\langle\Delta x\rangle_{t''-t'}}{\partial t'}. \quad (32)$$

These relations can be also written as

$$a(t-t') = \frac{\partial}{\partial t}\left[\theta(t-t')\frac{\partial\langle\Delta x\rangle_{t-t'}}{\partial t}\right], \quad (33)$$

$$b(t-t') = \frac{\partial}{\partial t}\Bigg[\theta(t-t')\Bigg(\frac{\partial\langle\Delta x^2\rangle_{t-t'}}{\partial t} -$$

$$-2\int_{t'}^{t} dt''\frac{\partial\langle\Delta x\rangle_{t-t''}}{\partial t''}\frac{\partial\langle\Delta x\rangle_{t''-t'}}{\partial t'}\Bigg)\Bigg]. \quad (34)$$

These relations make it possible to reproduce the explicit form of the time-nonlocal Fokker–Planck equation from Eq. (28) for the known cases. For the case of the ordinary diffusion in a stationary system, we have

$$\langle\Delta x\rangle_\tau = u\tau \quad \text{and} \quad \langle\Delta x^2\rangle_\tau = D\tau \quad (\tau = t-t'). \quad (35)$$

The substitution of the moments from Eq. (35) to Eq. (33) and Eq. (34) yields

$$a(\tau) = u\delta(\tau) \quad (36)$$

and

$$b(\tau) = D\delta(\tau). \quad (37)$$

After the substitution of kernels from Eqs. (35), (36) into Eq. (28), one obtains the local Fokker–Planck equation (20) with constant coefficients known as the Kramer's equation.

In the recent literature, the problem of a diffusion process which is consistent with Eq. (13) only for all times (i.e. $t_{\rm cor} = 0$) is investigated with the use of the formalism of fractional differential equations (see, e.g., [43]). The moments obtained in the frame of this approach are given by:

$$\langle\Delta x\rangle_\tau = \frac{A_\alpha u\tau}{\Gamma(1+\alpha)},$$

$$\langle\Delta x^2\rangle_\tau = \frac{2A_\alpha^2 u^2\tau^{2\alpha}}{\Gamma(1+\alpha)} + \frac{2D_\alpha\tau^\alpha}{\Gamma(1+\alpha)}. \quad (38)$$

Equations (28), (33), and (34) generate the fractional diffusion-advection equation [44, 46]

$$\frac{\partial f(x,t)}{\partial t} = {}_0\mathbf{D}_t^{1-\alpha}\left(-A_\alpha U\frac{\partial}{\partial x} + D_\alpha\frac{\partial^2}{\partial x^2}\right)f(x,t), \quad (39)$$

where the Riemann–Liouville operator ${}_0\mathbf{D}_t^{1-\alpha}$ is defined by

$${}_0\mathbf{D}_t^{1-\alpha}\phi(x,t) = \frac{1}{\Gamma(\alpha)}\frac{\partial}{\partial t}\int_0^t \mathrm{d}t'\frac{\phi(x,t')}{(t-t')^{1-\alpha}}. \quad (40)$$

At $u = 0$, Eq. (39) reduces to the fractional diffusion equation [43]. These particular examples confirm the self-consistency of Eqs. (28), (33), and (34).

Below, we will neglect the advective term ($\langle\Delta x\rangle_\tau = 0$). The time nonlocal diffusion equation follows from Eq. (28) and reads

$$\frac{\partial f(x,t)}{\partial t} = \int_0^t dt' K(t-t')\frac{\partial^2 f(x,t)}{\partial x^2}, \quad (41)$$

where the kernel is defined by (see Eq. (34)):

$$K(\tau) = \frac{1}{2}\frac{\partial}{\partial\tau}\left[\Theta(\tau)\frac{\partial\langle\Delta x^2\rangle_\tau}{\partial\tau}\right]. \quad (42)$$

The Laplace transformation of this kernel results in

$$K(s) = \frac{s^2\langle\Delta x^2\rangle_s}{2}. \quad (43)$$

Equation (41) is a particular case of the more general equation:

$$\frac{\partial f(x,t)}{\partial t} = \int_0^t K(t-t')\widehat{\mathcal{L}}f(k,t)\mathrm{d}t', \quad (44)$$

where the operator $\widehat{\mathcal{L}}$ acts on the spatial variables. It was shown in [47] that the Laplace transform of the formal solution of Eq. (44) reads

$$\tilde f(x,s) = \frac{1}{\tilde K(s)}\tilde F\left(x, \frac{s}{\tilde K(s)}\right), \quad (45)$$

where $F(x,t)$ is a solution of the auxiliary equation

$$\frac{\partial F(x,t)}{\partial t} = \widehat{\mathcal{L}}F(x,t) \quad (46)$$

with the same initial conditions as those for Eq. (44). In the case under consideration, $f(x, t = 0) = \delta(x -$

$x')$, $\widehat{\mathcal{L}} = \partial^2/\partial x^2$ and, therefore, the function $F(x,t)$ is defined by Eq. (9) with parameters $d = 1$ and $D = 1$. The Laplace transform of this solution of the ordinary diffusion equation reads:

$$\tilde{F}(x,s) = \frac{1}{2\sqrt{s}} \cdot \exp(- \mid \Delta x \mid \sqrt{s}) \; . \tag{47}$$

Substituting in Eq. (45), we find

$$\tilde{f}(x,s) = \frac{1}{\sqrt{2s^3\langle\Delta x^2\rangle_s}} \exp\left(- \mid \Delta x \mid \sqrt{\frac{2}{s\langle\Delta x^2\rangle_s}}\right). \tag{48}$$

Here, Eq. (43) was taken into account in order to replace the Laplace transform $\tilde{K}(s)$ in Eq. (45) with the Laplace transform of the mean square displacement.

4. Calculations of the Displacement Moments

The next important question is where the quantity $\langle\Delta x^2\rangle_t$ can be taken from. One of the ways is to measure the mean square displacements in experiments. On the other hand, they can be calculated on the basis of the microscopic treatment. However, such calculations require to know many details about the system under consideration and can be performed for the specific cases. Therefore, it looks quite reasonable to use the Langevin approach which makes it possible to perform a more general phenomenological treatment.

The Langevin equation [48] (the English translation of this paper is presented in [49]) is Newton's second law applied to a Brownian particle of mass m:

$$\frac{\partial^2 x}{\partial t^2} = -\gamma\frac{\partial x}{\partial t} + R(t), \tag{49}$$

where the first term on the right-hand side describes the action of a friction force (γ is the friction coefficient), and $R(t)$ is the random component uncorrelated with the velocity, and the mean value $\langle R(t)\rangle = 0$. The fluctuation of the random force is defined by:

$$\langle R(t)R(t')\rangle = \frac{k_\mathrm{B}T\gamma}{m}\delta(t-t'). \tag{50}$$

The well-known expression for the mean square displacement which follows from the solution of Eq. (49) reads (in [48], the expression for $\partial\langle\Delta x^2\rangle_t/\partial t$ is presented) :

$$\langle\Delta x^2\rangle_t = \frac{2k_\mathrm{B}T}{m}t_\mathrm{rel}^2\left(\frac{t}{t_\mathrm{rel}} - [1-\mathrm{e}^{-t/t_\mathrm{rel}}]\right), \tag{51}$$

where $t_\mathrm{rel} = m/\gamma$.

Within the Langevin approach, the mean square displacement exhibits the ballistic behavior at short times and the linear dependence on time in the asymptotic region. This property is a direct consequence of the random force fluctuation given by Eq. (50). The Fourier transformation of the delta-function is independent of the frequency. Therefore, in this case, the random force is called the white noise by analogy with the spectrum of light. Nevertheless, as was shown in [52–54], the more general way to use the Langevin approach is to assume the time nonlocality in the definition of the friction force:

$$\frac{\partial^2 x(t)}{\partial t^2} = -\int\limits_0^t \gamma(t-t')\frac{\partial x(t')}{\partial t'}\mathrm{d}t' + R(t). \tag{52}$$

The fluctuation of the random force is related to the kernel in Eq. (52) by the fluctuation-dissipation theorem [53, 54]:

$$\langle R(t)R(t')\rangle = \frac{k_\mathrm{B}T}{m}\gamma(t-t'). \tag{53}$$

In this general case, the Fourier transform of the random force correlation function is the frequency dependent, therefore, this noise is referred as colored (some examples of colored noise can be found in [55]).

The solution of Eq. (52) can be obtained with the use of the Laplace transformation technique, and the calculation of a mean square displacement (see, e.g., [56]) yields

$$\langle\Delta x^2\rangle_t = 2k_\mathrm{B}T\int\limits_0^t H(t')\mathrm{d}t'. \tag{54}$$

As usual, the Maxwell initial velocity distribution was suggested here. In Eq. (54), the function $H(t)$ is the inverse Laplace transform of the following expression:

$$\widetilde{H}(s) = \frac{1}{s(s+\tilde{\gamma}(s))}, \tag{55}$$

where $\tilde{\gamma}(s)$ is the Laplace transform of the kernel in Eq. (52).

The Laplace transform of Eq. (54) with Eq. (55) yields the relation

$$\frac{\langle x^2\rangle_s}{2k_\mathrm{B}T} = \frac{1}{s}\widetilde{H}(s) = \frac{1}{s^2(s+\tilde{\gamma}(s))} =$$

$$= \frac{1}{s^3}(1 - \frac{\tilde{\gamma}(s)}{s} + \ldots). \tag{56}$$

Suppose that, at short times, the mean square displacement can be expanded in a following Taylor series:

$$\langle x^2\rangle_t = \sum_{i=0}^{\infty} a_i t^{\mu_i - 1} \ = a_0 t^2 + a_1 t^3 + a_2 t^4 \cdots \ , \qquad (57)$$

where $\mu_0 = 3$, $\mu_1 = 4$, *etc.* Then the Laplace transform $\langle x^2\rangle_s$ can be written, as $s \to \infty$, in the form [57]

$$\langle x^2\rangle_s = \sum_{i=0}^{\infty} a_i \Gamma(\mu_i) \frac{1}{s^{\mu_i}} = 2a_0 \frac{1}{s^3} + 6a_1 \frac{1}{s^4} + 24a_2 \frac{1}{s^5} \cdots \ . \qquad (58)$$

One can see from the comparison of Eq. (56) and Eq. (58) that the coefficient $a_0 = 1/2$ is independent of the kernel $\gamma(t)$. Therefore, it follows from Eq. (57) that $\langle \Delta x^2\rangle_{t\sim 0} \sim t^2$, i.e., the ballistic short time behavior is a general property of the particle motion in the frame of the generalized Langevin approach.

The long-time behavior of the mean square displacement is defined by the small-s behavior of its Laplace transform [57]. Suppose that $\tilde{\gamma}(s)_{s\sim 0} \sim s^{\alpha-1}$. The substitution of this expression into Eq. (56) yields $\langle x^2\rangle_{s\sim 0} \sim 1/(s^{\alpha+1}(1+s^{\alpha-2}))$. If $\alpha < 2$, we can neglect the second term in brackets ($s^{\alpha-2}_{s\sim 0} \ll 1$). Therefore, applying the inverse Laplace transformation, we get $\langle \Delta x^2\rangle_{t\to\infty} \sim t^{\alpha}$, i.e., the generalized Langevin approach allows us to obtain the nonclassical long-time asymptotics of the mean square displacement.

5. Crossing from Ballistic to Fractional Diffusion with Memory

5.1. General case

The ballistic short-time behavior of the mean square displacement is a general property of the diffusion process independently of its asymptotics. Therefore, it is reasonable to extract this contribution from the general solution in Eq. (48). To find the appropriate general solution (see [58]), we will split $\tilde{f}(x,s)$ into two parts, $\tilde{f}_I(x,s)$ and $\tilde{f}_{\rm II}(x,s)$, such that the first part is constructed to agree with the existence of a ballistic regime.

Substituting Eq. (58) up to $O(s^{-4})$ in Eq. (48) yields

$$\tilde{f}_{\rm I}(x,s)_{s\to\infty} = \frac{1}{2\sqrt{a_0}} \exp\left[-\frac{|\,\Delta x\,|}{\sqrt{a_0}}\left(s - \frac{3a_1}{2a_0}\right)\right] \ . \qquad (59)$$

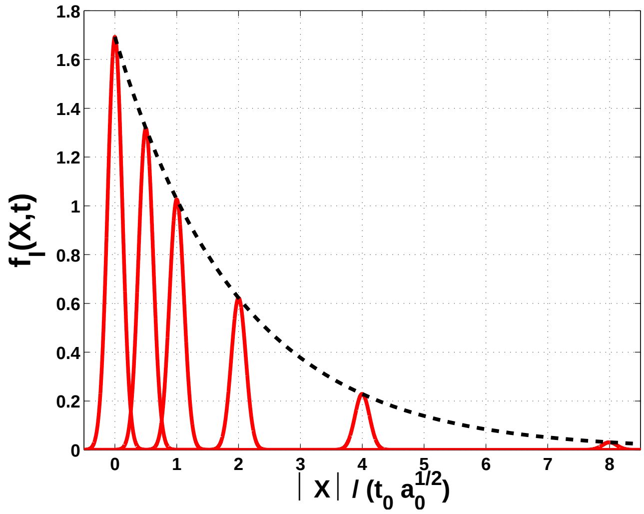

Fig. 1. The time evolution of the function $f_{\rm I}(X,t)$ defined by Eq. (60) for time intervals t/t_0 =0.5, 1, 2, 4, 8 (the time scale $t_0 = a_0/(3a_1)$). The δ-function is graphically represented by narrow Gaussians

The inverse Laplace transformation of this result gives

$$f_{\rm I}(x,t) = \frac{1}{2} \exp\left(\frac{3a_1}{2a_0} t\right) \delta(|\,\Delta x\,| - \sqrt{a_0} t) \ . \qquad (60)$$

Not surprisingly, this partial solution corresponds to a deterministic propagation. Note that, in order to avoid the exponential divergence in time, we must have $a_1 < 0$ in the expansion (57). Schematically, the time evolution of this term is shown in Fig. 1, where the δ-function is graphically represented as a narrow Gaussian. The dashed line represents the exponential decay of the integral over the δ-function.

Having found $\tilde{f}_{\rm I}(x,s)$, we can now write $\tilde{f}_{\rm II}(x,s)$ simply as

$$\tilde{f}_{\rm II}(x,s) = \tilde{f}(x,s) - \tilde{f}_{\rm I}(x,s) \ . \qquad (61)$$

Calculating this difference explicitly, we find

$$\tilde{f}_{\rm II}(x,s) = \frac{1}{2}\left\{\sqrt{\frac{2}{s^3\langle x^2\rangle_s}} \times \right.$$

$$\times \exp\left[-\,|\,x\,|\left(\sqrt{\frac{2}{s\langle x^2\rangle_s}} - \frac{s}{\sqrt{a_0}}\right)\right] -$$

$$\left. - \frac{1}{\sqrt{a_0}} \exp\left(\frac{3a_1}{2a_0^{3/2}}\,|\,x\,|\right)\right\} \exp\left(-\frac{|\,x\,|}{\sqrt{a_0}} s\right) \equiv$$

$$\equiv \tilde{\Phi}(x,s)\exp\Big(-\frac{|\,x\,|}{\sqrt{a_0}}s\Big). \tag{62}$$

The inverse Laplace transformation of Eq. (62) yields

$$f_{\rm II}(x,t) = \Phi\Big(x, t-|\,x\,|/\sqrt{a_0}\Big)\Theta(\sqrt{a_0}t-|\,x\,|). \tag{63}$$

The importance of this result is that the explicit Heaviside function is taking upon itself the discontinuity in the solution $f_{\rm II}(x,t)$. The exact value of the function $\Phi\Big(x, t-|\,x\,|/\sqrt{a_0}\Big)$ at the point $|\,x\,|=\sqrt{a_0}t$ can be calculated using the initial-value theorem and is given by

$$\Phi(x,0) = -\left\{\frac{3}{4}\frac{a_1}{a_0^{3/2}} + \frac{1}{2\sqrt{a_0}}\Big[\frac{27}{8}\Big(\frac{a_1}{a_0}\Big)^2 - 6\frac{a_2}{a_0}\Big]t\right\}\times$$

$$\times\exp\left(\frac{3}{2}\frac{a_1}{a_0}t\right). \tag{64}$$

Summing together results (60) and (63) in the time domain, we get a general solution of the non-Markovian problem with a short-time ballistic behavior in the form

$$f(x,t) = \frac{1}{2}\exp\left(\frac{3a_1}{2a_0}t\right)\delta(|\,x\,|-\sqrt{a_0}t)+$$

$$+\Phi\left(x, t-\frac{|\,x\,|}{\sqrt{a_0}}\right)\Theta(\sqrt{a_0}t-|\,x\,|). \tag{65}$$

The diffusion repartition of the probability distribution function occurs inside the spatial diffusion domain which increases in a deterministic way. The first term in Eq. (65) corresponds to the propagating δ-function which is inherited from the initial conditions, and it lives at the edge of the ballistically expanding domain. The function $\Phi(x,t)$ in the time domain is a continuous function and can be evaluated numerically, for example, by using the direct integration method [59]. Below, we demonstrate this calculation with explicit examples.

5.2. Common asymptotic diffusion

In the case of the common asymptotic diffusion, the mean square displacement is given by Eq. (51). It is convenient to introduce variables $\langle\xi^2\rangle = \langle\Delta x^2\rangle/(k_{\rm B}T)$ and $\tau = t/t_{\rm rel}$. Then Eq. (51) reads

$$\langle\xi^2\rangle_\tau = 2\big[\tau - (1-{\rm e}^{-\tau})\big]. \tag{66}$$

In this case, the Taylor expansion of the mean square displacement is given by:

$$\langle\xi^2\rangle_\tau = \tau^2 - \frac{1}{3}\tau^3 + \ldots. \tag{67}$$

Therefore, the coefficient $a_0 = 1$ and the coefficient $a_1 = -1/3$ in Eq. (57), and the ballistic contribution to the pdf (see Eq. (59)) is defined by:

$$f_{\rm I}(\xi,\tau) = \frac{1}{2}\exp\big(-\tau/2\big)\delta(|\,\xi\,|-\tau). \tag{68}$$

In order to derive the second contribution to the pdf $f_{\rm II}(\xi,\tau)$, it is necessary to perform the Laplace transformation of Eq. (66):

$$\langle\xi^2\rangle_s = \frac{2}{s^2(s+1)}. \tag{69}$$

The substitution of numerical values of the coefficients a_0 and a_1 and the Laplace transform of the mean-square displacement from Eq. (69) to Eq. (62) yields

$$\tilde{f}_{\rm II}(\xi,s) = \frac{1}{2}\left[\tilde{f}'_{\rm II}\Big(\xi, s+\frac{1}{2}\Big)\tilde{f}''_{\rm II}\Big(\xi, s+\frac{1}{2}\Big)\right]\times$$

$$\times\exp\left[-\sqrt{2}\,|\,\xi\,|\left(s+\frac{1}{2}\right)\right], \tag{70}$$

where

$$\tilde{f}'_{\rm II}(\xi,s) = \frac{\exp\Big[-|\,\xi\,|\,(\sqrt{s^2-1/4}-s)\Big]}{\sqrt{s^2-1/4}} \tag{71}$$

and

$$\tilde{f}''_{\rm II}(\xi,s) = s\tilde{f}'_{\rm II}(\xi,s) - 1 - \frac{1}{2}\tilde{f}'_{\rm II}(\xi,s). \tag{72}$$

From the initial-value theorem, $f'_{\rm II}(\xi,t=0)=1$, therefore, it follows from Eq. (72) that

$$f''_{\rm II}(\xi,\tau) = \frac{\partial}{\partial\tau}f'_{\rm I}(\xi,\tau) - \frac{1}{2}f'_{\rm I}(\xi,\tau). \tag{73}$$

The inverse Laplace transform of Eq. (71) is given by [60]

$$f'_{\rm I}(\xi,\tau) = I_0\left(\frac{1}{2}\sqrt{\tau^2+2\,|\,\xi\,|\,\tau}\right), \tag{74}$$

where $I_0(z)$ is the modified Bessel function of the first kind. Taking the first and second translation theorems into account, the inverse Laplace transformation of Eq. (70) yields

$$f_{\rm II}(\xi,t) = \frac{1}{4}\exp(-\tau/2)\left[I_0\left(\frac{1}{2}\sqrt{\tau^2-\xi^2}\right)+\right.$$

$$+\tau\frac{I_1\left(\frac{1}{2}\sqrt{\tau^2-\xi^2}\right)}{\sqrt{\tau^2-\xi^2}}\Bigg]\Theta(\tau-\mid\xi\mid). \qquad (75)$$

The asymptotic behavior of a modified Bessel function of the first kind is defined by $I_\nu(z)_{z\to\infty}\sim\exp(z)/\sqrt{2\pi z}$; therefore, Eq. (75) is reduced in the long-time limit to the solution of the common diffusion equation given by Eq. (9) with $d=1$.

5.3. Interpolation for all times

In order to connect the short- and long-time limits of the mean square displacement, it was proposed to use simple analytic expressions with desired asymptotic properties [61, 64]. To interpolate the mean square displacement, the formula

$$\langle\Delta x^2\rangle_t=2D_\alpha t_0^\alpha\frac{(t/t_0)^2}{[1+(t/t_0)]^{2-\alpha}}, \qquad (76)$$

where $0\le\alpha\le 2$ and t_0 is the crossover characteristic time, was proposed in [58]. Law (76) describes the ballistic regime at $t\ll t_0$ and the fractional diffusion at $t\gg t_0$.

We now introduce now dimensionless variables $\langle\xi^2\rangle_\tau=\langle\Delta x^2\rangle_t/(2D_\alpha t_0^\alpha)$ and $\tau=t/t_0$. With these variables, the last equation looks as

$$\langle\xi^2\rangle_\tau=\frac{\tau^2}{(1+\tau)^{2-\alpha}}. \qquad (77)$$

The Taylor expansion of (76) is given by

$$\langle\xi^2\rangle_\tau=\tau^2-(2-\alpha)\tau^3+\frac{1}{2}(3-\alpha)(2-\alpha)\tau^4+\dots. \qquad (78)$$

The substitution of these expansion coefficients into Eq. (60) yields the first term in the expression for the probability distribution function (65)

$$f_{\rm I}(\xi,t)=\frac{1}{2}\exp\left[-\frac{3(2-\alpha)}{2}\tau\right]\delta(\mid\xi\mid-\tau). \qquad (79)$$

The Laplace transform of Eq. (77) is

$$\langle\xi^2\rangle_s=\left(\frac{\alpha}{s}-1\right)\frac{1}{s}+\left[(\alpha-1)\left(\frac{\alpha}{s}-2\right)+s\right]\times$$

$$\times\frac{e^s}{s^\alpha}\Gamma(\alpha-1,s), \qquad (80)$$

where $\Gamma(a,s)$ is the incomplete gamma function. Note that the case $\alpha=2$ is special, since $\langle\xi^2\rangle_s=2/s^3$. Therefore, $f_{\rm II}(\xi,\tau)\equiv 0$ and it annuls the exponent in Eq. (79), leaving a ballistically propagating δ-function as a solution. For all other values of $\alpha<2$, the inverse Laplace transform of the function $\widetilde{\Phi}(\xi,s)$ which defines the diffusion process inside the expanded spatial domain should be evaluated, in general, numerically.

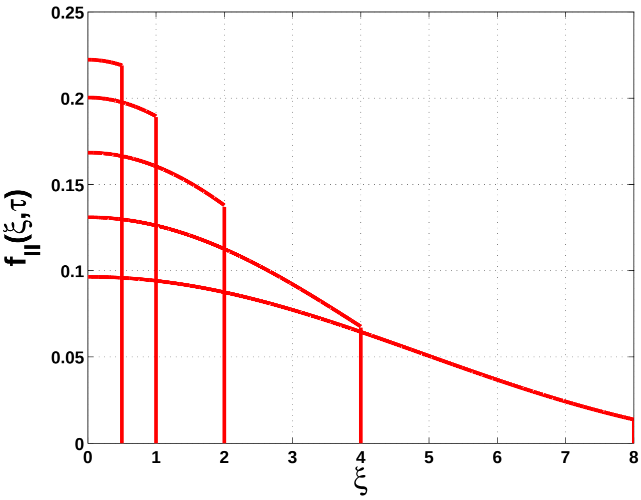

Fig. 2. Continuous part of pdf (75) for time intervals τ =.5, 1, 2, 4, 8. The reader should note that the full solution of the problem is the sum of the two solutions shown in this and the previous figure

The results of calculations following the method of Ref. [59] for the smooth part of the probability distribution function $f_{\rm II}(\xi,t)$ for different values of the parameter α are shown in Fig. 3. The reader should appreciate the tremendous role of memory. The ballistic part which is represented by the advancing and reducing δ-function sends backward the probability that it loses due to the exponential decay seen in Fig. 1. This 'back-diffusion' leads initially to a pdf with a maximum at the edge of the ballistically expanding domain. At later times the pdf begins to resemble that for a more regular diffusion. The effect strongly depends on α due to the appearance of α in the exponent in Eq. (79).

Nevertheless, the solution given by Eq. (75) for the Langevin mean square displacement shows a different behavior (compare Fig. 2 and the middle panel of Fig. 3 for $\alpha=1$). This is due to a relatively slow decay of the ballistic amplitude in Eq. 60) for this case in comparison with the interpolation by Eq. (76). Therefore, if the boundary propagation is defined by the first term in Eq. (57), the details of the near-boundary pdf behavior follows from the next correction in this expansion. Note that, for example, the ballistic peak of the pdf was observed in simulations of the test particle transport in pseudoturbulent fields [63].

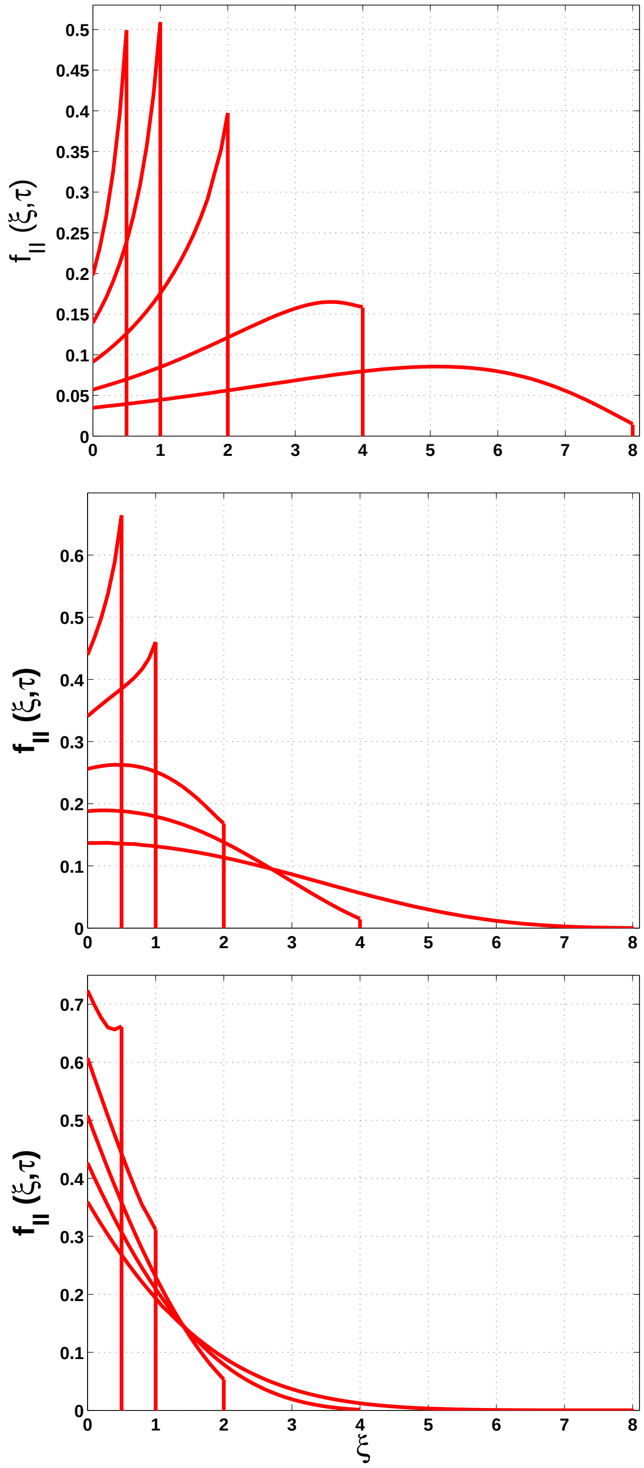

Fig. 3. Continuous part of pdf (63) for different values of the parameter α. Superdiffusion ($\alpha = 3/2$, upper panel), regular diffusion ($\alpha = 1$, middle panel), and subdiffusion ($\alpha = 1/2$, lower panel). Time intervals from the top to the bottom τ =0.5, 1, 2, 4, 8. The full solution of the problem is the sum of the two solutions shown in this figure and in Fig. (1)

For long times, the contribution from $f_{\rm I}(\xi,t)$ to the general solution tends to zero, and the solutions shown in Fig. 3 agree with the Markovian pdf obtained in the frame of a continuous-time random walk theory [64]. For the special case $\alpha = 0$, Eq. (80) reads as $\langle \xi^2 \rangle_s = 1/s + \left(1 - (2+s)e^s E_1(s)\right)$, where $E_1(s)$ is the exponential integral. The limiting behavior of the general solution from Eq. (65) $f(\xi,t)_{t \gg |\xi|/\sqrt{a_0}} \sim \Phi(\xi,t)$ can be evaluated analytically with the help of the final-value theorem $f(\xi) = (1/\sqrt{2})\exp(-\sqrt{2} \mid \xi \mid)$; this result coincides with the pdf from [64] under the same conditions.

6. Conclusion

The same mean square displacement (variance of the pdf) corresponds, in general, to different diffusion models. For the time non-local Fokker–Planck approach, we have shown how to deal with the diffusion processes that cross over from a ballistic to a fractional behavior for short and long times respectively, within the time non-local approach. The general solution (65) demonstrates the effect of the temporal memory in the form of a partition of the probability distribution function inside a spatial domain which increases in a deterministic way. The approach provides a solution that is valid at all times and, in particular, is free from the instantaneous action puzzle.

Unfortunately, the time non-local equation (41) is true only for one-dimensional systems. For higher dimensions, the pdf is not positive defined (as it is easy to see, this follows from properties of the wave equation solutions). For the two-dimensional persistent random walks, the pdf was found in [65], and it was shown that this function satisfies an inhomogeneous telegraph equation with a singular term located at the edge. Therefore, the used version of the time non-local Fokker–Planck equation should be corrected for higher dimensions, details will be discussed in the further publication.

This paper was partially supported by the Program of Fundamental Research of the Division of Physics and Astronomy of the National Academy of Sciences of Ukraine.

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Received 10.11.10

РІВНЯННЯ ФОККЕРА–ПЛАНКА З ПАМ'ЯТТЮ: ОБ'ЄДНАНИЙ ОПИС БАЛІСТИЧНИХ ТА ДИФУЗІЙНИХ ПРОЦЕСІВ

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Р е з ю м е

На основі немарковського узагальнення рівняння Фоккера–Планка запропоновано об'єднаний опис дифузійних процесів для довільних часів еволюції – від балістичної динаміки на малих часах до дробової (суб- чи супердифузії), або звичайної дифузії на великих часах. Встановлено зв'язок між немарковськими кінетичними коефіцієнтами зі спостережуваними величинами (середнім та середньоквадратичним зміщеннями). Обговорена проблема розрахунку кінетичних коефіцієнтів на основі рівняння Ланжевена. Знайдено розв'язок немарковського рівняння, що описує дифузійні процеси у реальному (координатному) просторі. Отримані розв'язки добре узгоджуються з результатами теорії випадкових неперервних у часі блукань, але, на відміну від останніх, послідовно описують також балістичний режим еволюції.