

THE STUDY OF FRACIONAL ANOMALOUS DIFFUSION LAWS ON THE HYPERSPHERE

ABDELGHANI DAOUDI^{1*}, MABROUK BENHAMOU² and EL HASSAN EL KINANI¹

ABSTRACT. In this work, we present an extended mathematical study of the fractional anomalous diffusion of a target particle which moves on a curved surface. To do explicit calculations, we assumed that this particle moves on a hypersphere of radius R . We analyzed the particle dynamics using a generalized fractional Langevin equation approach, based on the Caputo fractional derivative and Laplace transform techniques. We derived three physical quantities: the mean square displacement (MSD), the time diffusion coefficient (TDC), and the velocity autocorrelation function (VACF). We performed exact calculations of their temporal evolution by selecting power-law and Dirac delta memory functions. The results obtained are expressed in terms of Mittag-Leffler-type functions. The introduction of the fractional derivative affected the behavior of ballistic regime. We plotted the dynamic quantities for different values fractional derivative order and analyzed their influence on diffusion scaling laws.

1. INTRODUCTION

Diffusion phenomena were initially approached through a macroscopic perspective, such as Fick's laws [1, 2], or a statistical approach based on the diffusion equation (random walk model) [3]. It should be noted that many experiments have shown the limitations of models based on Fick's laws. Another mechanistic approach was introduced by Langevin [4], which is physically equivalent to the diffusion equation approach. Later, Kubo [5] and Zwanzig [6] have generalized Langevin's work by writing a *generalized Langevin equation* (GLE) to quantitatively study the diffusion phenomena *with memory*.

We recall that there are types of diffusion, namely a normal (or Brownian) diffusion, which occurs in a non-complex system (gas or low-viscosity liquid); and an anomalous diffusion, which produces in complex systems (*crowded systems*). The first diffusion is fast, while the second is very slow due to extreme difficulty for a particle to move in the host medium. For normal diffusions, MSD of the target particle increases linearly with time. For anomalous diffusions, however,

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* Corresponding author.

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MSD, denoted as $W(t)$, obeys an unusual power law, that is $W(t) \sim t^\gamma$, where, $0 < \gamma < 1$ for *subdiffusion*, $\gamma > 1$ for *superdiffusion*, and $\gamma = 1$ for normal diffusion. It should be noted that the anomalous diffusion is inherent in disordered or heterogeneous systems [7, 8], such as colloidal solutions, fractals, polymer melts, gels, living systems, charge carrier transport in amorphous semiconductors, percolation structures, polymer networks, and generally complex systems containing entities moving on different time scales.

The modeling of anomalous diffusion has been actively investigated, especially in the last decade, using the generalized Langevin equation introduced by Kubo [9, 10]. Another approach to anomalous diffusion is the use of the generalized diffusion equation [11], the fractional diffusion equation [12, 27], the fractional Fokker-Planck equation [13], the Chapman-Kolmogorov equation [14], and the generalized fractional Langevin equation (FGLE) [15, 16, 17], involving the fractional derivative, where the order of integrals and derivatives is generalized to fractional order [18].

Nevertheless, the influence of the geometry of the host media on the diffusion phenomena is a question which has not been sufficiently studied in the past. Among possible geometries, we can quote surfaces of bubbles (gas-liquid interfaces) in foams, surfaces of droplets (oil-water interfaces) in Pickering emulsions [19], and cell-membranes [20] or vesicles surfaces. The diffusion of the target particle on surfaces with curvature, such as spheres, ellipsoids, torus, etc., is a daunting task. An experimental analysis of particle diffusion on the stability of Pickering oil-in-water emulsions stabilized by hydrophobized fumed silica was studied [21], which is equivalent to a process on a hypersphere, while a theoretical analysis of diffusion of particles confined to surfaces was recently studied [22, 23, 24].

In the present study, the anomalous diffusion on curved surfaces is investigated using *fractional generalized Langevin equation* (FGLE). To perform calculations, we consider a spherical surface as geometry and investigate the influence of the curvature on the diffusion properties and the behavior of physical quantities.

The first part of this paper is dedicated to useful generalities necessary for our study of the anomalous diffusion occurring in crowded systems. In the second part, we discuss the solution of FGLE on a hypersphere, using Laplace transformation technique. To derive the general relations describing the time evolution of the obtained quantities, the asymptotic behaviors in short and long time limits are analyzed. For analysis, we choose Dirac delta memory function and power-law memory function. Finally, we plot the dynamical quantities for different values of the fractional derivative order and analyze their effect.

2. PRELIMINARIES

Definition 2.1. Consider *two* dynamic quantities, A and B . Their individual time-average and time correlation function are defined by:

$$\langle A(t_0) \rangle = \lim_{\tau \rightarrow \infty} \frac{1}{\tau} \int_{-\tau/2}^{\tau/2} dt A(t + t_0), \quad (2.1)$$

$$\langle A(t_1) B(t_0) \rangle = \lim_{\tau \rightarrow \infty} \frac{1}{\tau} \int_{-\tau/2}^{\tau/2} dt A(t+t_1) B(t+t_0). \quad (2.2)$$

Here τ is essentially the time interval over which quantities A and B are measured experimentally.

Definition 2.2. We define MSD of a target particle by:

$$W(t) = \langle [\mathbf{r}(t) - \mathbf{r}(0)]^2 \rangle, \quad t \geq 0. \quad (2.3)$$

Here $\mathbf{r}(t) \in \mathbf{R}^d$ is the instantaneous position in Euclidean space of dimension d .

Definition 2.3. We define VACF of a target particle by:

$$c_{vv}(t) = \langle \mathbf{v}(t) \cdot \mathbf{v}(0) \rangle, \quad t \geq 0. \quad (2.4)$$

Here $\mathbf{v}(t) \in \mathbf{R}^d$ is the instantaneous velocity of the target particle.

Definition 2.4. We define TDC by:

$$D(t) = \frac{1}{2d} \frac{dW(t)}{dt}, \quad t \geq 0. \quad (2.5)$$

We present some basic concepts of the Riemann-Liouville (R-L) fractional integral as well as the definition of fractional derivative in the sense of Caputo (see, e.g., [18] for more details). The choice of the Caputo derivative is motivated by the fact that it allows for a more intuitive definition of initial conditions and that its derivative for a constant is zero.

Definition 2.5. Let $f : [a, b] \rightarrow \mathbf{R}$ be a continuous real function; its *Riemann-Liouville fractional integral*, of order $\alpha \in \mathbf{C}$, with $\Re[\alpha] > 0$, is defined by [18]:

$$I_a^\alpha f(x) = \frac{1}{\Gamma(\alpha)} \int_a^x (x-t)^{\alpha-1} f(t) dt. \quad (2.6)$$

Here a is a real number, and $\Gamma(\alpha)$ accounts for *Euler's gamma function*.

Definition 2.6. Let $f \in \mathbf{C}^n([a, b])$ and $\alpha \in]n-1, n[$, with $n \in \mathbf{N}$. *Caputo derivative*, of order α , of a sufficiently integrable function, f , is given by [18]:

$$({}^C D_a^\alpha f)(x) = \frac{1}{\Gamma(n-\alpha)} \int_a^x (x-t)^{n-\alpha-1} f^{(n)}(t) dt. \quad (2.7)$$

Definition 2.7. The *three-parameter Mittag-Leffler* (M-L) function, $E_{\alpha,\beta}^\delta(z)$, is defined by its series:

$$E_{\alpha,\beta}^\delta(z) = \sum_{n=0}^{+\infty} \frac{(\delta)_n}{\Gamma(\alpha n + \beta)} \frac{z^n}{n!}, \quad \alpha, \beta, \delta, z \in \mathbf{C}, \Re[\alpha] > 0, \Re[\beta] > 0, \Re[\gamma] > 0, \quad (2.8)$$

where $(\delta)_n$ is the *Pochhammer symbol*:

$$(\delta)_0 = 1, \quad (\delta)_n = \delta(\delta+1)(\delta+2)\dots(\delta+n-1). \quad (2.9)$$

We note that for $\beta = \delta = 1$, we recover the *classical* M-L function, $E_\alpha(z)$, and for $\delta = 1$ we obtain the *two-parameter* M-L function, $E_{\alpha,\beta}(z)$. These functions, essentially employed in solving fractional partial differential equations and

generalize the exponential function [25]. Their behaviors are as follows:

$$E_{\alpha,\beta}^{\delta}(-z) = \frac{z^{-\delta}}{\Gamma(\beta - \alpha\delta)}, \quad z \rightarrow +\infty. \quad (2.10)$$

$$E_{\alpha,\beta}^{\delta}(-z) = \frac{1}{\Gamma(\beta)}, \quad z \rightarrow 0. \quad (2.11)$$

Let us recall the Laplace transform of the M-L function and the Caputo derivative, which will be used in what follows:

Proposition 2.8. *The Laplace transform of the function $t^{\alpha k + \beta - 1} E_{\alpha,\beta}^{(k)}(\pm at^{\alpha})$ can be expressed as [25]:*

$$\int_0^{\infty} e^{-st} t^{\alpha k + \beta - 1} E_{\alpha,\beta}^{(k)}(\pm at^{\alpha})(s) dt = \frac{s^{\alpha - \beta} k!}{(s^{\alpha} \mp a)^{k+1}} (Re(s) > |a|^{1/\alpha}), \quad (2.12)$$

with,

$$E_{\alpha,\beta}^{(k)}(t) = \frac{d^k}{dt^k} (E_{\alpha,\beta}(t)). \quad (2.13)$$

We have the following result for the Laplace transform:

$$\mathcal{L} \{ t^{\beta-1} E_{\alpha,\beta}^{\delta}(\pm at^{\alpha}) \} (s) = \frac{s^{\alpha\delta - \beta}}{(s^{\alpha} \mp a)^{\delta}}, \quad (Re(s) > |a|^{1/\alpha}), \quad (2.14)$$

and,

$$\mathcal{L}^{-1} \left\{ \frac{s^{\alpha\delta - \beta}}{(s^{\alpha} \mp a)^{\delta}} \right\} (t) = t^{\beta-1} E_{\alpha,\beta}^{\delta}(\pm at^{\alpha}), \quad (Re(s) > |a|^{1/\alpha}). \quad (2.15)$$

Proposition 2.9. *Laplace transform of a stationary correlation function,*

$$\langle f(t) f(t') \rangle = \Phi(|t - t'|), \quad (2.16)$$

of an arbitrary function, $f(t)$, is [26]:

$$\langle \hat{f}(s) \hat{f}(s') \rangle = \frac{\hat{\Phi}(s) + \hat{\Phi}(s')}{s + s'}. \quad (2.17)$$

Here $\hat{\Phi}(s)$ is Laplace transform of function $\Phi(t)$.

3. FGLE ON HYPERSPHERE

3.1. Inertial generalized stochastic equation.

Consider a given target particle of instantaneous position, $\mathbf{r}(t) \in \mathbf{R}^d$, which moves on a hypersphere of radius R . The equation of this hypersphere is:

$$\mathcal{C}(\mathbf{r}) : \mathbf{r}^2(t) - R^2 = 0. \quad (3.1)$$

This hypersphere may be a rigid body or a soft one as an interface between two immiscible liquids. The curved geometry imposes a *constraint force*, $\mathbf{F}_{ex}(\mathbf{r})$ on the particle proportional to the derivative of the constraint, along with the perpendicular direction to the surface, preventing this particle from leaving the

hypersphere. The expression of such a force can be inspired from the analytic mechanics principles, and we have:

$$\mathbf{F}_{ex}(\mathbf{r}) = -\frac{\lambda}{2} \frac{\partial \mathcal{C}(\mathbf{r})}{\partial \mathbf{r}} = -\lambda \mathbf{r}(t), \quad (3.2)$$

where λ is a positive factor (*Lagrange multiplier*) which will be identified below. Then, such a force plays the role of an *elastic force*. We shall denote by $\mathbf{r}_0 = \mathbf{r}(0) \in \mathbf{R}^d$, the initial position of the target particle on hypersphere.

The diffusion process can be described using the so-called FGLE, introduced by Kubo and Zwanzig:

$$m_\alpha \cdot {}^c D_0^\alpha [\mathbf{v}(t)] = \left[-m \int_0^t dt_1 \gamma(t-t_1) \mathbf{v}(t_1) \right] - \lambda \mathbf{r}(t) + \mathbf{F}(t). \quad (3.3)$$

The preceding equation may be expressed, through identity: $\mathbf{r}(t) - \mathbf{r}_0 = \int_0^t dt_1 \mathbf{v}(t_1)$ in the form:

$$m_\alpha \cdot {}^c D_0^\alpha [\mathbf{v}(t)] = -m \left[\int_0^t dt_1 \gamma(t-t_1) \mathbf{v}(t_1) \right] - \lambda \left[\int_0^t dt_1 \mathbf{v}(t_1) \right] - \lambda \mathbf{r}_0 + \mathbf{F}(t), \quad (3.4)$$

with the notations:

$$0 < \alpha \leq 1, \quad m_\alpha = m \cdot \tau^{\alpha-1}. \quad (3.5)$$

Here $\mathbf{v}(t) \in \mathbf{R}^d$ denotes the velocity of the target particle, $\tau^{\alpha-1}$ is a homogeneity coefficient and $\gamma(t)$ accounts for the *memory function* expressing a delay of frictions with the molecules located on the spherical surface. There m accounts for the real mass of the target particle, m_α , for a generalized mass of this particle, τ , for a time-scale, and $\mathbf{F}(t) \in \mathbf{R}^d$, for a *stochastic force* (*Langevin force*), obeying the following three rules:

$$\langle \mathbf{F}(t) \rangle = 0, \quad t > 0, \quad (3.6)$$

$$\langle \mathbf{F}(t_1) \cdot \mathbf{F}(t_2) \rangle = 2dk_B T m \gamma(|t_1 - t_2|), \quad (3.7)$$

$$\langle \mathbf{v}(0) \cdot \mathbf{F}(t) \rangle = 0. \quad (3.8)$$

The last condition means that the velocity of the target particle and the stochastic force are *uncorrelated*. Indeed, such a force does not depend on the velocity of the moving particle.

We note that the above stochastic equation must be supplemented with the fact that the target particle moves but remains all the time on the hypersphere, that is we must have:

$$\mathbf{r}^2(t) = R^2, \quad t \geq 0. \quad (3.9)$$

By simple derivation of this equation with respect to time, we find the equivalent condition:

$$\mathbf{r}(t) \cdot \mathbf{v}(t) = 0, \quad t \geq 0. \quad (3.10)$$

3.2. Basic relationships.

3.2.1. *Formal expression of velocity autocorrelation function(VACF).* We begin by recalling that the Laplace transform of a convolution product, $(f * g)(t)$, is simply $\widehat{f}(s) \times \widehat{g}(s)$ and $\mathcal{L}({}^c D_0^\alpha [c_{vv}(t)])(s) = s^\alpha \widehat{c}_{vv}(s) - s^{\alpha-1} \langle \mathbf{v}_0^2 \rangle$.

Multiplying stochastic equation (3.4) by $\mathbf{v}(0)$ and taking into account the two conditions (3.8) and (3.10) gives the following integro-differential equation satisfied by VACF:

$${}^c D_0^\alpha [\langle \mathbf{v}(t) \cdot \mathbf{v}(0) \rangle] = - \int_0^t dt_1 \left[\frac{m}{m_\alpha} \gamma(t-t_1) + \frac{\lambda}{m_\alpha} \theta(t-t_1) \right] \langle \mathbf{v}(t_1) \cdot \mathbf{v}(0) \rangle , \quad (3.11)$$

which can rewrite as:

$${}^c D_0^\alpha [c_{vv}(t)] = - \int_0^t dt_1 \left[\frac{m}{m_\alpha} \gamma(t-t_1) + \frac{\lambda}{m_\alpha} \theta(t-t_1) \right] c_{vv}(t_1) . \quad (3.12)$$

Here $\theta(t)$ denotes the *Heaviside step function*. This is an equation describing the relaxation of $\langle \mathbf{v}(t) \rangle$ with respect to the initial value $\langle \mathbf{v}(0) \rangle$.

After applying the Laplace transforming to equation (3.12) we obtain:

$$\widehat{c}_{vv}(s) = \langle \mathbf{v}_0^2 \rangle \cdot \widehat{K}(s) , \quad (3.13)$$

with de notations:

$$\widehat{K}(s) = \frac{s^{\alpha-1}}{s^\alpha + \frac{m}{m_\alpha} \widehat{\gamma}(s) + \frac{\lambda}{m_\alpha} s^{-1}} , \quad (3.14)$$

and $\mathbf{v}_0 = \mathbf{v}(0)$.

Inverse Laplace transform of equality (3.13) yields:

$$c_{vv}(t) = \langle \mathbf{v}_0^2 \rangle K(t) . \quad (3.15)$$

Notice that $K(0) = 1$ for all diffusion processes, since $c_{vv}(0) = \langle \mathbf{v}_0^2 \rangle = d \frac{k_B T}{m}$.

3.2.2. Formal expression of velocity.

Laplace transforming stochastic equation (3.4):

$$\widehat{\mathbf{v}}(s) = \widehat{K}(s) \mathbf{v}_0 - \frac{\lambda}{m_\alpha} \widehat{G}(s) \mathbf{r}_0 + \frac{1}{m_\alpha} \widehat{g}(s) \widehat{\mathbf{F}}(s) , \quad (3.16)$$

with,

$$\widehat{g}(s) = \frac{1}{s^\alpha + \frac{m}{m_\alpha} \widehat{\gamma}(s) + \frac{\lambda}{m_\alpha} s^{-1}} , \quad (3.17)$$

$$\widehat{G}(s) = s^{-1} \widehat{g}(s) = \frac{s^{-1}}{s^\alpha + \frac{m}{m_\alpha} \widehat{\gamma}(s) + \frac{\lambda}{m_\alpha} s^{-1}} . \quad (3.18)$$

Inverse LT of equality (3.16) gives the instantaneous velocity of the target particle, namely:

$$\mathbf{v}(t) = K(t) \mathbf{v}_0 - \frac{\lambda}{m_\alpha} G(t) \mathbf{r}_0 + \frac{1}{m_\alpha} \int_0^t dt_1 g(t-t_1) \mathbf{F}(t_1) . \quad (3.19)$$

By averaging on the random force, $\mathbf{F}(t)$, we find that the time-average of velocity is given by:

$$\langle \mathbf{v}(t) \rangle = K(t) \langle \mathbf{v}_0 \rangle - \frac{\lambda}{m_\alpha} G(t) \langle \mathbf{r}_0 \rangle . \quad (3.20)$$

The value at $t = 0$ imposes the following additional identity: $G(0) = 0$, for all diffusion processes.

3.2.3. Formal expression of position.

A combination of relation $\widehat{\mathbf{v}}(s) = s\widehat{\mathbf{r}}(s) - \mathbf{r}_0$ and equality (3.16) allows the following formula:

$$\widehat{\mathbf{r}}(s) = \frac{\mathbf{r}_0}{s} + \widehat{H}(s) \mathbf{v}_0 - \frac{\lambda}{m_\alpha} \widehat{I}(s) \mathbf{r}_0 + \frac{1}{m_\alpha} \widehat{G}(s) \widehat{\mathbf{F}}(s) , \quad (3.21)$$

with the notations:

$$\widehat{I}(s) = s^{-1} \widehat{G}(s) = s^{-2} \widehat{g}(s) = \frac{s^{-2}}{s^\alpha + \frac{m}{m_\alpha} \widehat{\gamma}(s) + \frac{\lambda}{m_\alpha} s^{-1}} , \quad (3.22)$$

$$\widehat{H}(s) = s^\alpha \widehat{I}(s) = \frac{s^{\alpha-2}}{s^\alpha + \frac{m}{m_\alpha} \widehat{\gamma}(s) + \frac{\lambda}{m_\alpha} s^{-1}} . \quad (3.23)$$

Inverse Laplace transform of relation (3.21) gives the expression of the evolution of position in time:

$$\mathbf{r}(t) = \mathbf{r}_0 - \frac{\lambda}{m_\alpha} I(t) \mathbf{r}_0 + H(t) \mathbf{v}_0 + \frac{1}{m_\alpha} \int_0^t G(t-t_1) \mathbf{F}(t_1) dt_1 . \quad (3.24)$$

Averaging on the random force leads to the time-average of position:

$$\langle \mathbf{r}(t) \rangle = \left(1 - \frac{\lambda}{m_\alpha} I(t) \right) \mathbf{r}_0 + H(t) \langle \mathbf{v}_0 \rangle . \quad (3.25)$$

The above results indicate that the time-average, $\langle \mathbf{r}(t) \rangle$, belongs to a plan containing the initial position-vector and velocity, $\mathbf{r}_0$ and $\mathbf{v}_0$. Setting $t = 0$ in expression above gives the additional condition: $I(0) = H(0) = 0$, whatever is the kind of the diffusion process.

We state that, for larger times, the position correlation function, $\langle \mathbf{r}(t) \cdot \mathbf{r}(0) \rangle$, tends to 0. Then, this implies:

$$I(+\infty) = \frac{m_\alpha}{\lambda} , \quad (3.26)$$

for all times.

From relations (3.22) and (3.23), it follows that:

$$I(t) = \int_0^t G(t) dt , \quad G(t) = {}^{RL}I_0^\alpha [K(t)] , \quad I(t) = {}^{RL}I_0^{\alpha+1} [K(t)] . \quad (3.27)$$

3.2.4. Formal expressions of MSD and TDC.

Combining the relation (3.25) with the conditions $\mathbf{r}^2(t) = R^2$ and $\mathbf{r}_0 \cdot \mathbf{v}_0 = 0$, since the target particle moves on the hypersphere, yields the formal expression:

$$\langle \mathbf{r}(t) \cdot \mathbf{r}(0) \rangle = \left(1 - \frac{\lambda}{m_\alpha} I(t) \right) R^2. \quad (3.28)$$

From the formal expression of MSD, i.e., $W(t) = \langle \mathbf{r}^2(t) \rangle + \mathbf{r}_0^2 - 2 \langle \mathbf{r}(t) \cdot \mathbf{r}(0) \rangle$, we deduce that:

$$W(t) = 2R^2 \frac{\lambda}{m_\alpha} I(t). \quad (3.29)$$

By a simple derivation of this MSD with respect to time, we get the formal expression of TDC:

$$D(t) = \frac{R^2}{d} \frac{\lambda}{m_\alpha} G(t). \quad (3.30)$$

Since the adjustable constant, λ , to be computed does not depend on the particular stochastic process, it will be determined in the limit $\alpha \rightarrow 1$. We show that (For more detail see appendix):

$$\langle \mathbf{r}^2(t) \rangle = R^2 + 2 \frac{\lambda}{m} \left(d \frac{k_B T}{\lambda} - R^2 \right) I(t) + \left(\frac{\lambda}{m} \right)^2 \left(R^2 - d \frac{k_B T}{\lambda} \right) I^2(t). \quad (3.31)$$

Then, since $\langle \mathbf{r}^2(t) \rangle = R^2$, yields:

$$\lambda = d \frac{k_B T}{R^2}. \quad (3.32)$$

Therefore, the final expressions for MSD, TDC and VACF are:

$$W(t) = 2d \frac{k_B T}{m_\alpha} I(t), \quad (3.33)$$

$$D(t) = \frac{k_B T}{m_\alpha} G(t), \quad (3.34)$$

$$c_{vv}(t) = d \frac{k_B T}{m} K(t). \quad (3.35)$$

After some algebraic manipulation, we show that the above general relations can be rewritten as:

$$W(t) = 2 \frac{m}{m_\alpha} \times^{RL} I_0^{\alpha+1} [c_{vv}(t)], \quad (3.36)$$

$$D(t) = \frac{1}{d} \frac{m}{m_\alpha} \times^{RL} I_0^\alpha [c_{vv}(t)], \quad (3.37)$$

$$c_{vv}(t) = \frac{1}{2} \frac{m_\alpha}{m} \times^C D_0^{\alpha+1} [W(t)]. \quad (3.38)$$

3.2.5. Short and large times behaviors of dynamic quantities.

In order to study the asymptotic behaviors of the dynamic quantities, we analyze the behaviors of the relaxation functions $I(t)$, $G(t)$, and $K(t)$ starting from their Laplace transform and using the Tauberian theorems[16], which we recall here: the behavior of a function, $f(t)$, for $t \rightarrow +\infty$, can be extracted from that of its Laplace transform, $\hat{f}(s)$, for $s \rightarrow 0$, and the behavior of Laplace transform, $\hat{f}(s)$, for $s \rightarrow \infty$, can be deduced from that of function, $f(t)$, for $t \rightarrow 0$.

Therefore, in the limit $s \rightarrow 0$ and the condition: $s\hat{\gamma}(s) \rightarrow 0$, as $s \rightarrow 0$, which corresponds to the vanishing of memory effects at long times, we obtain:

$$\hat{I}(s) \sim \frac{m_\alpha}{\lambda} s^{-1}, \quad \hat{G}(s) \sim \frac{m_\alpha}{\lambda}, \quad \hat{K}(s) \sim \frac{m_\alpha}{\lambda} s^\alpha. \quad (3.39)$$

Inverse Laplace transform gives the large-time behaviors for the relaxations functions namely:

$$I(t) = \frac{m_\alpha}{\lambda}, \quad G(t) = 0, \quad K(t) = 0. \quad (3.40)$$

Substituting the expressions(3.40) into (3.33), (3.34), and (3.35) yields:

$$W(t) = 2R^2, \quad D(t) = 0, \quad c_{vv}(t) = 0 \quad (3.41)$$

For $s \rightarrow +\infty$, the denominator of Laplace transform of relaxation functions are dominated by the term s , provided that the condition, $s^\alpha \hat{\gamma}^{-1}(s) \rightarrow +\infty$, as $s \rightarrow +\infty$, is fulfilled, and we have the following behaviors:

$$\hat{I}(s) \sim s^{-2-\alpha}, \quad \hat{G}(s) \sim s^{-1-\alpha}, \quad \hat{K}(s) \sim s^{-1}. \quad (3.42)$$

Inverse Laplace transform allows the behaviors of the relaxations functions, at small time:

$$I(t) = \frac{1}{\Gamma(\alpha+2)} t^{\alpha+1}, \quad G(t) = \frac{1}{\Gamma(\alpha+1)} t^\alpha, \quad K(t) = 1. \quad (3.43)$$

From relations (3.43), (3.33), (3.34), and (3.35), we obtain the dynamic quantities of interest, at small time:

$$W(t) = 2d \frac{k_B T}{m_\alpha} \frac{1}{\Gamma(\alpha+2)} t^{\alpha+1}, \quad (3.44)$$

$$D(t) = \frac{k_B T}{m_\alpha} \frac{1}{\Gamma(\alpha+1)} t^\alpha, \quad (3.45)$$

$$c_{vv}(t) = d \frac{k_B T}{m}. \quad (3.46)$$

4. EXPLICIT RESULTS

Here, to obtain explicit results, we consider two specific forms for the frictional memory kernels: a local memory function describing normal particle diffusion during the diffusion process, and a power-law form to describe anomalous diffusion, which is consistent with the long-time behavior of the MSD, $W(t) \sim t^\gamma$. Finally, we analyze and plot the dynamic quantities associated with this study.

4.1. Diffusion without memory.

Consider a target particle that freely diffuses on a hypersphere of radius, R . If the nanoparticle radius is high enough or the nanoparticle concentration is very weak, the diffusion of the target nanoparticle is Brownian or normal, and then, the associated memory function is local, that is $\gamma(t) = \gamma_1 \delta(t)$ is the friction coefficient. Laplace transform of relaxation functions, for this case, are:

$$\widehat{I}(s) = \frac{s^{-2}}{s^\alpha + \frac{m}{m_\alpha} \gamma_1 + \frac{\lambda}{m_\alpha} s^{-1}}, \quad (4.1)$$

$$\widehat{G}(s) = \frac{s^{-1}}{s^\alpha + \frac{m}{m_\alpha} \gamma_1 + \frac{\lambda}{m_\alpha} s^{-1}}, \quad (4.2)$$

$$\widehat{K}(s) = \frac{s^{\alpha-1}}{s^\alpha + \frac{m}{m_\alpha} \gamma_1 + \frac{\lambda}{m_\alpha} s^{-1}}. \quad (4.3)$$

We emphasize that inverse Laplace transform of the above quantities are not known exactly, but we can obtain information, at least for small constant λ , defined in relation (3.32). In fact, this approximation remain valid for big hyperspheres. By expansion in powers of reduced parameter, λ/m_α , we get:

$$\widehat{I}(s) = \sum_{n=0}^{+\infty} \left(-\frac{\lambda}{m_\alpha} \right)^n \frac{s^{-n-2}}{\left(s^\alpha + \frac{m}{m_\alpha} \gamma_1 \right)^{n+1}}, \quad (4.4)$$

$$\widehat{G}(s) = \sum_{n=0}^{+\infty} \left(-\frac{\lambda}{m_\alpha} \right)^n \frac{s^{-n-1}}{\left(s^\alpha + \frac{m}{m_\alpha} \gamma_1 \right)^{n+1}}, \quad (4.5)$$

$$\widehat{K}(s) = \sum_{n=0}^{+\infty} \left(-\frac{\lambda}{m_\alpha} \right)^n \frac{s^{-n+\alpha-1}}{\left(s^\alpha + \frac{m}{m_\alpha} \gamma_1 \right)^{n+1}}. \quad (4.6)$$

By inversion Laplace transform and using formula (2.15), we find the following expressions for relaxation functions, $I(t)$, $G(t)$, and $K(t)$:

$$I(t) = t^{\alpha+1} \sum_{n=0}^{+\infty} \left[-\left(\frac{t}{\tau_R} \right)^{\alpha+1} \right]^n E_{\alpha,(\alpha+1)(n+1)+1}^{n+1} \left(-\left(\frac{t}{\tau_\alpha} \right)^\alpha \right), \quad (4.7)$$

$$G(t) = t^\alpha \sum_{n=0}^{+\infty} \left[-\left(\frac{t}{\tau_R} \right)^{\alpha+1} \right]^n E_{\alpha,(\alpha+1)(n+1)}^{n+1} \left(-\left(\frac{t}{\tau_\alpha} \right)^\alpha \right), \quad (4.8)$$

$$K(t) = \sum_{n=0}^{+\infty} \left[-\left(\frac{t}{\tau_R} \right)^{\alpha+1} \right]^n E_{\alpha,n(\alpha+1)+1}^{n+1} \left(-\left(\frac{t}{\tau_\alpha} \right)^\alpha \right), \quad (4.9)$$

with the *characteristic times*:

$$\tau_\alpha = \left(\frac{m}{m_\alpha} \gamma_1 \right)^{-1/\alpha}, \quad \tau_R = \left(\frac{\lambda}{m_\alpha} \right)^{-1/(\alpha+1)} = \left(d \frac{k_B T}{m_\alpha R^2} \right)^{-1/(\alpha+1)}. \quad (4.10)$$

We note that, since the choice of parameter, γ_1 , is arbitrary, we can choose it so that the time-scale, τ_α , remains below the second one, τ_R , i.e. $\tau_\alpha < \tau_R$. Using the

relations (4.7), (4.8) and (4.9) yields the time expressions of the desired dynamic quantities:

$$\frac{W(t)}{W_0} = 2 \left(\frac{t}{\tau_R} \right)^{\alpha+1} \sum_{n=0}^{+\infty} (-1)^n \left[\left(\frac{t}{\tau_R} \right)^{\alpha+1} \right]^n E_{\alpha, (\alpha+1)(n+1)+1}^{n+1} \left[- \left(\frac{t}{\tau_R} \right)^{\alpha} \right], \quad (4.11)$$

$$\frac{D(t)}{D_0} = \left(\frac{t}{\tau_\alpha} \right)^{\alpha} \sum_{n=0}^{+\infty} (-1)^n \left[\left(\frac{t}{\tau_\alpha} \right)^{\alpha+1} \right]^n E_{\alpha, (\alpha+1)(n+1)}^{n+1} \left[- \left(\frac{t}{\tau_\alpha} \right)^{\alpha} \right], \quad (4.12)$$

$$\frac{c_{vv}(t)}{c_{vv}(0)} = \sum_{n=0}^{+\infty} (-1)^n \left[\left(\frac{t}{\tau_R} \right)^{\alpha+1} \right]^n E_{\alpha, n(\alpha+1)+1}^{n+1} \left[- \left(\frac{t}{\tau_\alpha} \right)^{\alpha} \right]. \quad (4.13)$$

with the notations:

$$W_0 = d \frac{k_B T}{\lambda}, \quad D_0 = \frac{k_B T}{m \gamma_1}, \quad c_{vv}(0) = d \frac{k_B T}{m}. \quad (4.14)$$

Quantity D_0 denotes simply the the standard diffusion coefficient.

Now, let us examine the time behaviors of dynamic quantities of interest. To this end, we have to distinguish three kinds of time-regimes, which are the following:

Cas 1 : α -ballistic regime : For $t < \tau_R$, the dominant parts of dynamic quantities come from the value $n = 0$, in the associated series, and we find that:

$$W(t) = 2d \frac{k_B T}{m_\alpha} \frac{1}{\Gamma(\alpha+2)} t^{\alpha+1}, \quad (4.15)$$

$$D(t) = \frac{k_B T}{m_\alpha} \frac{1}{\Gamma(\alpha+1)} t^{\alpha}, \quad (4.16)$$

$$c_{vv}(t) = d \frac{k_B T}{m}. \quad (4.17)$$

Note that the introduction of the fractional derivative has affected the behavior of the quantities during the ballistic regime. More precisely, the MSD no longer behaves like t^2 but like $t^{\alpha+1}$ and the TDC no longer behaves like t but like t^α . We thus call this regime the α -ballistic regime. For $\alpha = 1$, we find the behavior of the ballistic regime $W(t) = d \frac{k_B T}{m} t^2$.

Cas 2 : Normal diffusive regime : By using the asymptotic behavior of M-L functions (2.10) and (2.11), we obtain for $\tau_R < t < \tau_\alpha$ that :

$$W(t) = 2dD_1 t, \quad (4.18)$$

$$D(t) = D_0, \quad (4.19)$$

$$c_{vv}(t) = 0. \quad (4.20)$$

In this regime, the target particle executes a normal diffusion.

Cas 3 : Saturation regime : By using the asymptotic behaviors of M-L functions (2.10) and (2.11), we get for $t > \tau_\alpha$ that:

$$W(t) = 2R^2, \quad (4.21)$$

$$D(t) = 0, \quad (4.22)$$

$$c_{vv}(t) = 0. \quad (4.23)$$

For long times, i.e. $t \rightarrow +\infty$, MSD saturates at the finite value, $2R^2$. This result is not surprising, since the hypersphere area is finite, and then, after a time of the order of τ_α , all sites of this hypersphere are visited by the target particle.

Finally, we report in Figures (1), (2) and (3), the variation in time of the reduced MSD, $W(t)/W_0$, the reduced TDC, $D(t)/D_0$ and the reduced VACF, $c_{vv}(t)/c_{vv}(0)$, versus the renormalized time, t/τ_α , for different values of α , and we interpret the effect of the fractional order α on the behavior of dynamic quantities.

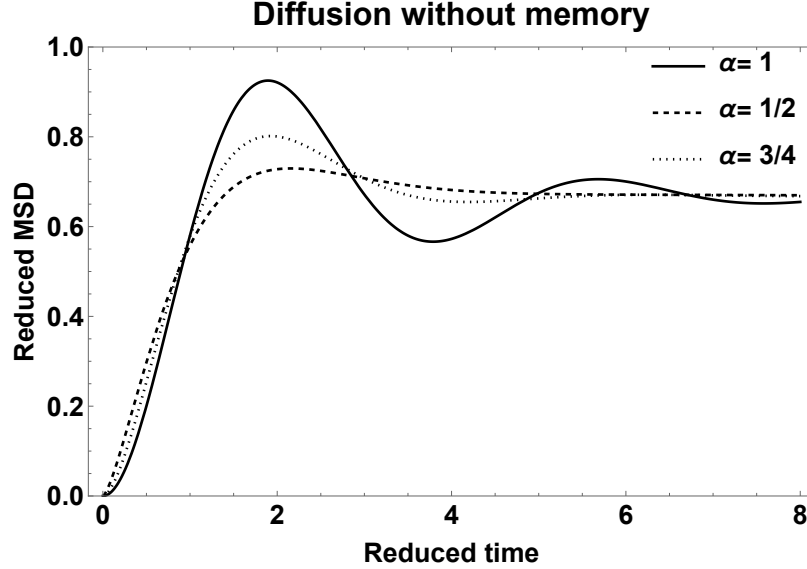


FIGURE 1. Graphical representation of the mean square displacement (4.11) for: $\tau_\alpha/\tau_R = 0.05$; $\alpha = 1$; $\alpha = 1/2$ and $\alpha = 3/4$.

Remark 4.1. When the constraint force is absent ($\lambda = 0$), the saturation phenomenon no longer occurs. Furthermore, increasing λ reduces the saturation time of the MSD.

The decrease in the fractional order α , leads to a faster saturation of the MSD, and to the disappearance of the oscillations.

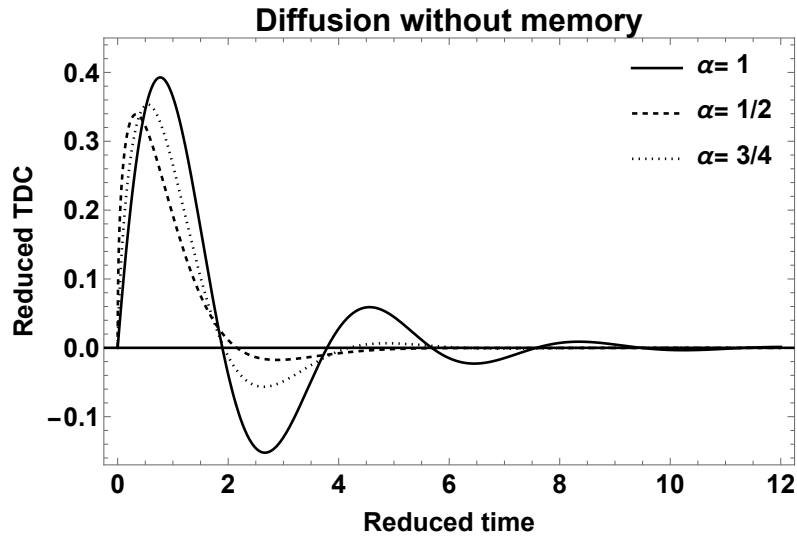


FIGURE 2. Graphical representation of the time diffusion coefficient (4.12) for : $\tau_\alpha/\tau_R = 0.05$; $\alpha = 1$; $\alpha = 1/2$ and $\alpha = 3/4$.

Remark 4.2. The graphical representation of the TDC clearly shows that this dynamical quantity tends to zero at infinity, since the motion of the target particle is completely blocked in the long term.

Figure (2) shows that as the fractional order α decreases, the TDC tends to zero more quickly, and increasing the parameter λ increases the oscillatory behavior.

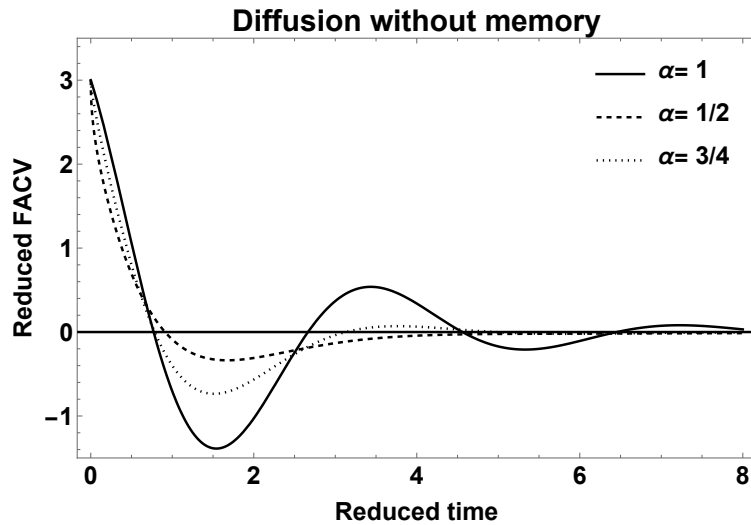


FIGURE 3. Graphical representation of the velocity autocorrelation function (4.13) for: $\tau_\alpha/\tau_R = 0.05$; $\alpha = 1$; $\alpha = 1/2$ and $\alpha = 3/4$.

Remark 4.3. The graphical representation of the VACF function shows that it tends to zero at infinity, because any correlation function must vanish at infinity.

The absence of force, i.e., $\lambda = 0$, causes the oscillatory behavior to disappear.

Figure (3) indicates that, as the fractional order α increases, or the parameter λ increases, oscillatory behavior appears.

4.2. Diffusion with memory.

It is noted that, for small radius or strong nanoparticle concentration, the target nanoparticle is trapped in a cage formed by the surrounding nanoparticles, and then, this nanoparticle executes rather a slow diffusion or subdiffusion. In this case, the memory function, $\gamma(t)$, is not trivial, and it is reasonable to choose for it a powers form of time, i.e. $\gamma(t)$, is a *powers function* of time, that is:

$$\gamma(t) = \frac{\gamma_\beta}{\Gamma(1-\beta)} t^{-\beta}, \quad 0 < \beta \leq 1. \quad (4.24)$$

Here $\gamma_\beta > 0$ is a *fractional relaxation rate*. To make a limit $\beta \rightarrow 1$, we rewrite the memory function as a derivation, in *distribution sense*:

$$\gamma(t) = \frac{\gamma_\beta}{\Gamma(2-\beta)} \frac{d}{dt} [t^{1-\beta} \theta(t)], \quad (4.25)$$

where $\theta(t)$ is the *Heaviside step function*. The limit $\beta \rightarrow 1$ then gives:

$$\gamma(t) \rightarrow \gamma_1 \delta(t), \quad \beta \rightarrow 1. \quad (4.26)$$

Laplace transform of memory function, defined in relation (4.24), is such that:

$$\hat{\gamma}(s) = \gamma_\beta s^{\beta-1}, \quad 0 < \beta \leq 1. \quad (4.27)$$

In this case, Laplace transform of the three relaxation functions, $K(t)$, $I(t)$ and $G(t)$ are as follows:

$$\hat{K}(s) = \frac{s^{\alpha-1}}{s^\alpha + \frac{m}{m_\alpha} \gamma_\beta s^{\beta-1} + \frac{\lambda}{m_\alpha} s^{-1}}, \quad (4.28)$$

$$\hat{I}(s) = \frac{s^{-2}}{s^\alpha + \frac{m}{m_\alpha} \gamma_\beta s^{\beta-1} + \frac{\lambda}{m_\alpha} s^{-1}}, \quad (4.29)$$

$$\hat{G}(s) = \frac{s^{-1}}{s^\alpha + \frac{m}{m_\alpha} \gamma_\beta s^{\beta-1} + \frac{\lambda}{m_\alpha} s^{-1}}. \quad (4.30)$$

As before, we formally develop them in powers of parameter, λ/m_α . Then, we have:

$$\hat{K}(s) = \sum_{n=0}^{+\infty} \left(-\frac{\lambda}{m_\alpha} \right)^n \frac{s^{-\beta(n+1)+\alpha}}{\left(s^{\alpha-\beta+1} + \frac{m}{m_\alpha} \gamma_\beta \right)^{n+1}}, \quad (4.31)$$

$$\hat{G}(s) = \sum_{n=0}^{+\infty} \left(-\frac{\lambda}{m_\alpha} \right)^n \frac{s^{-\beta(n+1)}}{\left(s^{\alpha-\beta+1} + \frac{m}{m_\alpha} \gamma_\beta \right)^{n+1}}, \quad (4.32)$$

$$\hat{I}(s) = \sum_{n=0}^{+\infty} \left(-\frac{\lambda}{m_\alpha} \right)^n \frac{s^{-\beta(n+1)-1}}{\left(s^{\alpha-\beta+1} + \frac{m}{m_\alpha} \gamma_\beta \right)^{n+1}}. \quad (4.33)$$

By inversion and using formula (2.15), we find the following expressions for the relaxation functions, $I(t)$, $G(t)$ and $K(t)$:

$$I(t) = t^{\alpha+1} \sum_{n=0}^{+\infty} \left[-\left(\frac{t}{\tau_R}\right)^{\alpha+1} \right]^n E_{\alpha-\beta+1, (\alpha+1)(n+1)+1}^{n+1} \left[-\left(\frac{t}{\tau_\beta}\right)^{\alpha-\beta+1} \right], \quad (4.34)$$

$$G(t) = t^\alpha \sum_{n=0}^{+\infty} \left[-\left(\frac{t}{\tau_R}\right)^{\alpha+1} \right]^n E_{\alpha-\beta+1, (\alpha+1)(n+1)}^{n+1} \left[-\left(\frac{t}{\tau_\beta}\right)^{\alpha-\beta+1} \right], \quad (4.35)$$

$$K(t) = \sum_{n=0}^{+\infty} \left[-\left(\frac{t}{\tau_R}\right)^{\alpha+1} \right]^n E_{\alpha-\beta+1, n(\alpha+1)+1}^{n+1} \left[-\left(\frac{t}{\tau_\beta}\right)^{\alpha-\beta+1} \right]. \quad (4.36)$$

with the time scale:

$$\tau_\beta = \left(\frac{m}{m_\alpha} \gamma_\beta \right)^{-1/(\alpha-\beta+1)}. \quad (4.37)$$

From the above series, we extract those of MSD, TDC and VACF and find:

$$\frac{W(t)}{W_0} = \left(\frac{t}{\tau_R}\right)^{\alpha+1} \sum_{n=0}^{+\infty} (-1)^n \left[\left(\frac{t}{\tau_R}\right)^{\alpha+1} \right]^n E_{\alpha-\beta+1, (\alpha+1)(n+1)+1}^{n+1} \left[-\left(\frac{t}{\tau_\beta}\right)^{\alpha-\beta+1} \right], \quad (4.38)$$

$$\frac{D(t)}{D_0} = \left(\frac{t}{\tau_1}\right)^\alpha \sum_{n=0}^{+\infty} (-1)^n \left[\left(\frac{t}{\tau_R}\right)^{\alpha+1} \right]^n E_{\alpha-\beta+1, (\alpha+1)(n+1)}^{n+1} \left[-\left(\frac{t}{\tau_\beta}\right)^{\alpha-\beta+1} \right], \quad (4.39)$$

$$\frac{c_{vv}(t)}{c_{vv}(0)} = \sum_{n=0}^{+\infty} \left[-\left(\frac{t}{\tau_R}\right)^{\alpha+1} \right]^n E_{\alpha-\beta+1, n(\alpha+1)+1}^{n+1} \left[-\left(\frac{t}{\tau_\beta}\right)^{\alpha-\beta+1} \right]. \quad (4.40)$$

Here

$$\tau_R = \left(\frac{\lambda}{m_\alpha} \right)^{-1/(\alpha+1)} = \left(\frac{dk_B T}{R^2 m_\alpha} \right)^{-1/(\alpha+1)}, \quad (4.41)$$

is a *second characteristic time*, which will be compared to the first one, τ_β , defined in formula (4.37). Since, constant λ is assumed to be small, we have necessarily $\tau_\beta < \tau_R$.

Amplitudes $W(t)/W_0$, D_0 and $c_{vv}(0)$ are respectively defined in relation (4.14) and $\tau_1 = \left(\frac{m\gamma_1}{m_\alpha} \right)^{1/\alpha}$.

Now, let us examine the time behaviors of dynamic quantities. To this end, we have to distinguish three kinds of time-regimes, which are the following:

Cas 1 : α -ballistic regime: For $t < \tau_\beta$, the dominant parts of dynamic quantities come from the value $n = 0$ in the associated series, and we find that:

$$W(t) = 2d \frac{k_B T}{m_\alpha} \frac{1}{\Gamma(\alpha+2)} t^{\alpha+1}, \quad (4.42)$$

$$D(t) = \frac{k_B T}{m_\alpha} \frac{1}{\Gamma(1+\alpha)} t^\alpha, \quad (4.43)$$

$$c_{vv}(t) = d \frac{k_B T}{m}. \quad (4.44)$$

As before, the introduction of the fractional derivative has affected the behavior of the quantities during the ballistic regime.

Cas 2 : Subdiffusion regime: By using the asymptotic behavior of M-L functions, we obtain for $\tau_\beta < t < \tau_R$ that :

$$W(t) = 2dD_\beta t^\beta, \quad (4.45)$$

$$D(t) = \beta D_\beta t^{\beta-1}, \quad (4.46)$$

$$c_{vv}(t) = 0. \quad (4.47)$$

Here

$$D_\beta = \frac{k_B T}{m\gamma_\beta \Gamma(\beta + 1)}, \quad (4.48)$$

accounts for the *generalized diffusion coefficient*.

The target particle executes a subdiffusion motion, because $W(t) \sim t^\beta$ and $\beta \leq 1$. Note that, in the limit $\beta \rightarrow 0$ we find the results of diffusion without memory, relations (4.18), (4.19) and (4.20).

Cas 3 : Saturation regime: By using the asymptotic behaviors of M-L functions, we get for $t > \tau_R$ that:

$$W(t) = 2R^2, \quad (4.49)$$

$$D(t) = 0, \quad (4.50)$$

$$c_{vv}(t) = 0. \quad (4.51)$$

As before, the MSD saturates at the finite value, $2R^2$.

Finally, we report in Figures (4), (5) and (6), the variation in time of the the reduced MSD, $W(t)/W_0$, the reduced TDC, $D(t)/D_0$ and the reduced VACF, $c_{vv}(t)/c_{vv}(0)$, versus the renormalized time, t/τ_α . for different values of α , and we will interpret the effect of the fractional order α and β on the behavior of dynamic quantities.

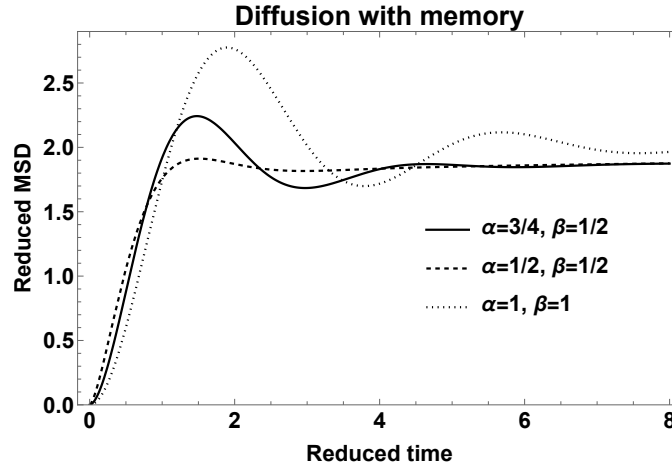


FIGURE 4. Graphical representation of the mean square displacement (4.38) for: $\tau_\alpha/\tau_R = 0.05$; $\alpha = 3/4, \beta = 1/2$; $\alpha = 1/2, \beta = 1/2$ and $\alpha = 3/4, \beta = 1/2$.

Remark 4.4. At the limit $\beta = 1$, we find the same behavior as in figure (1). Moreover, if the fractional order β decreases, the saturation of the MSD becomes faster and the oscillations disappear.

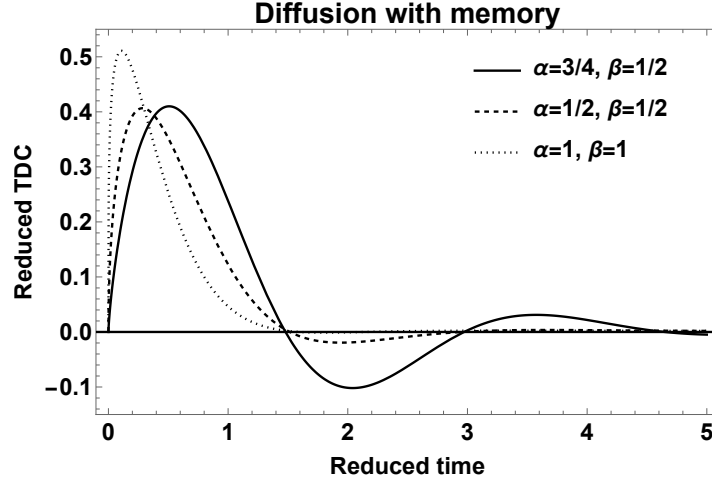


FIGURE 5. Graphical representation of the time diffusion coefficient (4.39) for: $\tau_\alpha/\tau_R = 0.05$; $\alpha = 3/4, \beta = 1/2$; $\alpha = 1/2, \beta = 1/2$ and $\alpha = 3/4, \beta = 1/2$.

Remark 4.5. In the limit $\beta = 1$, we find the same behavior as in figure (2). Moreover, if the fractional order β increases, the TDC quickly vanishes.

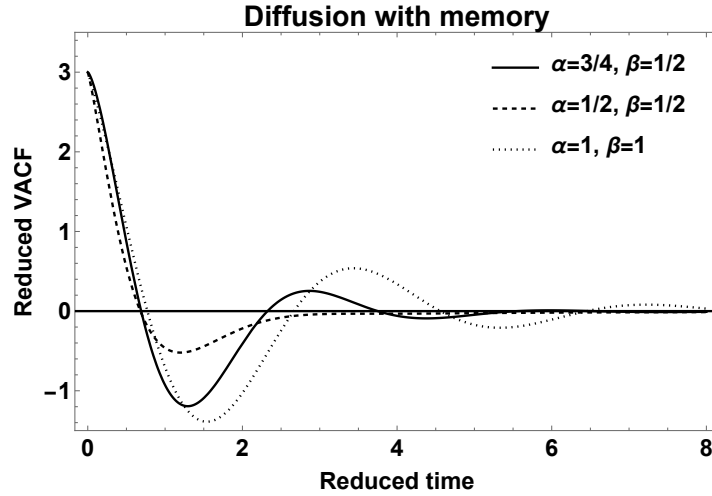


FIGURE 6. Graphical representation of the velocity autocorrelation function (4.40) for: $\tau_\alpha/\tau_R = 0.05$; $\alpha = 3/4, \beta = 1/2$; $\alpha = 1/2, \beta = 1/2$ and $\alpha = 3/4, \beta = 1/2$.

Remark 4.6. At the limit $\beta = 1$, we find the same behavior as in figure (3). Moreover, if the fractional order β increases, the oscillations become significant.

5. CONCLUSION

In this paper, we have investigated theoretically the diffusion of a target particle moving freely on a closed surface with its entities (molecules, traps and so on). It was assumed that this geometry is a crowded system, so that this diffusion is anomalous.

The receipt for this study is the use of the so-called generalized Langevin equation with fractional derivative. To do explicit calculation of the desired dynamic quantities, we have chosen a spherical geometry, with the aim of studying the drastic effects caused by the surface curvature on the time evolution of these physical quantities.

The condition of closed surfaces is essential to have a saturation of MSD, at larger times. For closed surfaces with arbitrary geometry, we easily show that the mean squared displacement is bounded from above by d^2 , where d is the greatest distance between points on these surfaces. For example, for spherical bodies, this distance simply equals their radius, R .

The main result is that, at shorter times, the target particle undergoes an anomalous diffusion due to the curvature of the space on which it moves. On the one hand, the fractional derivative affected the ballistic regime that we called α -ballistic regime. On the other hand, at larger times, the mean squared displacement saturates to a finite value, and in this case, the target particle could visit all points on the sphere.

Finally, it would be interesting to extend the present study to other curved geometries and generalize the investigation to the case where the anomalous diffusion process is governed by a fractional diffusion equation.

APPENDIX A. Identification of the Lagrange multiplier.

From expression (3.21), we obtain:

$$\begin{aligned} \langle \hat{\mathbf{r}}(s) \cdot \hat{\mathbf{r}}(s') \rangle &= \frac{R^2}{ss'} - \frac{\lambda R^2}{m} \left(\frac{\hat{I}(s)}{s'} + \frac{\hat{I}(s')}{s} \right) + \frac{\lambda^2 R^2}{m^2} \hat{I}(s) \hat{I}(s') + \hat{G}(s) \hat{G}(s') \langle \mathbf{v}_0^2 \rangle \\ &\quad + d \frac{k_B T}{m} \hat{G}(s) \hat{G}(s') \frac{\hat{\gamma}(s) + \hat{\gamma}(s')}{s + s'}. \end{aligned} \quad (\text{A.1})$$

We have used the result that (2.17):

$$\langle F(s) F(s') \rangle = dk_B T m \frac{\hat{\gamma}(s) + \hat{\gamma}(s')}{s + s'}, \quad (\text{A.2})$$

and expression:

$$\langle \mathbf{v}_0^2 \rangle = dk_B T / m. \quad (\text{A.3})$$

Now, in order to transform further the above equality, we first demonstrate that:

$$\hat{G}(s) \hat{G}(s') \frac{\hat{\gamma}(s) + \hat{\gamma}(s')}{s + s'} = \frac{\hat{I}(s)}{s'} + \frac{\hat{I}(s')}{s} - \frac{\hat{I}(s) + \hat{I}(s')}{s + s'} - \frac{\lambda}{m} \hat{I}(s) \hat{I}(s') - \hat{G}(s) \hat{G}(s'), \quad (\text{A.4})$$

and obtain:

$$\begin{aligned} \langle \hat{\mathbf{r}}(s) . \hat{\mathbf{r}}(s') \rangle &= \frac{R^2}{ss'} + \frac{\lambda}{m} \left(d \frac{k_B T}{m} - R^2 \right) \left(\frac{\hat{I}(s)}{s'} + \frac{\hat{I}(s')}{s} \right) + \frac{\lambda^2}{m^2} \left(R^2 - d \frac{k_B T}{\lambda} \right) \hat{I}(s) \hat{I}(s') \\ &\quad - d \frac{k_B T}{m} \frac{\hat{I}(s) + \hat{I}(s')}{s + s'}. \end{aligned} \quad (\text{A.5})$$

LT inverse of the above equality leads to the expression of the expected position correlation function:

$$\begin{aligned} \langle \mathbf{r}(t) . \mathbf{r}(t') \rangle &= R^2 + \frac{\lambda}{m} \left(d \frac{k_B T}{\lambda} - R^2 \right) [I(t) + I(t')] + \frac{\lambda^2}{m^2} \left(R^2 - d \frac{k_B T}{\lambda} \right) I(t) I(t') \\ &\quad - d \frac{k_B T}{m} I(|t - t'|). \end{aligned} \quad (\text{A.6})$$

For $t = t'$, we obtain

$$\langle \mathbf{r}^2(t) \rangle = R^2 + 2 \frac{\lambda}{m} \left(d \frac{k_B T}{\lambda} - R^2 \right) I(t) + \frac{\lambda^2}{m^2} \left(R^2 - d \frac{k_B T}{\lambda} \right) I^2(t). \quad (\text{A.7})$$

Since $\mathbf{r}^2(t) = R^2$, thus $\langle \mathbf{r}^2(t) \rangle = R^2$, we must have the following relation giving the Lagrange multiplier, namely:

$$\lambda = d \frac{k_B T}{R^2}. \quad (\text{A.8})$$

Availability of data and materials. No data was used for the research described in the article.

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¹ LABORATORY OF MATHEMATICS, ANALYSIS AND CONTROL OF DYNAMICAL SYSTEMS, DEPARTMENT OF MATHEMATICS, FACULTY OF SCIENCES, MOULAY ISMAIL UNIVERSITY, ZITOUNE, 11201, MEKNES, MOROCCO.

Email address: abdelg.daoudi@edu.umi.ac.ma

² DYNAMICS OF COMPLEX SYSTEMS TEAM, PHYSICS DEPARTMENT, FACULTY OF SCIENCES, P.O. BOX 11201, MEKNES, MOROCCO.

Email address: m.benhamou@umi.ac.ma

³ LABORATORY OF MATHEMATICS, ANALYSIS AND CONTROL OF DYNAMICAL SYSTEMS, DEPARTMENT OF MATHEMATICS, FACULTY OF SCIENCES, MOULAY ISMAIL UNIVERSITY, ZITOUNE, 11201, MEKNES, MOROCCO.

Email address: elkinani_67@yahoo.com