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Study of Diffusion of Nanoparticles on Bounded Curved Surfaces from Langevin Equation Theory: Exact Results

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ABSTRACT: This work deals with an extended mathematical study of the diffusion laws of a target particle which moves on a bounded curved surface. For explicit calculations, we considered a sphere of small radius, R. Dynamics of the target particle was studied considering three physical quantities : The mean square displacement (MSD), the time diffusion coefficient (TDC) and the velocity autocorrelation function (VACF). The study is conducted within the framework of the Langevin equation theory and Laplace transform techniques. In this study, we distinguished two regimes, namely *inertial* and *non-inertial* regimes. For the two regimes and for normal and anomalous diffusions, we found that, at large times, the movement of the target particle is completely blocked, and then, MSD saturates to a finite value, i.e. $2R^2$. Finally, the obtained results suggest that the surface curvature induces drastic changes of the diffusion laws in comparison with the diffusion laws in infinite spaces.

Key Words: Nanoparticles, diffusion, curved spaces, Langevin equation theory.

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1. Introduction

The diffusion is a very general phenomenon producing in nature, [1,2,3] and which corresponds to the spreading of species as colloidal particles, atoms or molecules in some media, due to an energetic excitation provided by heat. Depending on the structure of the host environment in which these species move, their spread may be fast or slow. At room temperature, the diffusion phenomenon is very significant in a gas, weaker in a liquid and practically absent in a solid. For example, to observe a diffusion phenomenon in a solid or in a crystal, it is necessary to bring the material to higher temperatures which are close to $1000^{\circ}C$.

We recall that the diffusion phenomena were first studied by a macroscopic approach, such as *Fick's laws*, [4] or by a statistical approach based on the *diffusion equation* (random walk model) [5,6]. We emphasize that many experiments have showed the limits of diffusion models based on Fick's laws.

It is noted that there exists another mechanistic approach introduced by Langevin, [3,5] which is physically equivalent to the diffusion equation approach. More exactly, Langevin [3] had written a classical equation of motion of the moving target particle, where in addition to the friction force and the external force derived from a potential energy, the author had introduced a random or stochastic force (*unknown* force) exerted by the molecules of the ambient environment on this target particle. It is this approach that we used in our investigation.

Much later, two famous physicists, Kubo [5] and Zwanzig [7] had generalized the Langevin's work, by writing a so-called *Generalized Langevin Equation* (GLE) to quantitatively study the diffusion phenomena with memory (anomalous diffusions). Mathematically speaking, this GLE is an integro-differential equation.

In fact, we distinguish *two* types of diffusion: normal (or *Brownian*) diffusion taking place in a medium which is not complex (gas or liquids of low viscosity) and anomalous diffusion producing in complex (or *crowded*) media. The first diffusion is fast, while the second is very slow due to the extreme difficulty that the target particle moves, due to the presence of many mobile or immobile traps.

For normal diffusions, MSD(or*position variance*) of the target particle, increases linearly with time. This increasing law was found to be in good agreement with experimental observations, for the most diffusion processes. More exactly, in this case, MSD scales with time as

$$MSD = 2dD_0 t \, . \tag{1.1}$$

Such a relationship constitutes the *Einstein's formula*, where D_0 accounts for the *diffusion coefficient*, expressed in m^2/s unit, $\mathbf{r}(t) \in \mathbf{R}^d$ for the instantaneous position-vector of the considered target particle, t for time and d for the Euclidean dimension of the host medium. This process is often called Gaussian process, because the associated probability density is Gaussian. We note that the above behavior takes place for long times, that is to say beyond the relaxation time, depending on the details of the diffusion process and the physical characteristics of the ambient environment.

For an anomalous diffusion, however, MSD obeys, at large time, an unusual power law, i.e.

$$MSD = 2dD_\alpha t^\alpha \, . \tag{1.2}$$

Here the *anomalous exponent*, $0 < \alpha < 1$, for *subdiffusions*, and $1 < \alpha < 2$, for *superdiffusions*, but α may take values greater than or equal to 2. For example, $\alpha = 2$ corresponds to *ballistic diffusions*, and $\alpha = 3$, to *diffusions of Richardson type* (as atmospheric turbulences). There D_α, expressed in m^2/s^α unit, accounts for the *generalized diffusion coefficient* or *fractal diffusion coefficient* (if the host media have a fractal geometric structure).

We emphasize that the anomalous diffusion is not a Markov process, contrary to the normal diffusion. This results is due to the fact that the probability density is not Gaussian.

It is noted that the anomalous diffusion is inherent in disordered or heterogeneous systems, [8,9,10,11] such as colloidal solutions, fractals, polymer melts, gels, living systems, transport of charge carriers in amorphous semiconductors, percolation structures, polymer networks, and generally complex systems containing entities moving on different time scales. For example, when the target particle moves in a fractal structure of *fractal dimension*, d_f, and *spectral dimension*, d_s, the associated subdiffusion exponent is $\alpha = 2/d_w$, where $d_w = 2d_f/d_s$ is the *walk dimension*, and for a time-fractional Brownian diffusion,

$\alpha = 2H$, where H is the *Hurst number*. Also, we note that the case with $1 < \alpha < 2$ describes the super-diffusion, occurring in turbulent plasmas, Lévy flights, transport in polymers, biological media, electrons in porous media, colloidal suspensions, under certain conditions, granular media and so on. In any case, the subdiffusion or the superdiffusion exponent, α, is not a universal quantity, but it strongly depends on the relevant model parameters.

The influence of the geometry (curvature) 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, cell-membranes or vesicles surfaces. The diffusion of the target particle on surfaces with curvature, such as spheres, ellipsoids, torus, etc., is a daunting task [19,20].

The goal of the present work was precisely a quantitative study of the influence of the surface curvature on the diffusion laws concerning a target particle which moves on a *closed smooth surface*, in the presence of mobile or immobile traps located on this surface. Intuitively, the main characteristic is that, the diffusion is blocked at large time, whatever is the geometrical structure of the closed smooth surface. As result, MSD is bounded by the maximal distance between points on the surface. To do explicit calculations, we considered spherical soft surfaces, as bubbles surfaces, droplets surfaces, cell-membranes or vesicles (liposomes...) surfaces . For these closed geometries, the only pertinent geometrical parameter of discussion is the sphere radius, R.

For the sake of generalization, we have studied the diffusion processes producing on a sphere, without memory (normal diffusions) and with memory (anomalous diffusions). For normal and anomalous diffusions, we have considered both initial and non-inertial regimes. The used theoretical method for the study of the formed was based on GLE, and as we shall see below, for some choice of the memory function, the obtained results are expressed in terms of Mittag-Leffler functions [12].

The remaining presentation of this paper is organized as follows. Section 2 deals with useful preliminaries. In Sections 3 and 4, we study the inertial and non-inertial diffusions, respectively. Finally, some concluding remarks are drawn in the last section.

2. Preliminaries

Definition 2.1 *For any arbitrary function, $f(t)$, defined in interval $t \geq 0$, its Laplace transform (LT) is*

$$\widehat{f}(s) = \int_0^\infty dt e^{-st} f(t) \ . \tag{2.1}$$

Its inverse LT is then

$$f(t) = \frac{1}{2\pi i} \oint_\Gamma ds e^{st} \widehat{f}(s) \ . \tag{2.2}$$

Γ *stands for the* integration contour *in* complex plan, $\mathbf{C}$, *with $Re[s] > 0$. We recall that LT of a* convolution product, $(f * g)(t)$, *is simply $\widehat{f}(s) \times \widehat{g}(s)$, and that of the first derivation of any function, $f(t)$, is $s\widehat{f}(s) - f(0)$.*

Definition 2.2 *MSD of a target particle, denoted as $W(t)$, is defined by*

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

Here $\mathbf{r}(t) \in \mathbf{R}^d$ is its instantaneous position-vector in Euclidean space, of dimension d. Naturally, we have: $W(t) > 0$, for all time $t > 0$, and $W(0) = 0$ (absence of the movement at initial time $t = 0$). Depending on the nature of the diffusion process, $W(t)$ is bounded or not at large time, that is for $t \to +\infty$.

Definition 2.3 *We define VACF, denoted as $c_{vv}(t)$, by*

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

Here $\mathbf{v}(t) \in \mathbf{R}^d$ *is the instantaneous velocity of the target particle. At the initial time, that is for* $t = 0$*, we have* $c_{vv}(0) = \langle \mathbf{v}_0^2 \rangle$*, where* $\mathbf{v}_0 \in \mathbf{R}^d$ *is the initial velocity of the target particle. Like any correlation function, VACF must vanish at infinite time, i.e.* $c_{vv}(t) \to 0$*, for* $t = +\infty$*. Of course, the behavior form of VACF, at large time limit, depends mainly on the nature of the diffusion process. In some cases, VACF may posses finite or infinite zeros (oscillations).*

Definition 2.4 *We define the* time diffusion coefficient *(TDC), denoted as* $D(t)$*, by*

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

Proposition 2.1 *LT of the stationary correlation function,*

$$\langle f(t) f(t') \rangle = \Phi(|t - t'|) \, . \tag{2.6}$$

of any arbitrary function, $f(t)$*, is [13]*

$$\left\langle \widehat{f}(s) \widehat{f}(s') \right\rangle = \frac{\widehat{\Phi}(s) + \widehat{\Phi}(s')}{s + s'} \, . \tag{2.7}$$

Here $\widehat{\Phi}(s)$ *is LT of function* $\Phi(t)$*.*

3. Inertial Diffusion Study

3.1. Inertial generalized stochastic equation

Consider a given target particle, of instantaneous position-vector, $\mathbf{r}(t) \in \mathbf{R}^3$, which moves on a sphere, of radius R. The equation of this sphere is:

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

This sphere may be rigid body or soft one as an interface between two immiscible liquids. The curved geometry imposes to the target particle a *constraint force*, $\mathbf{F}_{ext}(\mathbf{r})$, along the perpendicular direction to the surface, preventing it from leaving the sphere. The expression of such a force can be inspired from the *Analytic Mechanics Principles*, and we have

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

Here λ is a *positive* factor (*Lagrange multiplier*) which will be identified below. Then, the external force plays the role of an *elastic force*. We shall denote by $\mathbf{r}_0 = \mathbf{r}(0) \in \mathbf{R}^3$, the initial position of the target particle on the sphere.

The diffusion process (with memory) we are interested in can be described using GLE, due to Kubo [5] and Zwanzig [7]

$$m \frac{d\mathbf{v}(t)}{dt} = -m \int_0^t dt' \gamma(t - t') \mathbf{v}(t') - \lambda \mathbf{r}(t) + \mathbf{F}(t) \, . \tag{3.3}$$

Here $\mathbf{v}(t) \in \mathbf{R}^3$ denotes the instantaneous velocity of the target particle and $\gamma(t)$ is the instantaneous *memory function*, expressing a delay of frictions with the molecules or other particles (traps) located on the spherical surface, and $\mathbf{F}(t) \in \mathbf{R}^3$ is the *stochastic force* (*Langevin force*), obeying the following *three* rules

$$\langle \mathbf{F}(t) \rangle = 0 \, , \qquad t > 0 \, . \tag{3.4}$$

$$\langle \mathbf{F}(t) . \mathbf{F}(t') \rangle = 3 k_B T m \gamma(|t - t'|) \, . \tag{3.5}$$

$$\langle \mathbf{v}(0) . \mathbf{F}(t) \rangle = 0 \,. \tag{3.6}$$

Condition (3.6) 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 particles.

We note that the stochastic equation (3.3) must be supplemented by taking into account the fact that the target particle moves but remains all the time on the surface of the sphere, of equation

$$\mathbf{r}^2(t) = R^2 \,, \qquad\qquad t \geq 0 \,. \tag{3.7}$$

Par simple derivation of the above relation, with respect to time, we find the equivalent condition

$$\mathbf{r}(t) . \mathbf{v}(t) = 0 \,, \qquad\qquad t \geq 0 \,. \tag{3.8}$$

We note that the diffusion process without memory corresponds to the particular form of the memory function, $\gamma(t) = \gamma_1 \delta(t)$.

Now, scalarly multiplying the stochastic equation (3.3) by $\mathbf{v}(0)$ and making a time-average over the random force, $\mathbf{F}(t)$, gives the following integro-differential equation satisfied by VACF,

$$\frac{dc_{vv}(t)}{dt} = -\int_0^t dt' \left[\gamma(t-t') + \frac{\lambda}{m} \theta(t-t') \right] c_{vv}(t') \,. \tag{3.9}$$

Here $\theta(t)$ denotes the *Heaviside step function.* To derive dynamic equation (3.9), we have used the trivial identity

$$\mathbf{r}(t) - \mathbf{r}_0 = \int_0^t dt \mathbf{v}(t) \,. \tag{3.10}$$

and the fact that $\mathbf{r}_0 . \mathbf{v}_0 = 0$, since the particle moves on the sphere, all the time.

Laplace transforming integro-differential equation (3.9) leads to LT of VACF, namely

$$\widehat{c}_{vv}(s) = \left\langle \mathbf{v}_0^2 \right\rangle \widehat{g}(s) \,. \tag{3.11}$$

with

$$\widehat{g}(s) = \frac{1}{s + \widehat{\gamma}(s) + \frac{\lambda}{m} s^{-1}} \,. \tag{3.12}$$

which is LT of relaxation function, $g(t)$. We have used the fact that LT of the first derivative of VACF, $c_{vv}(t)$, is $s\widehat{c}_{vv}(s) - \left\langle \mathbf{v}_0^2 \right\rangle$. Formula (3.11) implies that

$$c_{vv}(t) = \left\langle \mathbf{v}_0^2 \right\rangle g(t) \,. \tag{3.13}$$

Notice that $g(0) = 1$, for all diffusion processes, since $c_{vv}(0) = \left\langle \mathbf{v}_0^2 \right\rangle$.

3.2. Basic relationships

3.2.1. Formal expression of velocity.

Laplace transforming stochastic equation (3.3) gives LT of velocity,

$$\widehat{\mathbf{v}}(s) = \widehat{g}(s) \mathbf{v}_0 - \frac{\lambda}{m} \widehat{G}(s) \mathbf{r}_0 + \frac{1}{m} \widehat{g}(s) \widehat{\mathbf{F}}(s) \,. \tag{3.14}$$

with

$$\widehat{G}(s) = s^{-1} \widehat{g}(s) = \frac{s^{-1}}{s + \widehat{\gamma}(s) + \frac{\lambda}{m} s^{-1}} \,. \tag{3.15}$$

which represents LT of the second relaxation function, $G(t)$.

Inverse LT of equality (3.14) yields the instantaneous velocity of the target particle, namely

$$\mathbf{v}(t) = g(t) \mathbf{v}_0 - \frac{\lambda}{m} G(t) \mathbf{r}_0 + \frac{1}{m} \int_0^t g(t-t') \mathbf{F}(t') dt' \,. \tag{3.16}$$

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

$$\langle \mathbf{v}(t)\rangle = g(t)\langle \mathbf{v}_0\rangle - \frac{\lambda}{m}G(t)\,\mathbf{r}_0 \,. \tag{3.17}$$

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

3.2.2. Formal expression of velocity correlation function.

From relation (3.14) and by averaging on the random force, we get

$$\begin{aligned}\langle \mathbf{v}(s).\mathbf{v}(s')\rangle &= \widehat{g}(s)\widehat{g}(s')\left\langle \mathbf{v}_0^2\right\rangle + \frac{\lambda^2R^2}{m^2}\widehat{G}(s)\widehat{G}(s') \\ &+\frac{1}{m^2}\widehat{g}(s)\widehat{g}(s')\langle \mathbf{F}(s).\mathbf{F}(s')\rangle \,. \end{aligned} \tag{3.18}$$

We have used identity: $\mathbf{r}_0.\mathbf{v}_0=0$. Applying general formula (2.7) to function $\Phi(t)=3k_BTm\gamma(t)$ yields

$$\langle \mathbf{F}(s).\mathbf{F}(s')\rangle = 3k_BTm\frac{\widehat{\gamma}(s)+\widehat{\gamma}(s')}{s+s'} \,. \tag{3.19}$$

Explicitly, we obtain

$$\begin{aligned}\langle \mathbf{v}(s).\mathbf{v}(s')\rangle &= \widehat{g}(s)\widehat{g}(s')\left\langle \mathbf{v}_0^2\right\rangle + \frac{\lambda^2R^2}{m^2}\widehat{G}(s)\widehat{G}(s') \\ &+3\frac{k_BT}{m}\widehat{g}(s)\widehat{g}(s')\frac{\widehat{\gamma}(s)+\widehat{\gamma}(s')}{s+s'} \,. \end{aligned} \tag{3.20}$$

Now, we try to transform the last contribution in the above expression. To this end, we first write

$$\begin{aligned}&\widehat{g}(s)\widehat{g}(s')\frac{\widehat{\gamma}(s)+\widehat{\gamma}(s')}{s+s'} \\ &= \widehat{g}(s)\widehat{g}(s')\frac{\widehat{g}^{-1}(s)+\widehat{g}^{-1}(s')-\left(s+s'+\frac{\lambda}{m}\left(s^{-1}+s'^{-1}\right)\right)}{s+s'} \,. \end{aligned} \tag{3.21}$$

Some algebra gives

$$\widehat{g}(s)\widehat{g}(s')\frac{\widehat{\gamma}(s)+\widehat{\gamma}(s')}{s+s'} = \frac{\widehat{g}(s)+\widehat{g}(s')}{s+s'} - \widehat{g}(s)\widehat{g}(s') - \frac{\lambda}{m}\widehat{G}(s)\widehat{G}(s') \,. \tag{3.22}$$

and then,

$$\begin{aligned}\langle \widehat{\mathbf{v}}(s).\widehat{\mathbf{v}}(s')\rangle &= \left(\left\langle \mathbf{v}_0^2\right\rangle - 3\frac{k_BT}{m}\right)\widehat{g}(s)\widehat{g}(s') + \frac{\lambda^2}{m^2}\left(R^2-3\frac{k_BT}{\lambda}\right)\widehat{G}(s)\widehat{G}(s') \\ &+3\frac{k_BT}{m}\frac{\widehat{g}(s)+\widehat{g}(s')}{s+s'} \,. \end{aligned} \tag{3.23}$$

Inverse LT of the above equality leads to the general expression of the desired velocity correlation function,

$$\begin{aligned}\langle \mathbf{v}(t).\mathbf{v}(t')\rangle &= \left(\left\langle \mathbf{v}_0^2\right\rangle - 3\frac{k_BT}{m}\right)g(t)g(t') + \frac{\lambda^2}{m^2}\left(R^2-3\frac{k_BT}{\lambda}\right)G(t)G(t') \\ &+3\frac{k_BT}{m}g(|t-t'|) \,. \end{aligned} \tag{3.24}$$

Such an expression suggests that this correlation function is not stationary, due to the two first terms, but the last one is rather a stationary function, since it is invariant under translations of time.

The above obtained velocity correlation function is simplified, when the initial velocities obey the Maxwell distribution, and we have $\left\langle \mathbf{v}_0^2\right\rangle = 3k_BT/m$. Therefore, in this case, we get

$$\langle \mathbf{v}(t).\mathbf{v}(t')\rangle = \frac{\lambda^2}{m^2}\left(R^2-3\frac{k_BT}{\lambda}\right)G(t)G(t') + 3\frac{k_BT}{m}g(|t-t'|) \,. \tag{3.25}$$

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 (3.14) allows the following formula

$$\widehat{\mathbf{r}}(s) = \frac{\mathbf{r}_0}{s} + \widehat{G}(s)\mathbf{v}_0 - \frac{\lambda}{m}\widehat{I}(s)\mathbf{r}_0 + \frac{1}{m}\widehat{G}(s)\widehat{\mathbf{F}}(s) \ . \tag{3.26}$$

with the notation

$$\widehat{I}(s) = s^{-1}\widehat{G}(s) = s^{-2}\widehat{g}(s) = \frac{s^{-2}}{s + \widehat{\gamma}(s) + \frac{\lambda}{m}s^{-1}} \ . \tag{3.27}$$

which is LT of relaxation function, $I(t)$.

Inverse LT of equality (3.26) gives the expression of the time evolution of the instantaneous position, that is

$$\mathbf{r}(t) = \mathbf{r}_0 + G(t)\mathbf{v}_0 - \frac{\lambda}{m}I(t)\mathbf{r}_0 + \frac{1}{m}\int_0^t dt' G(t-t')\mathbf{F}(t') \ . \tag{3.28}$$

Averaging the above equality on the random force, $\mathbf{F}(t)$, leads to the time-average of position,

$$\langle \mathbf{r}(t)\rangle = \left(1 - \frac{\lambda}{m}I(t)\right)\mathbf{r}_0 + G(t)\langle \mathbf{v}_0\rangle \ . \tag{3.29}$$

The above result clearly indicates that the time-average, $\langle \mathbf{r}(t)\rangle$, belongs to the 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: $I(0) = 0$, whatever is the kind of the diffusion process.

3.2.4. Formal expression of position correlation function.

From expression (3.26), we obtain

$$\begin{aligned}\langle \widehat{\mathbf{r}}(s).\widehat{\mathbf{r}}(s')\rangle &= \frac{R^2}{ss'} - \frac{\lambda R^2}{m}\left[\frac{\widehat{I}(s)}{s'} + \frac{\widehat{I}(s')}{s}\right] + \frac{\lambda^2 R^2}{m^2}\widehat{I}(s)\widehat{I}(s') \\ &+ \widehat{G}(s)\widehat{G}(s')\left\langle \mathbf{v}_0^2\right\rangle + 3\frac{k_B T}{m}\widehat{G}(s)\widehat{G}(s')\frac{\widehat{\gamma}(s) + \widehat{\gamma}(s')}{s+s'} \ . \end{aligned} \tag{3.30}$$

We have used relation (3.19) defining the quantity $\left\langle \widehat{\mathbf{F}}(s).\widehat{\mathbf{F}}(s')\right\rangle$ and expression $\left\langle \mathbf{v}_0^2\right\rangle = 3k_B T/m$.

On the other hand, we easily show the following equality

$$\begin{aligned}\widehat{G}(s)\widehat{G}(s')\frac{\widehat{\gamma}(s) + \widehat{\gamma}(s')}{s+s'} &= \frac{\widehat{I}(s)}{s'} + \frac{\widehat{I}(s')}{s} - \frac{\widehat{I}(s) + \widehat{I}(s')}{s+s'} \\ &- \frac{\lambda}{m}\widehat{I}(s)\widehat{I}(s') - \widehat{G}(s)\widehat{G}(s') \ . \end{aligned} \tag{3.31}$$

and get

$$\begin{aligned}\langle \widehat{\mathbf{r}}(s).\widehat{\mathbf{r}}(s')\rangle &= \frac{R^2}{ss'} - \frac{\lambda R^2}{m}\left[\frac{\widehat{I}(s)}{s'} + \frac{\widehat{I}(s')}{s}\right] + \frac{\lambda^2}{m^2}\left(R^2 - 3\frac{k_B T}{\lambda}\right)\hat{I}(s)\hat{I}(s') \\ &+ 3\frac{k_B T}{m}\left[\frac{\widehat{I}(s)}{s'} + \frac{\widehat{I}(s')}{s} - \frac{\widehat{I}(s) + \widehat{I}(s')}{s+s'}\right] \ . \end{aligned} \tag{3.32}$$

Inverse LT of the above equality leads to the expression of the expected position correlation function,

$$\begin{aligned}\langle \mathbf{r}(t).\mathbf{r}(t')\rangle &= R^2 + \frac{\lambda}{m}\left(3\frac{k_B T}{\lambda} - R^2\right)[I(t) + I(t')] \\ &+ \frac{\lambda^2}{m^2}\left(R^2 - 3\frac{k_B T}{\lambda}\right)I(t)I(t') - 3\frac{k_B T}{m}I(|t-t'|) \ . \end{aligned} \tag{3.33}$$

For $t = t'$, we obtain

$$\left\langle \mathbf{r}^2(t)\right\rangle = R^2 + 2\frac{\lambda}{m}\left(3\frac{k_BT}{\lambda} - R^2\right) I(t) + \frac{\lambda^2}{m^2}\left(R^2 - 3\frac{k_BT}{\lambda}\right) I^2(t) \ . \tag{3.34}$$

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

$$\lambda = 3\frac{k_BT}{R^2} \ . \tag{3.35}$$

The position correlation function is then given by

$$\left\langle \mathbf{r}(t) . \mathbf{r}(t')\right\rangle = R^2 - 3\frac{k_BT}{m} I(|t-t'|) \ . \tag{3.36}$$

Therefore, on a sphere, both velocity and position correlation functions are stationary functions.

Setting $t' = 0$ in the above equality, to get

$$\left\langle \mathbf{r}(t) . \mathbf{r}(0)\right\rangle = R^2 - 3\frac{k_BT}{m} I(t) \ . \tag{3.37}$$

In definitive, we find that MSD writes as

$$W(t) = \mathbf{r}_0^2 + \left\langle \mathbf{r}^2(t)\right\rangle - 2\left\langle \mathbf{r}(t) . \mathbf{r}(0)\right\rangle = 6\frac{k_BT}{m} I(t) \ . \tag{3.38}$$

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

$$D(t) = \frac{1}{6}\frac{dW(t)}{dt} = \frac{k_BT}{m} G(t) \ . \tag{3.39}$$

3.2.5. Short and large times behaviors of dynamic quantities.

According to *Complex Analysis*, the behavior of a function, $f(t)$, for $t \to +\infty$, can be extracted from that of its LT, $\widehat{f}(s)$, for $s \to 0$, and the behavior of LT, $\widehat{f}(s)$, for $s \to +\infty$, can be deduced from that of function, $f(t)$, for $t \to 0$.

Therefore, in the limit $s \to 0$, we expect the following behaviors, for any choice of the memory function,

$$\widehat{I}(s) \sim \frac{m}{\lambda} s^{-1} \ , \qquad \widehat{W}(s) = 2\left\langle \mathbf{v}_0^2\right\rangle \widehat{I}(s) \sim 6\frac{k_BT}{\lambda} s^{-1} \ . \tag{3.40}$$

$$\widehat{G}(s) \sim \frac{m}{\lambda} \ , \qquad \widehat{D}(s) = \frac{k_BT}{m}\widehat{G}(s) \sim \frac{k_BT}{\lambda} \ . \tag{3.41}$$

$$\widehat{g}(s) \sim \frac{m}{\lambda} s \ , \qquad \widehat{c}_{vv}(s) = 3\frac{k_BT}{m}\widehat{g}(s) \sim 3\frac{k_BT}{\lambda} s \ . \tag{3.42}$$

provided that the condition: $s\widehat{\gamma}(s) \to 0$, as $s \to 0$, is ensured. Inverse LT gives the large-time behaviors (*saturation regime*) for MSD, TDC and VACF, namely

$$W(t) \to 2R^2 \ , \qquad t \to +\infty \ . \tag{3.43}$$

$$D(t) \to 0 \ , \qquad t \to +\infty \ . \tag{3.44}$$

$$c_{vv}(t) \to 0 \ , \qquad t \to +\infty \ . \tag{3.45}$$

Therefore, as it should be, MSD is a bounded function from above and saturates to the finite value $2R^2$, at infinite time. In fact, this result is not surprising, since the area of the sphere is finite. We emphasize that such a limit value can be obtained rapidly noting that, for $t \to +\infty$, the position correlation function, $\left\langle \mathbf{r}(t) . \mathbf{r}(0)\right\rangle$, tends to 0, and according to formula (3.38), MSD goes, in this same limit, to $\mathbf{r}_0^2 + \left\langle \mathbf{r}^2(t)\right\rangle = 2R^2$.

For $s \to +\infty$, the denominator of LT of relaxation functions are dominated by the term s, provided that the condition, $s^{-1}\widehat{\gamma}(s) \to +\infty$, as $s \to +\infty$, is fulfilled, and we have the following behaviors

$$\widehat{I}(s) \sim s^{-3}\,, \qquad \widehat{W}(s) = 2\left\langle \mathbf{v}_0^2\right\rangle \widehat{I}(s) \sim 2\left\langle \mathbf{v}_0^2\right\rangle s^{-3}\,. \tag{3.46}$$

$$\widehat{G}(s) \sim s^{-2}\,, \qquad \widehat{D}(s) = \frac{1}{3}\left\langle \mathbf{v}_0^2\right\rangle \widehat{G}(s) \sim \frac{1}{3}\left\langle \mathbf{v}_0^2\right\rangle s^{-2}\,. \tag{3.47}$$

$$\widehat{g}(s) \sim s^{-1}\,, \qquad \widehat{c}_{vv}(s) = \left\langle \mathbf{v}_0^2\right\rangle \widehat{g}(s) \sim \left\langle \mathbf{v}_0^2\right\rangle s^{-1}\,. \tag{3.48}$$

Inverse LT allows the behaviors of dynamic quantities of interest, at small time,

$$W(t) = \left\langle \mathbf{v}_0^2\right\rangle t^2\,, \qquad t \to 0\,. \tag{3.49}$$

$$D(t) = \frac{1}{3}\left\langle \mathbf{v}_0^2\right\rangle t\,, \qquad t \to 0\,. \tag{3.50}$$

$$c_{vv}(t) = \left\langle \mathbf{v}_0^2\right\rangle = 3\frac{k_B T}{m}\,, \qquad t \to 0\,. \tag{3.51}$$

In fact, these behaviors characterize the so-called *ballistic regime*. Therefore, at the beginning times, the target particle does not felt the presence of the sphere curvature.

To summarize, we list below the essential results dealt with diffusions on a sphere, for any form of the memory function,

$$\left\langle \mathbf{v}(t)\,.\mathbf{v}(t')\right\rangle = \left\langle \mathbf{v}_0^2\right\rangle g(|t-t'|)\,, \qquad c_{vv}(t) = \left\langle \mathbf{v}(t)\,.\mathbf{v}(0)\right\rangle = \left\langle \mathbf{v}_0^2\right\rangle g(t)\,,$$

$$\left\langle \mathbf{r}(t)\,.\mathbf{r}(t')\right\rangle = R^2 - \left\langle \mathbf{v}_0^2\right\rangle I(|t-t'|)\,, \qquad \left\langle \mathbf{r}(t)\,.\mathbf{r}(0)\right\rangle = R^2 - \left\langle \mathbf{v}_0^2\right\rangle I(t)\,,$$

$$W(t) = 2\left\langle \mathbf{v}_0^2\right\rangle I(t)\,, \qquad D(t) = \frac{1}{6}\frac{dW(t)}{dt} = \frac{1}{3}\left\langle \mathbf{v}_0^2\right\rangle G(t)\,.$$

These various dynamic quantities are calculated by the only knowledge of the relaxation function, $g(t)$, because of the relationships

$$G(t) = \int_0^t g(t')\,dt'\,, \qquad I(t) = \int_0^t G(t')\,dt'.$$

We recall that $g(1) = 1$, $G(0) = 0$ and $I(0) = 0$, for all diffusion processes.

Finally, we note that the relaxation function, $g(t)$, will be calculated through its LT, $\widehat{g}(s)$, defined in relation (3.12), with $\lambda = 3k_B T/R^2$. Expression of $\widehat{g}(s)$ contains LT of the memory function, $\widehat{\gamma}(s)$. Therefore, expression $g(t)$ depends mainly on the particular choice of the form of the memory function, thus on the adequate model used for the description of the diffusion process.

3.2.6. Inertial normal diffusion in an infinite space.

Before accomplishing the study of the influence of the surface curvature on the diffusion process, it will be instructive to first recall the basic results dealt with a pure Brownian process. In this case, the random movement of the target particle obeys all the above derived basic relationships, but with a *local memory function*, i.e. $\gamma(t) = \gamma_1\delta(t)$, where the friction coefficient γ_1 is the *relaxation rate*. Its inverse, $\tau_1 = \gamma_1^{-1}$, denotes the *relaxation time*. The associated stochastic dynamic equation is simply given by

$$m\frac{d\mathbf{v}(t)}{dt} = -m\gamma_1\mathbf{v}(t) + \mathbf{F}(t)\,. \tag{3.52}$$

Here the stochastic force, $\mathbf{F}(t)$, obeys the three constraints (3.4), (3.5) and (3.6).

Therefore, in this particular case, LT of the relaxation functions are simply given by

$$\widehat{g}(s) = \frac{1}{\gamma_1 + s}\,. \tag{3.53}$$

$$\widehat{G}(s) = \frac{s^{-1}}{\gamma_1 + s} . \tag{3.54}$$

$$\widehat{I}(s) = \frac{s^{-2}}{\gamma_1 + s} . \tag{3.55}$$

Then, inverse LT gives MSD, TDC and VACF,

$$W(t) = 6\frac{k_B T}{m}\left(\frac{e^{-\gamma_1 t} - 1 + \gamma_1 t}{\gamma_1^2}\right) , \qquad t \geq 0 . \tag{3.56}$$

$$D(t) = \frac{k_B T}{m\gamma_1}\left(1 - e^{-\gamma_1 t}\right) , \qquad t \geq 0 . \tag{3.57}$$

$$c_{vv}(t) = \left\langle \mathbf{v}_0^2 \right\rangle e^{-\gamma_1 t} , \qquad \left\langle \mathbf{v}_0^2 \right\rangle = c_{vv}(0) = 3\frac{k_B T}{m} . \tag{3.58}$$

For shorter times (*ballistic regime*), i.e. for $t << \tau_1$, the above results stipulate that MSD, TDC and VACF behave according to the general behaviors (3.49), (3.50) and (3.51).

For larger times (*saturation regime*), i.e. for $t >> \tau_1$, these dynamic quantities behave rather as

$$W(t) = 6D_0 t , \qquad t >> \tau_1 . \tag{3.59}$$

$$D(t) = D_0 , \qquad t >> \tau_1 . \tag{3.60}$$

$$c_{vv}(t) = 0 , \qquad t >> \tau_1 . \tag{3.61}$$

Here $D_0 = k_B T/m\gamma_1$ accounts for the *bare diffusion coefficient*. Relation (3.59) is termed *Einstein's formula*, which results from a compromise between the thermal fluctuations (through the thermal energy, $k_B T$) and the dissipation (through the friction coefficient, γ_1).

3.2.7. Inertial normal diffusion on a sphere.

Now, consider a target particle which freely diffuses on a sphere, of radius, R. As in the last paragraph, the memory function is local, i.e. $\gamma(t) = \gamma_1 \delta(t)$. Then, its associated Standard Langevin Equation is the following

$$m\frac{d\mathbf{v}(t)}{dt} = -m\gamma_1 \mathbf{v}(t) - \lambda \mathbf{r} + \mathbf{F}(t) , \qquad \lambda = 3\frac{k_B T}{R^2} . \tag{3.62}$$

Also, the stochastic force, $\mathbf{F}(t)$, obeys the three constraints (3.4), (3.5) and (3.6). The above stochastic differential equation describes a *Langevin oscillator*, of *characteristic frequency*, $\omega_0 = \sqrt{\lambda/m}$. Explicitly, we have $\omega_0 = \sqrt{3k_B T/mR^2}$.

In this case, we have simply,

$$\widehat{c}_{vv}(s) = \left\langle \mathbf{v}_0^2 \right\rangle \frac{1}{s + \gamma_1 + \frac{\lambda}{m}s^{-1}} = \left\langle \mathbf{v}_0^2 \right\rangle \frac{s}{(s - s_1)(s - s_2)} . \tag{3.63}$$

with *two poles*

$$s_{1,2} = -\frac{\gamma_1}{2} \pm i\omega . \tag{3.64}$$

where the *frequency*, ω, is as follows

$$\omega = \sqrt{\frac{\lambda}{m} - \frac{\gamma_1^2}{4}} , \qquad \gamma_1 < 2\sqrt{\frac{\lambda}{m}} . \tag{3.65}$$

The above frequency may be rewritten as

$$\omega = \sqrt{\frac{3k_B T}{m}}\frac{1}{RR_c}\sqrt{R_c^2 - R^2} , \qquad R < R_c . \tag{3.66}$$

with the typical sphere radius, $R_c = \sqrt{12 k_B T / m\gamma_1^2}$. Therefore, VACF is obtained by inverse LT, and we find that

$$c_{vv}(t) = \left\langle \mathbf{v}_0^2 \right\rangle e^{-\gamma_1 t/2} \left\{ \cos(\omega t) - \frac{\gamma_1}{2\omega} \sin(\omega t) \right\} , \qquad R < R_c . \tag{3.67}$$

This expression clearly shows that VACF is a *quasi-periodic* function of time, t. This is the so-called *underdamped regime*, with $R < R_c$ (*small sphere*). The above expression indicates that VACF, $c_{vv}(t)$, has an *infinity of zeros*, for

$$\tan(\omega t_0) = 2\frac{\omega}{\gamma_1} . \tag{3.68}$$

The first zero is between 0 and $\pi/2\omega$, the second one, between $\pi/2\omega$ and $3\pi/2\omega$, and so on. Therefore, VACF *is not* a monotone function of time. Also, it exhibits an *infinity of minimums and maximums*, for

$$\tan(\omega t_m) = 2\frac{m\gamma_1\omega}{m\gamma_1^2 - 2\lambda} . \tag{3.69}$$

Comparing zeros equation (3.68) and extremums equation (3.69) and using the fact that function, $\tan(x)$, is an increasing function of its variable, we deduce that the first minimum is at the right of the first zero, the first maximum, at the right of the second zero, and so on. For *overdamped regime*, with $R > R_c$ (*big sphere*), we find that the associated VACF is given by

$$c_{vv}(t) = \left\langle \mathbf{v}_0^2 \right\rangle e^{-\gamma_1 t/2} \left\{ \cosh(\widehat{\omega} t) - \frac{\gamma_1}{2\widehat{\omega}} \sinh(\widehat{\omega} t) \right\} , \qquad R > R_c . \tag{3.70}$$

with a *second frequency*

$$\widehat{\omega} = \sqrt{3\frac{k_B T}{m}} \frac{1}{R R_c} \sqrt{R^2 - R_c^2} , \qquad R > R_c . \tag{3.71}$$

Expression (3.70) suggests that, for big spheres, i.e. $R > R_c$, VACF, $c_{vv}(t)$, possesses only *one zero*, t_0, for

$$\tanh(\omega t_0) = 2\frac{\widehat{\omega}}{\gamma_1} . \tag{3.72}$$

Also, VACF presents only *one minimum*, $t_{\min}$, for

$$\tanh(\widehat{\omega} t_{\min}) = 2\frac{m\gamma_1\widehat{\omega}}{2\lambda - m\gamma_1^2} . \tag{3.73}$$

The *critical regime* with $\gamma_1 = 2\sqrt{\lambda/m}$ or equivalently with $R = R_c$ (*medium sphere*) corresponds to the following VACF

$$c_{vv}(t) = \left\langle \mathbf{v}_0^2 \right\rangle e^{-\gamma_1 t/2} \left(1 - \frac{\gamma_1 t}{2} \right) , \qquad R = R_c . \tag{3.74}$$

Such a function has only *one zero*, for $t_0 = 2/\gamma_1$, and only *one extremum* which is a minimum, for $t_{\min} = 4/\gamma_1$.

Notice that, for all time-regimes we described above, $c_{vv}(t) \to 0^-$, for $t \to +\infty$.

MSD may also be calculated using LT techniques. Explicitly, we find that

$$\widehat{W}(s) = 6\frac{k_B T}{m} \frac{1}{s(s - s_1)(s - s_2)} . \tag{3.75}$$

An inverse LT leads to

$$W(t) = 2R^2 \left[1 - e^{-\gamma_1 t/2} \left\{ \cos(\omega t) + \frac{\gamma_1}{2\omega} \sin(\omega t) \right\} \right] , \qquad R < R_c . \tag{3.76}$$

for the *overdamped regime* (small sphere). It is straightforward to see that, at it should be, MSD is always positive definite, for all time. Of course, it has *no zero*, but exhibits an *infinity of extremums*, for $t_n = n\pi/\omega$, with $n \in \mathbf{N}$.

For the *underdamped regime* (big sphere), we find that MSD is the following

$$W(t) = 2R^2 \left[1 - e^{-\gamma_1 t/2}\left\{\cosh(\widehat{\omega}t) + \frac{\gamma_1}{2\widehat{\omega}}\sinh(\widehat{\omega}t)\right\}\right] , \qquad R > R_c . \tag{3.77}$$

We show that, in this case, MSD is also positive definite and has *one zero* which is at the same time a *minimum*, for $t = 0$. Then, MSD is an increasing monotone function of time.

For the *critical regime* (medium sphere), we find that

$$W(t) = 2R^2 \left[1 - e^{-\gamma_1 t/2}\left(1 + \frac{\gamma_1 t}{2}\right)\right] , \qquad R = R_c . \tag{3.78}$$

As in the *underdamped regime* case, MSD is positive definite and has *one zero* which is at the same time a *minimum*, for $t = 0$. Then, MSD is an increasing monotone function of time.

Now, we are interested in the determination of TDC expression. The latter can be obtained using its direct relation with MSD, relation (2.5). Then, we have

$$D(t) = \frac{k_B T}{m\omega}\sin(\omega t)\, e^{-\gamma_1 t/2} , \qquad R < R_c . \tag{3.79}$$

for the *overdamped regime*. The above expression indicates that TDC is a pseudo-periodic function of time, and remains confined between two exponential curves of equations $y = \pm k_B T e^{-\gamma_1 t/2}/m\omega$, for all time. In addition, it has an *infinity of zeros*, for $t_n = n\pi/\omega$, with $n \in \mathbf{N}$. Also, it exhibits an infinity for extremums, for

$$\tan(\omega t_m) = 2\frac{\omega}{\gamma_1} . \tag{3.80}$$

Notice that, as it should be, extremums of TDC coincide with zeros of VACF, and its zeros, with extremums of MSD.

For the *underdamped regime*, we find that

$$D(t) = \frac{k_B T}{m\widehat{\omega}}\sinh(\widehat{\omega}t)\, e^{-\gamma_1 t/2} , \qquad R > R_c . \tag{3.81}$$

Such an expression shows that TDC vanishes only for $t = 0$, and exhibits only *one extremum* which is a *minimum*, for

$$\tanh(\widehat{\omega}t_{\min}) = \frac{2\widehat{\omega}}{\gamma_1} . \tag{3.82}$$

As it should be, such a minimum coincides with the zero of MSD.

Finally, for the *critical regime*, we find that the associated TDC is as follows

$$D(t) = \frac{k_B T}{m} t e^{-\gamma_1 t/2} , \qquad R = R_c . \tag{3.83}$$

In this case, TDC vanishes, for $t = 0$, and exhibits only *one* extremum which is a *maximum*, for $t_{\max} = 2/\gamma_1$. As it should be, this maximum is identical to the only zero of VACF.

Let us comment more the above results.

Firstly, they clearly indicate that the diffusion on a sphere is different from a diffusion in an infinite space. Therefore, the surface curvature plays an important role and affects drastically the diffusion laws.

Secondly, at shorter times and for all regimes, the diffusion is ballistic. This means that, at early times, the target particle does not felt the curvature of the surface.

Thirdly, at very large time, that is for $t >> \tau_1 = 1/\gamma_1$, and for all regimes, MSD saturated to a finite value which equals $2R^2$. This result is not surprising, since the sphere area is finite, and then, after a time of the order of τ_1, all sites of this sphere are visited by the target particle.

Finally, we note that, in the limit $R \to \infty$ (absence of curvature), *Lagrange multiplier*, λ goes to 0, and then, the respective expressions (3.56), (3.57) and (3.58) of MSD, TDC and VACF are recovered.

3.2.8. Inertial subdiffusion in an infinite spaces.

Reconsider now GLE defined in relation (3.3) and assume that the memory function, $\gamma(t)$, is a *power function* of time, that is

$$\gamma(t) = \frac{\gamma_\alpha}{\Gamma(1-\alpha)} t^{-\alpha} \ , \qquad 0 < \alpha < 1 \, . \tag{3.84}$$

Here $\gamma_\alpha > 0$ is a *fractional relaxation rate* and $\Gamma(1-\alpha)$ denotes the *Euler gamma function* of argument $1-\alpha$ we have introduced for reasons commodity. The positivity of subdiffusion exponent, α, is compatible with the decreasing of the memory function in time. As we shall see below, the above chosen memory is compatible with the fact that MSD, $W(t)$, behaves at large time, i.e. $t \to \infty$, as: $W(t) \sim t^\alpha$.

It is noted that the limit $\alpha \to 1$ must be done carefully, by simply rewriting the memory function as a derivation, in *distribution sense*, that is

$$\gamma(t) = \frac{\gamma_\alpha}{\Gamma(2-\alpha)} \frac{d}{dt}\left[t^{1-\alpha}\theta(t)\right] . \tag{3.85}$$

where $\theta(t)$ is the *Heaviside step function*. The limit $\alpha \to 1$ then gives

$$\gamma(t) \to \gamma_1 \delta(t) \ , \qquad \alpha \to 1 \, . \tag{3.86}$$

We recover well the local memory function of a normal diffusion.

LT of the memory function, defined in relation (3.84), is

$$\widehat{\gamma}(s) = \gamma_\alpha s^{\alpha-1} \ , \qquad 0 < \alpha < 1 \, . \tag{3.87}$$

The above expression shows that $s\widehat{\gamma}(s) \to 0$, for $s \to 0$. Then, LTs of the three relaxation functions, $g(t)$, $G(t)$ and $I(t)$ are the following

$$\widehat{g}(s) = \frac{1}{s+\widehat{\gamma}(s)} = \frac{1}{s+\gamma_\alpha s^{\alpha-1}} \, . \tag{3.88}$$

$$\widehat{G}(s) = \frac{s^{-1}}{s+\widehat{\gamma}(s)} = \frac{s^{-1}}{s+\gamma_\alpha s^{\alpha-1}} \, . \tag{3.89}$$

$$\widehat{I}(s) = \frac{s^{-2}}{s+\widehat{\gamma}(s)} = \frac{s^{-2}}{s+\gamma_\alpha s^{\alpha-1}} \, . \tag{3.90}$$

Recall that LT of function $t^{\beta'-1}E_{\alpha',\beta'}\left(\pm a t^{\alpha'}\right)$ is $s^{\alpha'-\beta'}.\left(s^{\alpha'} \mp a\right)^{-1}$, with $Re\,[s] > |a|^{1-\alpha'}$. Here $E_{\alpha,\beta}(z)$ denotes the Wiman's function [12]. Then, performing inverse TL yields the *exact* expressions of VACF, TDC and MSD,

$$W(t) = 2d\frac{k_BT}{m} I(t) = 6\frac{k_BT}{m} t^2 E_{2-\alpha,3}\left(-\left(\frac{t}{\tau_\alpha}\right)^{2-\alpha}\right) \ , \qquad t > 0 \, . \tag{3.91}$$

$$D(t) = \frac{k_BT}{m} G(t) = \frac{k_BT}{m} t E_{2-\alpha,2}\left(-\left(\frac{t}{\tau_\alpha}\right)^{2-\alpha}\right) \ , \qquad t > 0 \, . \tag{3.92}$$

$$c_{vv}(t) = d\frac{k_BT}{m} g(t) = 3\frac{k_BT}{m} E_{2-\alpha}\left(-\left(\frac{t}{\tau_\alpha}\right)^{2-\alpha}\right) \ , \qquad t > 0 \, . \tag{3.93}$$

Here

$$\tau_\alpha = \gamma_\alpha^{-1/(2-\alpha)} \, . \tag{3.94}$$

is a *characteristic time*, and

$$E_\alpha(z) = \sum_{n=0}^{+\infty} \frac{z^n}{\Gamma(\alpha n+1)} \ , \qquad |z| < 1, \ \ z \in \mathbf{C} \, . \tag{3.95}$$

$$E_{\alpha,\beta}(z) = \sum_{n=0}^{+\infty} \frac{z^n}{\Gamma(\alpha n+\beta)} \,, \qquad |z|<1, \;\; z\in \mathbf{C}\,. \tag{3.96}$$

are *one-parameter*, $E_\alpha(z)$, and *two-parameter*, $E_{\alpha,\beta}(z)$, Mittag-Leffler (ML) functions, respectively [12].

We emphasize that, in the limit $\alpha\to 1$, we recover naturally expressions (3.56), (3.57) and (3.58) of MSD, TDC and VACF relatively to a Brownian diffusion. To this end, we have used the particular ML-functions

$$E_1(-z) = e^{-z} \,, \quad E_{1,2}(-z) = \frac{1-e^{-z}}{z} \,, \quad E_{1,3}(-z) = \frac{e^{-z}-1+z}{z^2}\,. \tag{3.97}$$

together with the notation $\tau_1 = \gamma_1^{-1}$.

We shall need the following known asymptotic series of these functions,

$$E_\alpha(-z) = -\sum_{n=1}^{+\infty} \frac{(-z)^{-n}}{\Gamma(1-\alpha n)} \,, \qquad |z|>1\,. \tag{3.98}$$

$$E_{\alpha,\beta}(-z) = -\sum_{n=1}^{+\infty} \frac{(-z)^{-n}}{\Gamma(\beta-\alpha n)} \,, \qquad |z|>1\,. \tag{3.99}$$

For shorter times, i.e. for $t << \tau_\alpha$, we find that

$$W(t) = 3\frac{k_BT}{m}t^2 \,, \qquad D(t) = \frac{k_BT}{m}t \,, \qquad c_{vv}(t) = \left\langle \mathbf{v}_0^2 \right\rangle = 3\frac{k_BT}{m}\,. \tag{3.100}$$

We have used definitions (3.95) and (3.96). This is the ballistic regime. We note that the above behaviors are independent of the particular form of the memory function.

For larger times, i.e. $t >> \tau_\alpha$, we find that

$$W(t) = 6D_\alpha t^\alpha \,, \qquad t >> \tau_\alpha\,. \tag{3.101}$$

$$D(t) = D_\alpha \alpha t^{\alpha-1} \,, \qquad t >> \tau_\alpha\,. \tag{3.102}$$

$$c_{vv}(t) = 3D_\alpha \alpha(\alpha-1)\,t^{\alpha-2} \,, \qquad t >> \tau_\alpha\,. \tag{3.103}$$

Here

$$D_\alpha = \frac{k_BT}{m\gamma_\alpha}\frac{1}{\Gamma(\alpha+1)}\,. \tag{3.104}$$

accounts for the *generalized diffusion coefficient*. Formulas (3.103) and (3.104) suggest the integral expression for this generalized diffusion coefficient

$$D_\alpha = \frac{1}{\Gamma(\alpha+1)}\int_0^{+\infty} dt\,\partial_t^{\alpha-1}\left[c_{vv}(t)\right]\,. \tag{3.105}$$

Here

$$\partial_t^{\alpha-1}\left[c_{vv}(t)\right] = \frac{d}{dt}\left[\int_0^t dx\,(t-x)^{\alpha-1}c_{vv}(x)\right]\,. \tag{3.106}$$

is the fractional Riemann-Liouville derivative [15] of order $1-\alpha$ of function $c_{vv}(t)$.

As conclusion, the chosen memory function defined in relation (3.85) describes well the subdiffusion process characterized by an exponent α whose values vary between 0 and 1.

3.2.9. Inertial subdiffusion on a sphere.

In this case, LTs of the three relaxation functions, $g(t)$, $G(t)$ and $I(t)$ are as follows

$$\widehat{g}(s) = \frac{1}{s+\widehat{\gamma}(s)+\frac{\lambda}{m}s^{-1}} = \frac{1}{s+\gamma_\alpha s^{\alpha-1}+\frac{\lambda}{m}s^{-1}}. \tag{3.107}$$

$$\widehat{G}(s) = \frac{s^{-1}}{s+\widehat{\gamma}(s)+\frac{\lambda}{m}s^{-1}} = \frac{s^{-1}}{s+\gamma_\alpha s^{\alpha-1}+\frac{\lambda}{m}s^{-1}}. \tag{3.108}$$

$$\widehat{I}(s) = \frac{s^{-2}}{s+\widehat{\gamma}(s)+\frac{\lambda}{m}s^{-1}} = \frac{s^{-2}}{s+\gamma_\alpha s^{\alpha-1}+\frac{\lambda}{m}s^{-1}}. \tag{3.109}$$

with parameter $\lambda = 3k_BT/R^2$.

Since inverses LT of the above functions cannot be determined exactly, we formally develop them in powers of the reduced parameter λ/m. Then, we obtain

$$\widehat{g}(s) = \sum_{n=0}^{\infty}\left(-\frac{\lambda}{m}\right)^n \frac{s^{-(2n+1)}}{\left[1+\gamma_\alpha s^{\alpha-2}\right]^{n+1}}. \tag{3.110}$$

$$\widehat{G}(s) = \sum_{n=0}^{\infty}\left(-\frac{\lambda}{m}\right)^n \frac{s^{-(2n+2)}}{\left[1+\gamma_\alpha s^{\alpha-2}\right]^{n+1}}. \tag{3.111}$$

$$\widehat{I}(s) = \sum_{n=0}^{\infty}\left(-\frac{\lambda}{m}\right)^n \frac{s^{-(2n+3)}}{\left[1+\gamma_\alpha s^{\alpha-2}\right]^{n+1}}. \tag{3.112}$$

Recall that LT of function $t^{\beta'-1}E^{\delta'}_{\alpha',\beta'}\left(-at^{\alpha'}\right)$ is $s^{-\beta'}.\left(1+as^{-\alpha'}\right)^{-\delta'}$, where $E^{\delta}_{\alpha,\beta}(z)$ is the Prabhakar's function [15]. By inversion, we find the following expressions for the relaxation functions, $I(t)$, $G(t)$ and $g(t)$, from which we extract those of MSD, TDC and TDC. Then, the results are expressed in terms of the following function series

$$W(t) = 6\frac{k_BT}{m}t^2\sum_{n=0}^{\infty}(-1)^n\left(\frac{t}{\tau_R}\right)^{2n} E^{n+1}_{2-\alpha,2n+3}\left[-\left(\frac{t}{\tau_\alpha}\right)^{2-\alpha}\right]. \tag{3.113}$$

$$D(t) = \frac{k_BT}{m}t\sum_{n=0}^{\infty}(-1)^n\left(\frac{t}{\tau_R}\right)^{2n} E^{n+1}_{2-\alpha,2n+2}\left[-\left(\frac{t}{\tau_\alpha}\right)^{2-\alpha}\right]. \tag{3.114}$$

$$c_{vv}(t) = 3\frac{k_BT}{m}\sum_{n=0}^{\infty}(-1)^n\left(\frac{t}{\tau_R}\right)^{2n} E^{n+1}_{2-\alpha,2n+1}\left[-\left(\frac{t}{\tau_\alpha}\right)^{2-\alpha}\right]. \tag{3.115}$$

Here $\tau_R = \omega_0^{-1} = \sqrt{m/\lambda} = R\sqrt{m/3k_BT}$ is a *second characteristic time*, which will be compared to the first one, τ_α, defined in formula (3.94). Since the sphere radius, R, has a fixed value, we can choose the amplitude of the memory function, γ_α, in such a way that $\tau_\alpha < \tau_R$.

Remark that in the limit $\omega_0 \to 0$, the only dominant terms of the above function series correspond to the particular value $n = 0$, and then, the above expressions reduce to those relative to subdiffusions in an infinite space (with no oscillation). For this reduction, we have used formula: $E^1_{\alpha',\beta'}(z) = E_{\alpha',\beta'}(z)$, whatever are the values of argument z and parameters α' and β'.

Now, let us examine the time behaviors of the dynamic quantities or interest, with the chosen memory function defined in relation (3.84). We have to distinguish three kinds of time-regimes which are the following:

Ballistic regime: For *beginning times*, that is for $t << \tau_\alpha < \tau_R$, the dominant parts of the dynamic quantities come from the value $n = 0$, and we find that

$$W(t) = 3\frac{k_BT}{m}t^2\ , \qquad 0 \le t << \tau_\alpha < \tau_R. \tag{3.116}$$

$$D(t) = \frac{k_B T}{m} t \;, \qquad 0 \le t << \tau_\alpha < \tau_R \,. \tag{3.117}$$

$$c_{vv}(t) = 3\frac{k_B T}{m} \;, \qquad 0 \le t << \tau_\alpha < \tau_R \,. \tag{3.118}$$

Subdiffusive regime: For *intermediate times*, that is for $\tau_\alpha << t << \tau_R$, and by using the asymptotic behavior M-L functions, we obtain

$$W(t) = 6D_\alpha t^\alpha \;, \qquad \tau_\alpha << t << \tau_R \,. \tag{3.119}$$

$$D(t) = D_\alpha \alpha t^{\alpha-1} \;, \qquad \tau_\alpha << t << \tau_R \,. \tag{3.120}$$

$$c_{vv}(t) = 3D_\alpha \alpha (\alpha - 1) t^{\alpha-2} \;, \qquad \tau_\alpha << t << \tau_R \,. \tag{3.121}$$

Here D_α accounts for the *generalized diffusion coefficient* , defined in relation(3.104). The above behaviors suggest that the target particle is subject to an *anomalous diffusion*.
Saturation regime: For *long times*, that is for $t >> \tau_R$, and by using the asymptotic behaviors of M-L functions, we obtain the following long time behaviors

$$W(t) = 6\frac{k_B T}{\lambda} = 2R^2 \;, \qquad t >> \tau_R \,. \tag{3.122}$$

$$D(t) = 0 \;, \qquad t >> \tau_R \,. \tag{3.123}$$

$$c_{vv}(t) = 0 \;, \qquad t >> \tau_R \,. \tag{3.124}$$

Therefore, similarly to the standard Langevin oscillator, MSD relatively to a generalized Langevin oscillator *saturates* to a finite value, that is $2R^2$, as $t \to +\infty$.

Series (3.113), (3.114) and (3.115) may exploited to find closer forms for MSD, TDC and VACF, for small, intermediate and long times.

Using the properties of the three-parameter ML-function, for small and long times, we find the following closer forms

$$W(t) = 6\frac{k_B T}{m} t^2 E_{2,3}\left[-\left(\frac{t}{\tau_R}\right)^2\right] \;, \qquad t < \tau_\alpha \,. \tag{3.125}$$

$$D(t) = \frac{k_B T}{m} t E_{2,2}\left[-\left(\frac{t}{\tau_R}\right)^2\right] \;, \qquad t < \tau_\alpha \,. \tag{3.126}$$

$$c_{vv}(t) = 3\frac{k_B T}{m} E_{2,1}\left[-\left(\frac{t}{\tau_R}\right)^2\right] \;, \qquad t < \tau_\alpha \,. \tag{3.127}$$

for smaller times.

$$W(t) = 6\frac{k_B T}{m} t^\alpha \tau_\alpha^{2-\alpha} . E_{\alpha,\alpha+1}\left[-\left(\frac{\tau_\alpha}{\tau_R}\right)^2 \left(\frac{t}{\tau_\alpha}\right)^\alpha\right] \;, \qquad t > \tau_\alpha \,. \tag{3.128}$$

$$D(t) = \frac{k_B T}{m} t^{\alpha-1} \tau_\alpha^{2-\alpha} . E_{\alpha,\alpha}\left[-\left(\frac{\tau_\alpha}{\tau_R}\right)^2 \left(\frac{t}{\tau_\alpha}\right)^\alpha\right] \;, \qquad t > \tau_\alpha \,. \tag{3.129}$$

$$c_{vv}(t) = 3\frac{k_B T}{m} . t^{\alpha-2} \tau_\alpha^{2-\alpha} . E_{\alpha,\alpha-1}\left[-\left(\frac{\tau_\alpha}{\tau_R}\right)^2 \left(\frac{t}{\tau_\alpha}\right)^\alpha\right] \;, \qquad t > \tau_\alpha \,. \tag{3.130}$$

for intermediate and long times.

Notice that the behaviors of the dynamic quantities, for ballistic, intermediate and long times are well recovered.

4. Non-Inertial Diffusion Study

4.1. Non-inertial generalized stochastic equation

When the particles diffuse in a very crowded medium, the acceleration term in the generalized Langevin stochastic equation (3.3) can be ignored. Within the framework of this crude approximation, this equation reduces to the so-called *Abel equation* which is the following

$$m\int_0^t dt'\gamma\left(t-t'\right)\mathbf{v}\left(t'\right)=-\lambda\mathbf{r}\left(t\right)+\mathbf{F}\left(t\right)\,. \tag{4.1}$$

At this stage, the *Lagrange multiplier*, λ, is unknown and will be computed below. Here $\mathbf{F}\left(t\right)$ is the stochastic force obeying identities (3.4) and (3.5).

In this section, we are interested in the evolution of MSD, only. In this case, we find that LT of relaxation function, $I\left(t\right)$, is

$$\widehat{I}\left(s\right)=\frac{s^{-2}}{\widehat{\gamma}\left(s\right)+\frac{\lambda}{m}\frac{1}{s}}\,. \tag{4.2}$$

Here $\widehat{\gamma}\left(s\right)$ accounts for LT of the memory function, $\gamma\left(t\right)$.

4.2. Formal expression of position

We easily find that the instantaneous position is the following

$$\mathbf{r}\left(t\right)=\left(1-\frac{\lambda}{m}I\left(t\right)\right)\mathbf{r}_0+\frac{1}{m}\int_0^t dt'G\left(t-t'\right)\mathbf{F}\left(t'\right)\,. \tag{4.3}$$

Then, averaging over the stochastic force gives

$$\left\langle\mathbf{r}\left(t\right)\right\rangle=\left(1-\frac{\lambda}{m}I\left(t\right)\right)\mathbf{r}_0\,. \tag{4.4}$$

The value of position for $t=0$ implies that $I\left(0\right)=0$.

4.3. Formal expression of position correlation function

Using LT techniques gives the following expression for the position correlation

$$\begin{aligned}\left\langle\mathbf{r}\left(t\right).\mathbf{r}\left(t'\right)\right\rangle &= R^2+\frac{\lambda^2}{m^2}\left(R^2-3\frac{k_BT}{\lambda}\right)I\left(t\right)I\left(t'\right)\\ &+\frac{\lambda}{m}\left(R^2-3\frac{k_BT}{\lambda}\right)\left[I\left(t\right)+I\left(t'\right)\right]-3\frac{k_BT}{m}I\left(\left|t-t'\right|\right)\,.\end{aligned} \tag{4.5}$$

For $t=t'$, we get

$$\left\langle\mathbf{r}^2\left(t\right)\right\rangle=R^2+\frac{\lambda^2}{m^2}\left(R^2-3\frac{k_BT}{\lambda}\right)I^2\left(t\right)+2\frac{\lambda}{m}\left(R^2-3\frac{k_BT}{\lambda}\right)I\left(t\right)\,. \tag{4.6}$$

We have used the fact that $I\left(0\right)=0$. Since $\left\langle\mathbf{r}^2\left(t\right)\right\rangle=R^2$, at any time t, we conclude that the *Lagrange multiplier*, λ, is that defined in (3.35). Then, the position correlation function becomes

$$\left\langle\mathbf{r}\left(t\right).\mathbf{r}\left(t'\right)\right\rangle=R^2-3\frac{k_BT}{m}I\left(\left|t-t'\right|\right)\,. \tag{4.7}$$

Setting $t'=0$ into equality 4.7 yields

$$\left\langle\mathbf{r}\left(t\right).\mathbf{r}\left(0\right)\right\rangle=R^2-3\frac{k_BT}{m}I\left(t\right)\,. \tag{4.8}$$

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

$$W\left(t\right)=6\frac{k_BT}{m}I\left(t\right)\,. \tag{4.9}$$

4.4. Non-inertial normal diffusion

In this case, the memory function is local in time, i.e. $\gamma(t) = \gamma_1 \delta(t)$, and then, LT of relaxation function, $I(t)$, is as follows

$$\widehat{I}(s) = \frac{s^{-2}}{\gamma_1 + \frac{\lambda}{m}\frac{1}{s}} . \tag{4.10}$$

Inverse LT gives

$$I(t) = \frac{m}{\lambda}\left(1 - e^{-\frac{\lambda}{m\gamma_1}t}\right) , \qquad t \geq 0 . \tag{4.11}$$

Then, MSD is

$$W(t) = 2R^2\left(1 - e^{-\frac{\lambda}{m\gamma_1}t}\right) , \qquad t \geq 0 . \tag{4.12}$$

Let us discuss the obtained time evolution of MSD for a normal diffusion on a sphere.

Firstly, we emphasize that the above expression of MSD is conform with the same result obtained solving the diffusion equation satisfied by the probability density [16].

Secondly, as is should be, $W(t)$ vanishes at $t = 0$, and it is a monotone increasing function of time.

Thirdly, for infinite time, i.e. $t >> m\gamma_1/\lambda$, MSD saturates to the fixed value, $2R^2$. But for small time, i.e. $t << m\gamma_1/\lambda$, $W(t) = 6D_0 t$, where $D_0 = k_B T/m\gamma_1$ is the standard diffusion coefficient. Then, for smaller times, Einstein's law is recovered.

Finally, by simple derivation of the above expression of MSD, with respect to time, we have obtained the expression of TDC we do not write.

4.5. Non-inertial subdiffusion

As before, we adopt a similar form for the memory function, that defined in relation (3.84). In this case, LT of relaxation function, $I(t)$, is such that

$$\widehat{I}(s) = \frac{s^{-2}}{\widehat{\gamma}(s) + \frac{\lambda}{m}\frac{1}{s}} . \tag{4.13}$$

Inverse LT gives the expected result

$$I(t) = \frac{1}{\gamma_\alpha} t^\alpha E_{\alpha,\alpha+1}\left[-\left(\frac{t}{\tau}\right)^\alpha\right] . \tag{4.14}$$

with the *relaxation time*

$$\tau = \left(\frac{m\gamma_\alpha}{\lambda}\right)^{1/\alpha} = \left(\frac{m\gamma_\alpha R^2}{3k_B T}\right)^{1/\alpha} . \tag{4.15}$$

Therefore, MSD is as follows

$$W(t) = 6\frac{k_B T}{m}\frac{1}{\gamma_\alpha} t^\alpha E_{\alpha,\alpha+1}\left(-\left(\frac{t}{\tau}\right)^\alpha\right) . \tag{4.16}$$

Let us comment about the above result.

Firstly, notice that the dependence of the surface curvature of MSD is completely contained in relaxation time, τ.

Secondly, for small time, i.e. $t << \tau$, the target particle executes a subdiffusion, and we have in this limit: $W(t) = 6D_\alpha t^\alpha$, where D_α accounts for the fractional diffusion coefficient, defined in relation (3.104).

Thirdly, for long time, i.e. $t >> \tau$, MSD saturates, and we have: $W(t) = 2R^2$, independently of the particular form of the memory function. Finally, by simple derivation of the above expression of MSD, with respect to time, we have obtained the expression for TDC we do not write.

5. Conclusion

We recall that the presented work is concerned with a general mathematical study of the (normal and anomalous) diffusion laws, when the target particle moves on a bounded curved space. The target particle motion is provoked by its diffusion with molecules or other particles located on this surface. As noted before, the anomalous diffusion is a consequence of the complexity of the structure of the curved space, and it is very slow in comparison with the normal diffusion. As shown above, the main property is that, the diffusion of the target particle is blocked beyond some characteristic time depending on the structure of the bounded surface and the kind of the diffusion process.

To do explicit calculations, the considered bounded surface was assumed to be spherical. Dynamics of the target particle was studied through three physical quantities, namely the mean squared displacement, the time diffusion coefficient and the velocity autocorrelation function. The study is accomplished using the so-called (Standard and Generalized) Langevin Equation Theory, for both *inertial* and *non-inertial* regimes. The essential result is that, at large time, the movement of the target particle is completely blocked, and then, MSD saturates to a finite value, independently on the nature of the diffusion process.

The present investigation was motivated by the diffusion phenomenon within two kinds of systems. The first is the Pickering emulsions [17] which are dispersions of a liquid (dispersed phase) in another unlike liquid (continuous phase), as oil-in-water or water-in-oil emulsions, on the form of small droplets (of size about $50nm$). In this case, the droplets stability is ensured by adsorption of soft or solid particles (of size about $25nm$) onto their surfaces. The anchored particles form a fractal aggregate and execute a diffusion motion which is generally anomalous. The associated diffusion coefficient, D_p, scales as: $D_p \sim c^{2/d_F}$, where c is the concentration of the anchored particles (surface coverage) and $d_F < 2$ is the fractal dimension of the aggregate. The second system is a cell-membrane, [18] formed by a spontaneous organization of the phospholipid molecules. In addition, it contains other details as proteins and cholesterol molecules. Proteins move freely on the membrane surface and generally execute an anomalous diffusion due to the complex chemical structure of this membrane. In fact, such a diffusion is responsible for the formation of pores within the bilayer-membrane.

As last word, we emphasize that the present work could be generalized to *second-degree* surfaces as torus, ellipsoids and so on. Such a question is under consideration.

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Declarations

Conflict of interest. The authors declare that they have no competing of interests regarding the publication of this paper.
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