Gulf Journal of Mathematics
Vol 22, Issue 1 (2026) 1-24

ISSN: 2309-4966
https://doi.org/10.56947/gjom.v22i1.3805

EXACT THEORETICAL STUDY OF DIFFUSION OF NANOPARTICLES AROUND CELL-MEMBRANES

ABDELGHANI DAOUDI[1]*, MABROUK BENHAMOU[2] and EL HASSAN EL KINANI[1]

Abstract. The aim of this work is a theoretical study of normal and anomalous diffusion laws of nanoparticles moving freely around an interacting cell-membrane. Using generalized Langevin and Smoluchowski equations theories which are equivalent, we determined the time variation of mean square displacement, time diffusion coefficient and variance of position. The essential conclusion is that, at large times, the diffusion of the target nanoparticle is faster, in intermediate times, the movement is slow, and in large times, the movement is blocked in the perpendicular direction to the cell-membrane. Finally, the cage effect was also discussed.

Keywords. Fluid-membranes, Nanoparticles, Effective interactions, Diffusion laws
2020 Mathematics Subject Classification. Primary 82C31, 92C37; Secondary 60H10

1. Introduction

Cell-membranes play an important role in life, since they separate cells from their environment and act as a selective barrier for the import and export of materials that cells need. The cell-membranes are phospholipids bilayers combined with a variety of proteins and cholesterol [35, 27, 1, 34, 10, 15, 5, 32, 18, 17]. The liquid character of the cell membranes allows them to be considered as *two-dimensional fluid-membranes*.

In general, the fluid-membranes are not pure, but they are in the presence of various entities, such as proteins, small and macro-ions, impurities, or more complex structures [6]. For example, the suspensions of membranes used in detergency and cosmetics are usually in contact with numerous additives on the form of macromolecules or colloids, in order to improve their efficiency and to control their viscoelastic properties [20].

Physics of nanoparticles in contact with a fluctuating fluid-membrane has been extensively studied in recent theoretical works [4, 3, 16, 29]. More precisely, the authors were concerned with detailed investigations of the static and dynamic

Date: Received: Nov 8, 2025; Revised: Dec 6, 2025; Accepted: Dec 19, 2025.
* Corresponding author

organization of these nanoparticles that interact with impenetrable or penetrable soft interfaces. It is noted that the Brownian motion can be investigated using two physically equivalent approaches, namely the *Smoluchowski equation* [25, 26] and the *Langevin equation* [37, 8, 9, 28].

The present study can be regarded as a natural extension of that reported in Ref. [29], which was concerned with the variation of the nanoparticle density in space and time. In our work, the question was addressed to the determination of the time-evolution of three interesting dynamic quantities, which are the perpendicular *mean-square-displacement* (MSD), the perpendicular *time-diffusion-coefficient* (TDC) and the perpendicular *variance of position* (VP). We emphasize that the diffusion in parallel directions to the interface is not affected by the thermal fluctuations of the later, and therefore, this will not be discussed in this paper. In doing so, use was made of (i) the standard Langevin and standard Smoluchowski equations and (ii) the generalized Langevin and generalized Smoluchowski equations, for weak membrane-nanoparticles interactions. Method (i) corresponds to a fast (or Brownian) movement of the target nanoparticle, for which MSD obeys Einstein's law, and method (ii) to a slower one, termed *subdiffusion*, for which MSD behaves upon time t as t^{α}, with the *subdiffusion exponent* $0 < \alpha < 1$.

In our formulation, we assume that the nanoparticles are point-like and of *low-density*, and in addition, the fluid-membrane is assumed to be penetrable and the membrane-nanoparticle interaction is *attractive*.

The main obtained result is that, beyond this specific time, the movement of the nanoparticles in the perpendicular direction to the interface is completely blocked, due to their interaction with the fluid-membrane and the thermal fluctuations of the latter. Therefore, MSD saturates to a final value, at large time, for both cases (i) and (ii).

The remaining of the presentation of this paper is structured as follows. Section 2 is devoted to a succinct recall of the induced interactions between moving nanoparticles, in particular, the mean-force potential used in this study. Dynamics study is the aim of Sections 3 and 4. The final section summarizes the key results and provides broader conclusions.

2. Membrane-mediated interactions

Consider a dilute suspension of N identical nanoparticles which move around an interacting fluid-membrane. For simplicity, we assume that these nanoparticles are point-like. In fact, such an assumption makes sense only if the nanoparticle size is much smaller than the *membrane surface roughness*, $\xi_{\perp}$. Typically, the considered nanoparticles have a diameter of a *few* tens of nanometers, whereas the surface roughness lies in the micrometer range.

In the *Monge representation*, a point on the membrane surface can be described by a three-dimensional position-vector $\mathbf{r} = (\rho, z) \in \mathbf{R}^3$, where $\rho = (x, y) \in \mathbf{R}^2$ accounts for the transverse-vector and $z = h\,(x, y)$ for the perpendicular distance from the plane located at $z = 0$.

The total Hamiltonian, $\mathcal{H}[h]$, of the membrane-nanoparticles system is the following [16, 15]:

$$\mathcal{H}[h] = \mathcal{H}_m[h] + \mathcal{H}_{nn}[h] + \mathcal{H}_{nm}[h] \ . \tag{2.1}$$

The first contribution $\mathcal{H}_m[h]$ represents the usual *Canham-Helfrich free energy* [9, 21]:

$$\mathcal{H}_m[h] = \frac{1}{2}\int d\rho \left\{\kappa (\Delta h)^2 + \sigma (\nabla h)^2 + \mu h^2\right\} \ . \tag{2.2}$$

Here $\kappa > 0$ is the *bending rigidity constant* (or simply *bending modulus*), $\sigma > 0$ the *surface tension coefficient*, and $\mu > 0$ the *confinement parameter*.

With the help of the above Hamiltonian, $\mathcal{H}_m[h]$, one calculates the thermal-averages of the physical quantities as the *height-height correlation function*. The latter is given by the following thermal-average:

$$G(\rho_1 - \rho_2) = \langle h(\rho_1) h(\rho_2)\rangle_0 - \langle h(\rho_1)\rangle_0 . \langle h(\rho_2)\rangle_0 \ . \tag{2.3}$$

The notation $\langle ...\rangle_0$ denotes the thermal-average which is performed with the Canham-Helfrich Hamiltonian in the absence of nanoparticles, $\mathcal{H}_m[h]$.

We recall that the general expression of propagator, $G(\rho)$, is [4, 3, 16, 29]:

$$G(\rho) = \frac{k_B T}{2\pi\sqrt{\sigma^2 - 4\mu\kappa}} \left[K_0(\rho\xi_-) - K_0(\rho\xi_+)\right] \ , \tag{2.4}$$

with the two length-scales:

$$\xi_\pm = \left(\frac{\sigma \pm \sqrt{\sigma^2 - 4\mu\kappa}}{2\kappa}\right)^{-1/2} \ . \tag{2.5}$$

There $K_\nu(z)$ is the *second-order modified Bessel function* [19].

From the above propagator expression, one deduces that of the mean-squared-displacement, $\xi_\perp$. The result is:

$$\xi_\perp^2 = G(0) = \frac{k_B T}{2\pi\sqrt{\sigma^2 - 4\kappa\mu}} \ln\left(\frac{\xi_+}{\xi_-}\right) \ . \tag{2.6}$$

For tensionless fluid-membranes, that is for $\sigma = 0$, the corresponding *mean membrane roughness* is:

$$\xi_\perp = 2^{-3/2} (k_B T)^{1/2} (\kappa\mu)^{-1/4} \ . \tag{2.7}$$

We recall that the in-plane correlation length, $\xi_\parallel$, is related to $\xi_\perp$ by [4, 3, 16, 29]: $\xi_\parallel = 2^{1/2} (\kappa/\mu)^{1/4}$.

There $\mathcal{H}_{nn}[h]$ accounts for the direct nanoparticle-nanoparticle interaction. The latter can be ignored as long as we are concerned with very dilute dispersions (no mutual interactions between nanoparticles). The last contribution in equality (2.1), $\mathcal{H}_{nm}[h]$, is the nanoparticle-interface interaction. For the sake of simplicity, it is assumed that $\mathcal{H}_{nm}[h]$ is a contact interaction, and depends only on the relative perpendicular distances between the nanoparticles and the fluid-membrane, that is [16]:

$$\frac{\mathcal{H}_{mn}[h]}{k_B T} = -w \sum_{i=1}^{N} \delta(z_i - h(\rho)) \ , \qquad w > 0 \ . \tag{2.8}$$

Here δ denotes the one-dimensional Dirac's distribution. The nanoparticles positions are denoted as: $\{\mathbf{r}_i = (\rho_i, z_i)\ \ i = 1, ..., N\}$. There, $w > 0$ denotes the dimensionless *interface-coupling constant*. From dimensionality point of view, w is a characteristic length which plays the role of an *extrapolation length* as usually encountered in *Surface Critical Phenomena* [7, 12, 13].

On the other hand, the interaction energy between the nanoparticles generated by the membrane-undulations was found to be [16]:

$$\mathcal{U}(\mathbf{r}_1, ..., \mathbf{r}_N) = -k_B T \ln \left\langle e^{-\mathcal{H}_{cm}[h]/k_B T} \right\rangle_0 = \sum_{i=1}^{N} U_1(\mathbf{r}_i) + \frac{1}{2} \sum_{\{i,j\}} U_2(\mathbf{r}_i, \mathbf{r}_j) + ... \ . \tag{2.9}$$

Using the cumulant method utilized in *Statistical Field Theory* [22, 36], it has been shown [16] that the p-body effective potentials, U_p's, express in terms of the following functions:

$$\Phi_p(\mathbf{r}_1, \ldots, \mathbf{r}_N) = \left\langle \prod_{i=1}^{p} \delta(z_i - h(\boldsymbol{\rho}_i)) \right\rangle_0 \ . \tag{2.10}$$

Functions Φ_p's have been exactly computed in Ref. [16].

In this work, we are concerned only with the suspensions of nanoparticles of low-density. This means that the mutual interactions between nanoparticles can be neglected. Therefore, the only remaining interaction is $U_1(z)$, which describes the direct potential between the nanoparticles and the interface. Its expression was found to be [16]:

$$\frac{U(z)}{k_B T} = -w \Phi_1(z) \ , \qquad \Phi_1(z) = \frac{1}{\sqrt{2\pi}\xi_\perp} e^{-z^2/2\xi_\perp^2} \ . \tag{2.11}$$

Explicitly, we have:

$$U(z) = U_0 e^{-z^2/2\xi_\perp^2} \ , \qquad U_0 = -\frac{w}{\sqrt{2\pi}} \frac{k_B T}{\xi_\perp} \ . \tag{2.12}$$

Quantity $|U_0|$ represents the *potential-depth.*

Therefore, the membrane-undulations give rise to a Gaussian external potential whose expression was largely discussed in Ref. [16]. We simply note that, since the essential of the phenomenon occurs very close to the interface, that is for distances $|z| << \xi_\perp$, the above one-body potential become *harmonic*, that is:

$$U(z) \simeq U_0 + \frac{k}{2} z^2 \ , \qquad |z| << \xi_\perp \ , \tag{2.13}$$

with the elastic constant:

$$k = -\frac{U_0}{\xi_\perp^2} = \frac{w}{\sqrt{2\pi}\xi_\perp^3} k_B T > 0 \ . \tag{2.14}$$

Therefore, the constant k combines the thermal-fluctuations of the fluid-membrane, through its roughness, $\xi_\perp$, and the membrane nanoparticles interaction strength $w > 0$. We shall set below $k = m\omega^2$, where m is the common mass of the immersed nanoparticles and ω is their *characteristic frequency.*

Then, the *elastic force* experienced by the nanoparticles due to the presence of the fluid-membrane is $\mathbf{F}_e = -kz\mathbf{e}_z$. Here $\mathbf{e}_z$ denotes the unit vector along the perpendicular direction to the interface.

3. Diffusion study from Langevin equation

3.1. **Stochastic equation.**

Consider now a single nanoparticle (*target* or *tracer*) which moves near an attractive penetrable fluid-membrane. This target nanoparticle diffuses with the molecules of water (*host medium*) and it is subject to an external harmonic force, $-kz\mathbf{e}_z$, where z denotes the distance of the target nanoparticle to the interface, $\mathbf{e}_z$ is the *unit vector* along the z-direction, and $k = m\omega^2$ is the elastic constant, defined in relation (2.14).

The diffusion process (with memory) we are interested in can be described using non-inertial GLE, due to Kubo [25] and Zwanzig [37]:

$$0 = -m\int_0^t dt'\gamma\left(t-t'\right)v\left(t'\right) - kz\left(t\right) + F\left(t\right) \ . \tag{3.1}$$

Here $v\left(t\right) \in \mathbf{R}$ denotes the perpendicular instantaneous velocity of the target nanoparticle, and $\gamma\left(t\right)$ is the perpendicular *memory function*. There, $F\left(t\right) \in \mathbf{R}$ is a *stochastic* or *random force* that satisfies the following rules [25, 37] :

$$\left\langle F\left(t\right)\right\rangle = 0 \ , \tag{3.2}$$

$$\left\langle F\left(t\right).F\left(t'\right)\right\rangle = 2k_BTm\gamma\left(\left|t-t'\right|\right) \ . \tag{3.3}$$

3.2. **Formal expression of position.**

Laplace transform (LT) of stochastic equation (3.1) gives that of the perpendicular velocity, that is:

$$\widehat{v}\left(s\right) = -\frac{k}{m}\widehat{G}\left(s\right)z_0 + \frac{1}{m}\widehat{g}\left(s\right)\widehat{F}\left(s\right) \ , \tag{3.4}$$

with

$$\widehat{g}\left(s\right) = \frac{1}{\widehat{\gamma}\left(s\right) + \frac{k}{m}s^{-1}} = \int_0^{+\infty} g\left(t\right)e^{-ts}dt \ , \tag{3.5}$$

and

$$\widehat{G}\left(s\right) = s^{-1}\widehat{g}\left(s\right) = \frac{s^{-1}}{\widehat{\gamma}\left(s\right) + \frac{k}{m}s^{-1}} = \int_0^{+\infty} G\left(t\right)e^{-ts}dt \ , \tag{3.6}$$

which are LTs of two *relaxation functions*, denoted as $g\left(t\right)$ et $G\left(t\right)$. Notice that $g\left(t\right) = dG\left(t\right)/dt$. There, $\widehat{F}\left(s\right)$ is LT of the stochastic force, $F\left(t\right)$. We have used LT of convolution product of velocity and memory function that is: $\widehat{v}\left(s\right)\widehat{\gamma}\left(s\right)$.

Inverse LT of relation (3.4) gives the perpendicular component of the velocity, as a functional of the stochastic force:

$$v\left(t\right) = -\frac{k}{m}G\left(t\right)z_0 + \frac{1}{m}\int_0^t g\left(t-t'\right)F\left(t'\right)dt' \ . \tag{3.7}$$

Averaging this equation over random force gives the mean-velocity:

$$\left\langle v\left(t\right)\right\rangle = -\frac{k}{m}G\left(t\right)z_0 \ . \tag{3.8}$$

Since $\langle v(t)\rangle < 0$, the relaxation function, $G(t)$, is positive definite: $G(t) > 0$, for all times.

From relation $\widehat{v}(s) = s\widehat{z}(s) - z_0$ and formula (3.4), we deduce LT of the instantaneous vertical coordinate of the target nanoparticle, $\widehat{z}(s)$:

$$\widehat{z}(s) = \frac{z_0}{s} - \frac{k}{m}\widehat{I}(s)\, z_0 + \frac{1}{m}\widehat{G}(s)\,\widehat{F}(s) \ , \tag{3.9}$$

with

$$\widehat{I}(s) = s^{-1}\widehat{G}(s) = s^{-2}\widehat{g}(s) = \frac{s^{-2}}{\widehat{\gamma}(s) + \frac{k}{m}s^{-1}} = \int_0^{+\infty} I(t)\, e^{-ts} dt \ , \tag{3.10}$$

which is LT of the third *relaxation function*, $I(t)$. We then have the following equality:

$$I(t) = \int_0^t G(t')\, dt' \ . \tag{3.11}$$

We note that the relaxation function, $I(t)$, is an increasing function of time.

Inverse LT of equality (3.9) allows the time evolution of the instantaneous vertical position of the target nanoparticle, that is:

$$z(t) = \left(1 - \frac{k}{m}I(t)\right) z_0 + \frac{1}{m}\int_0^t G(t-t')\, F(t')\, dt' \ . \tag{3.12}$$

Such an expression indicates that the position is a functional of random force, $F(t)$. The value of $z(t)$ for $t = 0$ imposes the condition that: $I(0) = 0$.

Then, the average position of the target nanoparticle around the fluid-membrane is:

$$\langle z(t)\rangle = \left(1 - \frac{k}{m}I(t)\right) z_0 \ . \tag{3.13}$$

Since $0 \leq \langle z(t)\rangle \leq z_0$, for all time t, the relaxation function, $I(t)$, is bounded from below by 0, and from above, by factor, m/k.

3.3. **Formal expression of position correlation function.**

From expression (3.9), we obtain:

$$\begin{aligned}\langle \widehat{z}(s)\,\widehat{z}(s')\rangle \ &= \ \frac{z_0^2}{ss'} - \frac{k}{m}\left(\frac{\widehat{I}(s)}{s'} + \frac{\widehat{I}(s')}{s} - \frac{k}{m}\widehat{I}(s)\,\widehat{I}(s')\right) z_0^2 \\ &\quad + \frac{k_B T}{m}\widehat{G}(s)\,\widehat{G}(s')\,\frac{\widehat{\gamma}(s) + \widehat{\gamma}(s')}{s+s'} \ .\end{aligned} \tag{3.14}$$

We have used the result that [30]:

$$\langle F(s)\, F(s')\rangle = mk_B T\frac{\widehat{\gamma}(s) + \widehat{\gamma}(s')}{s+s'} \ . \tag{3.15}$$

Some non-trivial algebra gives the following LT of position correlation function:

$$\begin{aligned}\left\langle \widehat{z}\left(s\right)\widehat{z}\left(s'\right)\right\rangle &= \frac{z_0^2}{ss'}-\frac{k}{m}\left(\frac{\widehat{I}\left(s\right)}{s'}+\frac{\widehat{I}\left(s'\right)}{s}\right)z_0^2+\frac{k^2}{m^2}\left(z_0^2-\frac{k_BT}{k}\right)\widehat{I}\left(s\right)\widehat{I}\left(s'\right)\\ &+\frac{k_BT}{m}\left(\frac{\widehat{I}\left(s\right)}{s'}+\frac{\widehat{I}\left(s'\right)}{s}-\frac{\widehat{I}\left(s\right)+\widehat{I}\left(s'\right)}{s+s'}\right) . \end{aligned} \tag{3.16}$$

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

$$\begin{aligned}\left\langle z\left(t\right)z\left(t'\right)\right\rangle &= z_0^2+\frac{k}{m}\left(\frac{k_BT}{k}-z_0^2\right)\left(I\left(t\right)+I\left(t'\right)\right)\\ &+\frac{k^2}{m^2}\left(z_0^2-\frac{k_BT}{k}\right)I\left(t\right)I\left(t'\right)-\frac{k_BT}{m}I\left(\left|t-t'\right|\right) . \end{aligned} \tag{3.17}$$

In general, this correlation function is *not stationary*, but it becomes *stationary*, only for the particular value of the squared position: $z_0^2=k_BT/k$.

3.4. **Formal expressions of dynamic quantities.**

Setting $t'=t$ and $t'=0$ successively in formula (3.17) yields:

$$\left\langle z^2\left(t\right)\right\rangle = 2\frac{k_BT}{m}\left(1-\frac{k}{2m}I\left(t\right)\right)I\left(t\right)+z_0^2\left(1-\frac{k}{m}I\left(t\right)\right)^2 , \tag{3.18}$$

and

$$\left\langle z\left(t\right)z\left(0\right)\right\rangle = \left(1-\frac{k}{m}I\left(t\right)\right)z_0^2 . \tag{3.19}$$

Since the above correlation function must vanish at infinite time, i.e. for $t\rightarrow+\infty$, then, we have necessarily the condition that the relaxation function, $I\left(t\right)$, reaches its maximal value, which is m/k. Such an assertion will be demonstrated analytically below.

In definitive, combining relations (3.18) and (3.19) yields the formal expression of the perpendicular MSD, $W\left(t\right)=\left\langle\left[z\left(t\right)-z\left(0\right)\right]^2\right\rangle$:

$$W\left(t\right)=2\frac{k_BT}{m}\left(1-\frac{k}{2m}I\left(t\right)\right)I\left(t\right)+\left(\frac{k}{m}I\left(t\right)\right)^2z_0^2 , \tag{3.20}$$

and by a simple derivation of MSD, $W\left(t\right)$, with respect to time, t, we obtain the formal expression of TDC:

$$D\left(t\right)=\frac{1}{2}\frac{dW\left(t\right)}{dt}=\frac{k_BT}{m}\left(1-\frac{k}{m}G\left(t\right)\right)G\left(t\right)+\frac{k^2}{m^2}G\left(t\right)I\left(t\right)z_0^2 . \tag{3.21}$$

We find that the formal expression of VP is as follows:

$$\sigma^2\left(t\right)=<z^2\left(t\right)>-<z\left(t\right)>^2=2\frac{k_BT}{m}\left(1-\frac{k}{2m}I\left(t\right)\right)I\left(t\right) . \tag{3.22}$$

Now, in order to express MSD, TDC and VP on linear forms, we introduce a *new relaxation function*, $J(t)$, we define as:

$$J(t) = I(t) - \frac{k}{2m} I^2(t) \ . \tag{3.23}$$

LT of $J(t)$ is such that (see below):

$$\widehat{J}(s) = \frac{s^{-2}}{\widehat{\gamma}(s) + \frac{2k}{m} s^{-1}} \ . \tag{3.24}$$

Such a new formula suggests that $\widehat{J}(s)$ can be deduced from $\widehat{I}(s)$ changing simply k by $2k$.

Therefore, with these considerations, the dynamic quantities of interest take the following linear forms:

$$\left\langle z^2(t) \right\rangle = \left(1 - 2\frac{k}{m} J(t)\right) z_0^2 \ , \tag{3.25}$$

$$W(t) = 2\frac{k_B T}{m} J(t) + \frac{2k}{m} \left(I(t) - J(t)\right) z_0^2 \ , \tag{3.26}$$

$$D(t) = \frac{k_B T}{m} H(t) + \frac{k}{m} \left(G(t) - H(t)\right) z_0^2 \ , \tag{3.27}$$

$$\sigma^2(t) = 2\frac{k_B T}{m} J(t) \ , \tag{3.28}$$

with an additional *relaxation function*:

$$H(t) = \frac{dJ(t)}{dt} \tag{3.29}$$

whose LT is:

$$\widehat{H}(s) = s\widehat{J}(s) = \frac{s^{-1}}{\widehat{\gamma}(s) + \frac{2k}{m} s^{-1}} \ . \tag{3.30}$$

Now, let us comment about the above general results.

First, relation (3.23) suggests that $J(0) = 0$ and $J(t) < I(t)$, at any time t. In addition, $J(t) > 0$, at any time t, since $\sigma^2(t) > 0$.

Second, expression (3.26) demonstrates that MSD is always positive definite, since $I(t) > J(t)$.

Third, as it should be, $\sigma^2(t)$, is independent of the value of the starting position, z_0.

Fourth, for vanishing membrane-nanoparticles interaction, i.e. $k = 0$ (or equivalently $w = 0$), the traditional formal expressions of MSD, TDC and VP are recovered, in terms of the usual relaxation functions, $I_0(t)$ and $G_0(t)$, in the absence of the fluid-membrane.

Fifth, both $W(t)$ and $D(t)$ naturally increases with increasing z_0. Indeed, an increase of the latter gives more mobility in space of the tracer.

Sixth, $W(t)$ reaches its maximal value at infinite time, that is for $t \to +\infty$, which is $z_0^2 + k_B T/k$, since $I(t)$ and $J(t)$ saturate to finite values m/k and $m/2k$, respectively. But, in this limit, $D(t)$ vanishes and $\sigma^2(t)$ goes to $k_B T/k$.

Seventh, for $z_0 = z_0^*$, with $z_0^* = k_B T/k$, both $W(t)$ and $D(t)$ become:

$$W(t) = 2\frac{k_B T}{m} I(t) \ , \qquad D(t) = \frac{k_B T}{m} G(t) \ , \tag{3.31}$$

independently on the particular form of relaxation function, $J(t)$.

Finally, both $W(t)$ and $D(t)$ diminish, for $z_0 < z_0^*$. In fact, this diminishment indicates that the movement of the target nanoparticle is confined in a slab around the fluctuating fluid-membrane of roughness, $\xi_\perp$. The perpendicular size of this slab is of the order of $2z_0^*$. The first condition ($z_0 > z_0^*$) means that, initially, the diffusion starts far from the interface, while the second ($z_0 < z_0^*$), means that the diffusion starts close this interface. Therefore, for $z_0 < z_0^*$, the perpendicular diffusion is much slower, and for $z_0 > z_0^*$, it is rather more faster.

3.5. **Normal diffusion.**

For this case, the memory kernel is local in time, that is $\gamma(t) = \gamma_0 \delta(t)$, where γ_0 is the *relaxation rate* whose inverse is the relaxation time. Then, in this case, LTs of the relaxation functions, $I(t)$ and $G(t)$, are the following:

$$\widehat{I}(s) = \frac{s^{-2}}{\gamma_0 + \frac{k}{m} s^{-1}} \ , \qquad \widehat{G}(s) = \frac{s^{-1}}{\gamma_0 + \frac{k}{m} s^{-1}} \ . \tag{3.32}$$

Inverse LT gives:

$$I(t) = \frac{\tau}{\gamma_0}\left(1 - e^{-t/\tau}\right) \ , \qquad G(t) = \frac{1}{\gamma_0} e^{-t/\tau} \ , \qquad t \geq 0 \ , \tag{3.33}$$

with the *characteristic time*:

$$\tau = \frac{m\gamma_0}{k} \ . \tag{3.34}$$

Therefore, according to the general formulae (3.20), (3.21) and (3.22), and the explicit expression (3.33), the expected results are:

$$\frac{W(t)}{W_0} = \left(1 - e^{-2t/\tau}\right) + \eta\left(1 - e^{-t/\tau}\right)^2 \ , \tag{3.35}$$

$$\frac{D(t)}{D_0} = e^{-2t/\tau} + \eta e^{-t/\tau}\left(1 - e^{-t/\tau}\right) \ , \tag{3.36}$$

$$\frac{\sigma^2(t)}{\sigma_0^2} = 1 - e^{-2t/\tau} \ , \tag{3.37}$$

with the notations: $W_0 = \sigma_0^2 = k_B T/k$ and $D_0 = k_B T/m\gamma_0$. Quantity D_0 denotes simply the *usual diffusion coefficient*. There, the dimensionless parameter, η, is the distance ratio: $\eta = (z_0/z_0^*)^2$.

In addition, according to general formula (3.13) and explicit expression (3.33), we find that the mean position is: $\langle z(t) \rangle = e^{-t/\tau} z_0$.

Let us discuss the time evolution of the above obtained expressions for perpendicular MSD, TDC and VP.

First, both perpendicular MSD and TDC depend naturally on the starting distance of the tracer, z_0, through the parameter, η.

Second, for the particular value, $\eta = 1$, we have:

$$\frac{W(t)}{W_0} = 2\left(1 - e^{-t/\tau}\right) \ , \tag{3.38}$$

$$\frac{D(t)}{D_0} = e^{-t/\tau} \ . \tag{3.39}$$

Third, at large times, that is for $t >> \tau$, the perpendicular MSD saturates to the constant value: $z_0^2 + z_0^{2*}$.

Fourth, at small times, that is for $t << \tau$, $W(t) \to 2D_0 t$, where $D_0 = k_B T / m\gamma_0$ is the standard diffusion coefficient. Remark that this dominant part of MSD is independent of the initial vertical position, z_0. Therefore, at small times, the perpendicular MSD obeys Einstein's law.

Sixth, we easily show that $W(t)$ is an increasing monotone function from $t = 0$ to $t = +\infty$, since $D(t) > 0$, at any time t.

Seventh, $\sigma^2(t)$ is an increasing monotone function of time and saturates to a finite value, that is $\sigma^2(t) \to k/m$, as $t \to +\infty$.

Finally, we report in Figs. (1), (2) and (3), the reduced MSD, $W(t)/W_0$, the reduced TDC, $D(t)/D_0$, and the reduced VP, $\sigma^2(t)/\sigma_0^2$, versus the renormalized time, t/τ. Curves in Figs. (1) and (2) are drawn with three fixed values of the dimensionless ratio, η. All these curves deal with the normal diffusion.

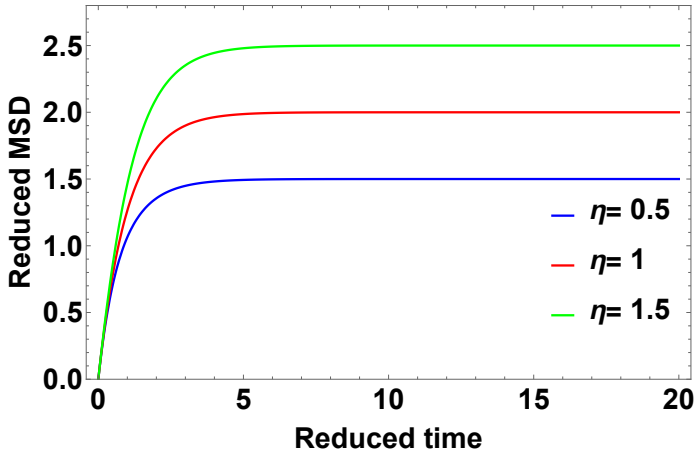

FIGURE 1. Reduced MSD, $W(t)/W_0$, (3.35) versus the dimensionless time, t/τ, for *three* values of the parameter, η: $\eta = 0.5$, $\eta = 1$ and $\eta = 1.5$.

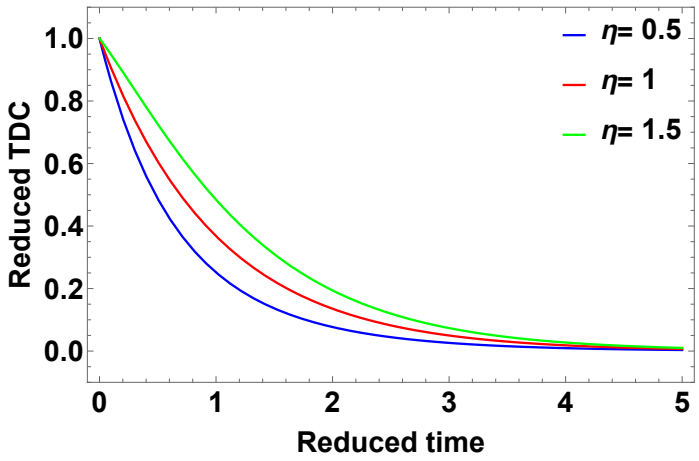

FIGURE 2. Reduced TDC, $D(t)/D_0$, (3.36) versus the dimensionless time, t/τ, for *three* values of the parameter, η: $\eta = 0.5$, $\eta = 1$ and $\eta = 1.5$.

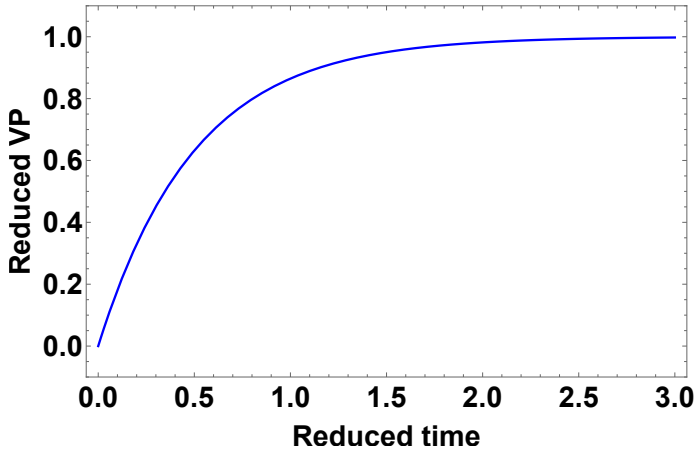

FIGURE 3. Reduced VP, $\sigma^2(t)/\sigma_0^2$, (3.37) versus the dimensionless time, t/τ .

3.6. **Diffusion with memory.**

To make explicit calculations, we assume the following form for the memory function:

$$\gamma(t) = \gamma_0 \delta(t) + \frac{\gamma_\alpha}{\Gamma(1-\alpha)} t^{-\alpha} , \qquad t \geq 0 . \tag{3.40}$$

Here, γ_α is a real positive constant. In equality above, factor $\Gamma(1-\alpha)$ was introduced for commodity, where $\Gamma(x)$ is the *Euler's gamma function*. Therefore, LT of the memory function is $\widehat{\gamma}(s) = \gamma_0 + \gamma_\alpha s^{\alpha-1}$. In this case, LTs of the relaxation functions of interest are the following:

$$\widehat{I}(s) = \frac{s^{-2}}{\gamma_0 + \gamma_\alpha s^{\alpha-1} + \frac{k}{m} s^{-1}} , \tag{3.41}$$

$$\widehat{J}(s)=\frac{s^{-2}}{\gamma_0+\gamma_\alpha s^{\alpha-1}+\frac{2k}{m}s^{-1}}\ , \tag{3.42}$$

$$\widehat{G}(s)=\frac{s^{-1}}{\gamma_0+\gamma_\alpha s^{\alpha-1}+\frac{k}{m}s^{-1}}\ , \tag{3.43}$$

$$\widehat{H}(s)=\frac{s^{-1}}{\gamma_0+\gamma_\alpha s^{\alpha-1}+\frac{2k}{m}s^{-1}}\ . \tag{3.44}$$

Now, it is possible from equalities (3.43) and (3.44) to extract the values, $G(0)$ and $H(0)$, of the relaxation functions, $G(t)$ and $H(t)$. Indeed, these particular values come from the behavior of LTs, $\widehat{G}(s)$ and $\widehat{H}(s)$, for $s\to\infty$ (*Tauber theorem*). Then, we find that:$G(0)=H(0)=1/\gamma_0$. Notice that the above inverse LTs cannot be determined exactly. To overcome this problem, we shall replace these LTs (3.41), (3.42), (3.43) and (3.44) by their respective formal series, assuming that ratio k/m is small enough. Physically, such an assumption makes sense only for very weak membrane fluctuations. Mathematically, this means that the nanoparticles have a strong memory amplitude. We emphasize that this kind of assumption was largely admitted in literature. To make expansions, with respect to parameter, k/m, we first write:

$$\frac{1}{\gamma_0+\gamma_\alpha s^{\alpha-1}+km^{-1}s^{-1}}=\frac{1}{\gamma_0+\gamma_\alpha s^{\alpha-1}}\frac{1}{1+\frac{km^{-1}s^{-1}}{\gamma_0+\gamma_\alpha s^{\alpha-1}}}\ , \tag{3.45}$$

and assume that $s^{-1}k/m$ is small enough compared to $\gamma_0+\gamma_\alpha s^{\alpha-1}$ term. As results we get:

$$\widehat{I}(s)=\frac{1}{\gamma_0}\sum_{n=0}^{\infty}\left(-\frac{k}{m\gamma_0}\right)^n\frac{s^{-n-2}}{\left(1+\frac{\gamma_\alpha}{\gamma_0}s^{\alpha-1}\right)^{n+1}}\ , \tag{3.46}$$

$$\widehat{J}(s)=\frac{1}{\gamma_0}\sum_{n=0}^{\infty}\left(-\frac{2k}{m\gamma_0}\right)^n\frac{s^{-n-2}}{\left(1+\frac{\gamma_\alpha}{\gamma_0}s^{\alpha-1}\right)^{n+1}}\ , \tag{3.47}$$

$$\widehat{G}(s)=\frac{1}{\gamma_0}\sum_{n=0}^{\infty}\left(-\frac{k}{m\gamma_0}\right)^n\frac{s^{-n-1}}{\left(1+\frac{\gamma_\alpha}{\gamma_0}s^{\alpha-1}\right)^{n+1}}\ , \tag{3.48}$$

$$\widehat{H}(s)=\frac{1}{\gamma_0}\sum_{n=0}^{\infty}\left(-\frac{2k}{m\gamma_0}\right)^n\frac{s^{-n-1}}{\left(1+\frac{\gamma_\alpha}{\gamma_0}s^{\alpha-1}\right)^{n+1}}\ . \tag{3.49}$$

Then, the corresponding inverse LTs are:

$$I(t)=\frac{t}{\gamma_0}\sum_{n=0}^{\infty}\left(-\frac{t}{\tau}\right)^n E^{n+1}_{1-\alpha,n+2}\left[-\left(\frac{t}{\tau_\alpha}\right)^{1-\alpha}\right]\ , \tag{3.50}$$

$$J(t)=\frac{t}{\gamma_0}\sum_{n=0}^{\infty}\left(-\frac{2t}{\tau}\right)^n E^{n+1}_{1-\alpha,n+2}\left[-\left(\frac{t}{\tau_\alpha}\right)^{1-\alpha}\right]\ , \tag{3.51}$$

$$G(t) = \frac{1}{\gamma_0} \sum_{n=0}^{\infty} \left(-\frac{t}{\tau}\right)^n E^{n+1}_{1-\alpha,n+1} \left[-\left(\frac{t}{\tau_\alpha}\right)^{1-\alpha}\right] , \tag{3.52}$$

$$H(t) = \frac{1}{\gamma_0} \sum_{n=0}^{\infty} \left(-\frac{2t}{\tau}\right)^n E^{n+1}_{1-\alpha,n+1} \left[-\left(\frac{t}{\tau_\alpha}\right)^{1-\alpha}\right] , \tag{3.53}$$

where, $E^{\delta}_{\alpha,\beta}(z)$ is the *three-parameter* Mittag-Leffler (M-L) function [31]. We have used LT of M-L [23]:

$$\mathcal{L}\left\{t^{\beta-1} E^{\delta}_{\alpha,\beta}\left(\pm a t^{\alpha}\right)\right\}(s) = \frac{s^{\alpha\delta-\beta}}{\left(s^{\alpha} \mp a\right)^{\delta}} , \qquad Re[s] > |a|^{1/\alpha} . \tag{3.54}$$

with the *new* scale-time:

$$\tau_\alpha = \left(\frac{\gamma_0}{\gamma_\alpha}\right)^{1/(1-\alpha)} . \tag{3.55}$$

Therefore, the problem depends on *two* scale-times, namely, τ, defined in relation (3.34), and τ_α.

We show in Appendix A that the four series (3.50) to (3.53) are uniformly convergent. This may be a justification of expansions (3.46) to (3.49), and formulae (3.50) to (3.53) may be viewed as solutions in terms of formal series. With these considerations, MSD, TCD and VP, defined respectively in relations (3.20), (3.21) and (3.22) write:

$$\frac{W(t)}{W_0} = 2 \sum_{n=0}^{\infty} (-1)^n \left[2^n + \eta\left(1 - 2^n\right)\right] \left(\frac{t}{\tau}\right)^{n+1} E^{n+1}_{1-\alpha,n+2} \left[-\left(\frac{t}{\tau_\alpha}\right)^{1-\alpha}\right] , \tag{3.56}$$

$$\frac{D(t)}{D_0} = \sum_{n=0}^{\infty} (-1)^n \left[2^n + \eta\left(1 + 2^n\right)\right] \left(\frac{t}{\tau}\right)^{n} E^{n+1}_{1-\alpha,n+1} \left[-\left(\frac{t}{\tau_\alpha}\right)^{1-\alpha}\right] , \tag{3.57}$$

$$\frac{\sigma^2(t)}{\sigma_0^2} = 2 \sum_{n=0}^{\infty} (-1)^n 2^n \left(\frac{t}{\tau}\right)^{n+1} E^{n+1}_{1-\alpha,n+2} \left[-\left(\frac{t}{\tau_\alpha}\right)^{1-\alpha}\right] , \tag{3.58}$$

with the notation: $\eta = \left(z_0/z_0^*\right)^2$. It is noted that MSD, TCD and VP depend on the characteristics of the fluid-membrane through the typical time, τ, or equivalently through the elastic constant, k. The latter scale respectively as: $\tau \sim \xi_\perp^3/w$ and $k \sim w/\xi_\perp^3$.

Notice that, for $\gamma_\alpha \to 0$ ($\tau_\alpha \to +\infty$), expressions (3.35), (3.36) and (3.37) are then recovered. For that, we have used the small argument behavior of the three-parameter M-L function.

In the following, without losing generality, we shall assume that $\tau_\alpha < \tau$. In fact, this is equivalent to choose a suitable value for parameter, γ_α, which models the non-trivial contribution of the memory function. Therefore, the analysis of the time behavior of the dynamic quantities, $W(t)$, $D(t)$ and $\sigma^2(t)$, depends on *three* kinds of time intervals, which are: (i) $t << \tau_\alpha$, (ii) $\tau_\alpha << t << \tau$ and (iii) $t >> \tau$:

Small time regime : In this regime, using behavior $E^{\delta}_{\alpha,\beta}(-z) \simeq [\Gamma(\beta)]^{-1}$ for $z \to 0$ yields:

$$W(t) = 2 D_0 t , \qquad t << \tau_\alpha , \tag{3.59}$$

$$D(t) = D_0 \ , \qquad t << \tau_\alpha \ , \tag{3.60}$$

$$\sigma^2(t) = 2D_0 t \ , \qquad t << \tau_\alpha \ . \tag{3.61}$$

Therefore, at the beginning times, the target nanoparticle does not feel the thermal fluctuations of the fluid-membrane, and then, its diffusion obeys the usual Einstein's law.

Intermediate time regime : In this regime, using behavior $E^{\delta}_{\alpha,\beta}(-z) \simeq [\Gamma(\beta - \alpha\delta)]^{-1} z^{-\delta}$ for $z \to +\infty$ gives:

$$W(t) = 2D_\alpha t^\alpha \ , \qquad \tau_\alpha << t << \tau \ , \tag{3.62}$$

$$D(t) = D_\alpha t^{\alpha-1} \ , \qquad \tau_\alpha << t << \tau \ , \tag{3.63}$$

$$\sigma^2(t) = 2D_\alpha t^\alpha \ , \qquad \tau_\alpha << t << \tau \ , \tag{3.64}$$

independently on the value of the initial position, z_0. Here,

$$D_\alpha = \frac{k_B T}{m\gamma_\alpha} \frac{1}{\Gamma(1+\alpha)} \tag{3.65}$$

denotes the *generalized diffusion coefficient*. Therefore, behavior (3.62) suggests that, for intermediate times, the movement of the target nanoparticle is *subdiffusive*, with a subdiffusion exponent $\alpha \in]0,1[$.

Saturation regime : For large times, that is for $t >> \tau$, by using the asymptotic behavior $E^{\delta}_{\alpha,\beta}(-z) \simeq [\Gamma(\beta - \alpha\delta)]^{-1} z^{-\delta}$ for $z \to +\infty$, we obtain:

$$W(t) \to z_0^2 + \frac{k_B T}{k} \ , \qquad t >> \tau \ , \tag{3.66}$$

$$D(t) \to 0 \ , \qquad t >> \tau \ , \tag{3.67}$$

$$\sigma^2(t) = \frac{k_B T}{k} \ , \qquad t >> \tau \ , \tag{3.68}$$

with the elastic constant $k = w k_B T/\sqrt{2\pi}\xi_\perp^3$. Then, knowing the initial position, z_0, behavior (3.66) may be an experimental measure of the membrane-nanoparticles coupling energy, w, at fixed temperature, T, and membrane roughness, $\xi_\perp$. Notice that the saturation regime is reached rapidly for strong coupling energy, w, since $\tau \sim \xi_\perp^3/w$.

Finally, we report in Figs. (4), (5) and (6), the reduced MSD, $W(t)/W_0$, the reduced TDC, $D(t)/D_0$ and the reduced VP, $\sigma^2(t)/\sigma_0^2$, versus the dimensionless time, t/τ, for diffusions with memory. MSD and TDC curves are drawn choosing three different values of parameter, η. All these curves reflect discussion we made above.

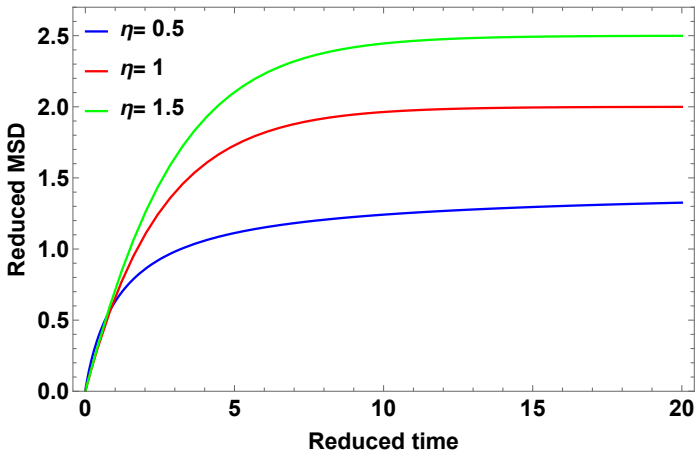

FIGURE 4. Reduced MSD, $W(t)/W_0$, (3.56) versus the dimensionless time, t/τ, for *three* values of the parameter, η: $\eta = 0.5$, $\eta = 1$ and $\eta = 1.5$.

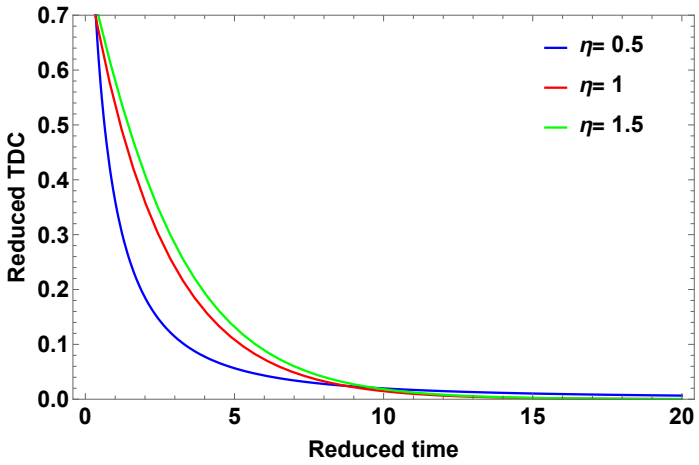

FIGURE 5. Reduced TDC, $D(t)/D_0$, (3.57) versus the dimensionless time, t/τ, for *three* values of the parameter, η: $\eta = 0.5$, $\eta = 1$ and $\eta = 1.5$.

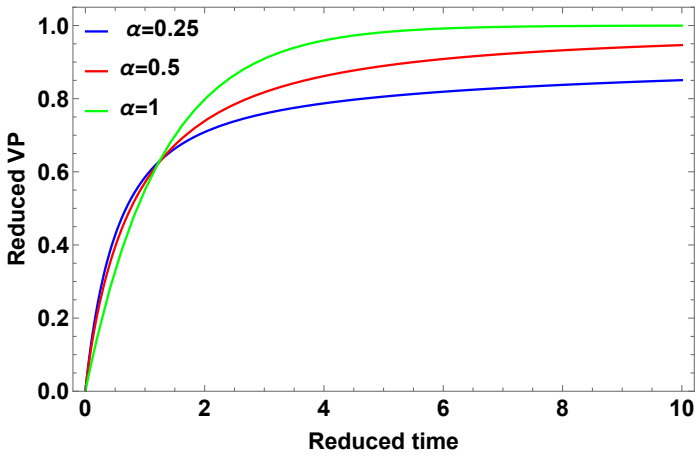

FIGURE 6. Reduced VP, $\sigma^2(t)/\sigma_0^2$, (3.58) versus the dimensionless time, t/τ, for three values of the parameter, α: $\alpha = 0.25$, $\alpha = 0.5$ and $\alpha = 1$.

4. Diffusion study from Smoluchowski equation

4.1. **Normal diffusion.**

The main quantity to consider is the *probability density*, $P(z,t)$, to find a target nanoparticle at position z, at time t, knowing that it was initially at position z_0, at initial time $t = 0$. This solves the well-known *Smoluchowski equation* (or *diffusion equation*) [14, 33], in the presence of an external potential, that is:

$$\frac{\partial P}{\partial t} = D_0 \frac{\partial^2 P}{\partial z^2} + \frac{1}{\zeta}\frac{\partial}{\partial z}\left(P\frac{\partial U}{\partial z}\right) \ , \tag{4.1}$$

with the initial condition:

$$P(z,0) = \delta(z - z_0) \ . \tag{4.2}$$

Here, $\zeta = m\gamma_0$ accounts for the *friction coefficient*, and $D_0 = k_B T/\zeta$, for the standard diffusion coefficient. For our case, the external potential is $U(z) = U_0 + kz^2/2$, and then, the above differential equation becomes:

$$m\gamma_0 \frac{\partial P}{\partial t} = k_B T \frac{\partial^2 P}{\partial z^2} + k\frac{\partial}{\partial z}(zP) \ . \tag{4.3}$$

Such an equation can be solved exactly, we simply recall its solution of Gaussian form [14]:

$$P(z,t) = \frac{1}{\sqrt{2\pi\sigma^2}} e^{-(z-\mu)^2/2\sigma^2} \ . \tag{4.4}$$

Here $\mu = \langle z \rangle = z_0 e^{-t/\tau}$, accounts for the centre of the Gaussian distribution, and $\sigma^2 = k_B T\left(1 - e^{-2t/\tau}\right)/k$, for its variance.

Notice that the initial condition (4.2) of the probability density is ensured. Indeed, in the limit $t \to 0$, we have:

$$P(z,t) \to (4\pi D_0 t)^{-1/2} \exp\left(-\frac{(z-z_0)^2}{4D_0 t}\right) \to \delta(z - z_0) \ , \tag{4.5}$$

A simple algebra gives the expressions of MSD, TDC and VP:

$$W(t) = \int_{-\infty}^{+\infty} (z - z_0)^2 P(z,t)\, dz = \frac{k_B T}{k}\left(1 - e^{-2t/\tau}\right) + \left(1 - e^{-t/\tau}\right)^2 z_0^2 \,, \quad (4.6)$$

$$D(t) = 2\frac{k_B T}{k}\frac{1}{\tau}e^{-2t/\tau} + \frac{2}{\tau}e^{-t/\tau}\left(1 - e^{-t/\tau}\right) z_0^2 \,, \quad (4.7)$$

$$\sigma^2(t) = \frac{k_B T}{k}\left(1 - e^{-2t/\tau}\right) \,. \quad (4.8)$$

We then recover the expressions of the dynamic quantities already obtained within the framework of standard Langevin equation theory, relations (3.35), (3.36) and (3.37).

We note that high-order moments of the variables z and $z - z_0$ can be computed exactly through the knowledge of their *exact* associated *generating functions*, $\Phi_i(\lambda)$, defined as:

$$\Phi_1(\lambda) = \left\langle e^{i\lambda z t}\right\rangle = \int_{-\infty}^{+\infty} e^{i\lambda z t} P(z,t)\, dz = e^{-\frac{1}{2}\sigma^2\lambda^2 + i z_0 \lambda e^{-t/\tau}} \,, \quad (4.9)$$

$$\Phi_2(\lambda) = \left\langle e^{i\lambda(z-z_0)t}\right\rangle = \int_{-\infty}^{+\infty} e^{i\lambda(z-z_0)t} P(z,t)\, dz = e^{-\frac{1}{2}\sigma^2\lambda^2 - i\lambda z_0\left(1-e^{-t/\tau}\right)} \,. \quad (4.10)$$

These moments are simply derivatives of the corresponding generating functions with respect to parameter, λ. We find that:

$$\langle z^n\rangle = n!\left(\frac{1}{i}\frac{d}{d\lambda}\right)^n \Phi_1(\lambda)\Big|_{\lambda=0} = n!\left(\frac{1}{i}\frac{d}{d\lambda}\right)^n \left(e^{-\frac{1}{2}\sigma^2\lambda^2 + i\lambda z_0 e^{-t/\tau}}\right)\Big|_{\lambda=0} \,, \quad (4.11)$$

$$\langle (z - z_0)^n\rangle = n!\left(\frac{1}{i}\frac{d}{d\lambda}\right)^n \Phi_2(\lambda)\Big|_{\lambda=0} \,. \quad (4.12)$$

4.2. **Diffusion with memory.**

The probability density, $P(z,t)$, to find a target nanoparticle at position z, at time t, solves the *generalized Smoluchowski equation* (GSE), that is:

$$m\int_0^t dt' \gamma(t - t')\frac{\partial P(z,t')}{\partial t'} = k_B T\frac{\partial^2 P}{\partial z^2} + k\frac{\partial}{\partial z}(zP) \,, \quad (4.13)$$

with the conditions (4.2) and $P(z,t) \to 0$, as $z \to \pm\infty$, at any time t.

Let us make some comments dealing with the above *two order* stochastic differential equation and its implications.

First, for $\gamma(t - t') = \gamma_0 \delta(t - t')$, we recover the standard Smoluchowski equation (4.1).

Second, the solution to the above second order differential equation depends strongly on the particular form of the chosen memory function, $\gamma(t)$.

Third, even the probability density, $P(z,t)$, is not completely known, all moments of the z-random variable:

$$\mathbf{M}_n(t) = \langle z^n\rangle = \int_{-\infty}^{+\infty} \mathbf{z}^n \mathbf{P}(z,t)\, \mathbf{dz} \,, \qquad \mathbf{n \geq 1} \quad (4.14)$$

can be calculated by multiplying the general differential equation (4.13) by $z^n (n \geq 1)$, and integrating over the variable z-variable, to find:

$$\begin{aligned} & m\left(\int_0^t dt' \gamma\left(t-t'\right) \frac{\partial}{\partial t'}\left[\int_{-\infty}^{+\infty} z^n P\left(z, t'\right) dz\right]\right) \\ = \quad & k_B T\left[\int_{-\infty}^{+\infty} z^n \frac{\partial^2 P\left(z, t\right)}{\partial z^2} dz\right]+k\left[\int_{-\infty}^{+\infty} z^n \frac{\partial\left(z P\left(z, t\right)\right)}{\partial z}\right] , \end{aligned} \tag{4.15}$$

The two integrals appearing in the rigth-hand side can be computed by integration by parts. As result, we have the following *one order* differential equations solved by moments $M_n\left(t\right)$:

$$m \int_0^t dt' \gamma\left(t-t'\right) \frac{\partial M_n\left(t'\right)}{\partial t'} = k_B T n\left(n-1\right) M_{n-2}\left(t\right) - n k M_n\left(t\right) \ , \quad n \geq 1 \ . \tag{4.16}$$

We have, by convention, $M_0\left(t\right) = \langle 1 \rangle = 1$, for all time t. Such equations indicate that, for $n \geq 3$, the moments M_{n-2} and M_n are *coupled*. These equations become linear only for $n = 1$. Notice that these equations can be solved using LT techniques.

Fourth, as examples, the first and second moments solve the following differential equations:

$$m \int_0^t dt' \gamma\left(t-t'\right) \frac{\partial M_1\left(t'\right)}{\partial t'} = -k M_1\left(t\right) \ , \tag{4.17}$$

$$m \int_0^t dt' \gamma\left(t-t'\right) \frac{\partial M_2\left(t\right)}{\partial t'} = 2 k_B T - 2 k M_2\left(t\right) \ . \tag{4.18}$$

Using LT techniques, we find that expression of $\langle z \rangle$ agrees well with that defined in relation (3.13) within the framework of Langevin formalism. Using the same techniques, the resolution of differential equation (4.18) gives $\langle z^2 \rangle$ which is found identical to that defined in relation (3.18). Result of computation of VP, $\sigma^2\left(t\right)$, is which is conform with its expression (3.28) already obtained using GLE approach. Finally, we deduce the expression of MSD, $W\left(t\right)$, and first derivating MSD with respect to time yields TDC, $D\left(t\right)$. The main conclusion is that, general results (3.56), (3.57) and (3.58) from GLE are then recovered. Therefore, the two theoretical approaches, GLE and GSE, are equivalent for a quantitative description of diffusion laws of moving nanoparticles around a fluctuating fluid-membrane.This exact equivalence is established in Appendix B.

All moments of any order satisfy an infinite number of coupled linear systems. Solving these systems is done step by step. Indeed, as indicated in equation (4.16), the obtaining of moment of order n requires the knowledge of moment of order $n-2$.

Sixth, at infinity, that is for $t \to +\infty$, all moments M_n saturate to finite values. For examples, $M_1\left(+\infty\right) = 0$, $M_2\left(+\infty\right) = k_B T / k$, which can be extracted directly from equations (4.17) and (4.18). For the other moments M_n, with $n \geq 3$, we demonstrate the following relations:

$$M_{2p+1}\left(+\infty\right) = 0 \ , \qquad\qquad p \in \mathbf{N}^* \ , \tag{4.19}$$

$$(2p-1)\,M_{2(p-1)}\,(+\infty)=\frac{k}{k_BT}M_{2p}\,(+\infty)\ , \qquad p\in\mathbf{N}^*\ . \tag{4.20}$$

Therefore, at infinite time, *odd order* moments vanish, but *even order* ones have non vanishing values. Functional relation (4.20) can be solved exactly and we find:

$$M_{2p}\,(+\infty)=(2p-1)!!\left(\frac{k}{k_BT}\right)^p=(2p-1)!!\,[M_2\,(+\infty)]^p\ . \tag{4.21}$$

Explicitly, the double factorial $(2p-1)!!$ is [12] :

$$(2p-1)!!=\frac{(2p-1)!}{2^{p-1}\,(p-1)!}\ , \tag{4.22}$$

with $p\geq 2$, simply express in terms of the second moment, $M_2\,(+\infty)=k/k_BT$.

Finally, according to the general formulae: $\sigma^2\,(t)=M_2\,(t)-M_1^2\,(t)$ and $W\,(t)=M_2\,(t)-2z_0M_1\,(t)+z_0^2$, we find that VP and MSD saturate respectively to the values: $W\,(t)\to z_0^2+(k/k_BT)$ and $\sigma^2\,(t)\to(k/k_BT)$, in agreement with limits (3.66)and (3.68) obtained from GLE theory.

5. Conclusion

We recall that the present work was concerned with a quantitative study of dynamic properties of very fine particles that move freely around a penetrable cell-membrane. We assumed that this cell-membrane and the nanoparticles mutually interact through attractive interactions that result from the contact interactions between the two systems and from the membrane-undulations.

More precisely, we were concerned with a theoretical study of the general diffusion of nanoparticles close to an attractive and penetrable cell-membrane, and the problem was to see how these fine particles diffuse in the perpendicular direction to this fluid-membrane.

The considered dynamic quantities were the mean square displacement, the time diffusion coefficient and the variance of position, and the essential problem to solve was their exact variations in time. The essential result is that, at large times, the movement is completed blocked in the perpendicular direction to the fluid-membrane.

Due to the incessant movement of nanoparticles, one assists to a *cage effect* along the perpendicular direction to this membrane in a region whose size is of the order of the membrane-roughness, $\xi_\perp$, interpreted as the typical size of holes and valleys of this membrane. The stay duration, denoted as τ_c, of a nanoparticle in its cage, formed by the neighboring nanoparticles, scales as: $\tau_c\sim\xi_\perp^{1/\alpha}$, where α is the subdiffusive exponent. This means that the target nanoparticle cannot escape from its cage during the interval time τ_c.

For a quantitative determination of the nature of the tracer diffusion with its environment, we have used two different approaches, namely the generalized Langevin and Smoluchowski equations theories which were found to be physically and mathematically equivalent.

Finally, we emphasize that, in this work, we have intentionally ignored the hydrodynamic effects felt by the fine particles which are caused by the membrane-undulations. In fact, such an assumption make good physical sense only when the thermal fluctuations of the fluid-membrane are weak enough. We recall that such effects have been studied in recent experiments [2]. We note that the modeling of a high-density nanoparticle suspension or when the thermal fluctuations of the fluid membrane are large can be based on the fractional Langevin equation [11].

Appendix A.

The goal of this to appendix is the demomstration of uniform convergence of series (3.50) to (3.53). Since the demonstrations are similar for these series, we shall restrict ourselves to series (3.50) representing the relaxation function, $I(t)$, only.

To this end, we first write this relaxation function on the following form:

$$I(t)=\sum_{n=0}^{+\infty}\sum_{k=0}^{+\infty}f_{k,n}\left(t\right)\ , \tag{A.1}$$

with:

$$f_{k,n}\left(t\right)=\left(-1\right)^{n}\left(\frac{t}{\tau}\right)^{n+1}\left(\frac{\tau}{\gamma_0}\right)\frac{\Gamma\left(n+k+1\right)}{\Gamma\left(n+1\right)\Gamma\left(\left(1-\alpha\right)k+n+2\right)}\frac{\left(-1\right)^{k}}{k!}\left(\frac{t}{\tau_\alpha}\right)^{(1-\alpha)k}\ . \tag{A.2}$$

To show that series (A.1) converges uniformly, we have to demonstrate that both series with respect to columns (keeping k fixed and summing over n) and series with respect to rows (summing over k, for fixed n) lead to a uniformly convergent series.

Let us begin by the study of the uniform convergence with respect to k, for fixed n. To this end, we first set:

$$a_k=\left(-1\right)^{n}\left(\frac{\tau}{\gamma_0}\right)\frac{\Gamma\left(n+k+1\right)}{\Gamma\left(n+1\right)\Gamma\left(\left(1-\alpha\right)k+n+2\right)}\frac{\left(-1\right)^{k}}{k!}\ . \tag{A.3}$$

Then, we have the ratio:

$$\left|\frac{a_{k+1}\left(\frac{t}{\tau_\alpha}\right)^{(1-\alpha)(k+1)}}{a_{k,n}\left(\frac{t}{\tau_\alpha}\right)^{(1-\alpha)k}}\right|=\left(\frac{t}{\tau_\alpha}\right)^{1-\alpha}.\frac{\left(n+k+1\right)}{\left(k+1\right)}.\left|\frac{\Gamma\left(k-\alpha k+n+2\right)}{\Gamma\left(k-\alpha k+n+2+1-\alpha\right)}\right|\ . \tag{A.4}$$

Using the asymptotic behavior of Euler' gamma function, $\Gamma\left(z\right)$ [24]:

$$\frac{\Gamma\left(z+a\right)}{\Gamma\left(z+b\right)}=z^{a-b}\left[1+\mathcal{O}\left(\frac{1}{z}\right)\right]\ ,\qquad |\arg\left(z+a\right)|\leq\pi\ ,\qquad |z|\longmapsto\infty \tag{A.5}$$

yields:

$$\left|\frac{a_{k+1}\left(\frac{t}{\tau_\alpha}\right)^{(1-\alpha)(k+1)}}{a_{k,n}\left(\frac{t}{\tau_\alpha}\right)^{(1-\alpha)k}}\right|\simeq\left(\frac{t}{\tau_\alpha}\right)^{1-\alpha}.\frac{\left(n+k+1\right)}{\left(k+1\right)}.\left(k-\alpha k\right)^{\alpha-1}\ . \tag{A.6}$$

Given $\alpha < 1$, we then obtain:

$$\lim_{k\to+\infty}\left|\frac{a_{k+1}\left(\frac{t}{\tau_\alpha}\right)^{(1-\alpha)(k+1)}}{a_{k,n}\left(\frac{t}{\tau_\alpha}\right)^{(1-\alpha)k}}\right| = 0\ . \tag{A.7}$$

This complete the demomstration of the uniform convergene of series (A.1) with respect to k, for fixed n.

Now, to demomstrate the uniform convergence of series (A.1) with respect to n, for fixed k, we first set:

$$a_n = (-1)^n\left(\frac{\tau}{\gamma_0}\right).\frac{\Gamma(n+k+1)}{\Gamma(n+1)\,\Gamma((1-\alpha)k+n+2)}\frac{(-1)^k}{k!}\ . \tag{A.8}$$

Therefore, we get:

$$\left|\frac{a_{n+1}\left(\frac{t}{\tau}\right)^{n+2}}{a_n\left(\frac{t}{\tau}\right)^{n+1}}\right| = \left(\frac{t}{\tau}\right).\frac{(n+k+1)}{(n+1)}.\frac{1}{(1-\alpha)k+n+2}\ . \tag{A.9}$$

Then, we have:

$$\lim_{n\to+\infty}\left|\frac{a_{n+1}\left(\frac{t}{\tau}\right)^{n+2}}{a_n\left(\frac{t}{\tau}\right)^{n+1}}\right| = 0\ . \tag{A.10}$$

Consequently, series (A.1) converges uniformly with respect to n for fixed k.

This completes the proof of the uniform convergence of all the considered series (3.50) to (3.53).

Appendix B.

The aim of this appendix is to show the quantitative equivalence between Langevin equation and Smoluchowski equation. For demonstration, we restrict ourselves to particle systems with no memory, only. This means that the corresponding memory is local in time, i.e., $\gamma(t-t') = \gamma_0\delta(t-t')$. In this case, the perpendicular position writes:

$$z(t) = \left(1-\frac{k}{m}I(t)\right)z_0 + \frac{1}{m}\int_0^t G(t-t')\,F(t')\,dt'\ , \tag{B.1}$$

which is a function of time, t, and a functional of random force, $F(t)$. Here, $G(t) = \gamma_0^{-1}e^{-t/\tau}$ and $I(t) = \tau\gamma_0^{-1}\left(1-e^{-t/\tau}\right)$, are relaxation functions, with the characteristic time, $\tau = m\gamma_0/k$.

In this case, the random force distribution is *Gaussian*, that is:

$$\mathcal{P}[F] = \mathcal{P}_0\exp\left\{-\frac{1}{4m\gamma_0 k_B T}\int_0^t dt_1\int_0^t dt_2 F(t_1)\,\delta(t_1-t_2)\,F(t_2)\right\}\ . \tag{B.2}$$

Here, $\mathcal{P}_0$ is a normalization constant. Such a distribution is conform to conditions (3.2) and (3.3) with local memory function.

The probability density, $P(z,t)$, to find a target nanoparticle at position z, at time t, knowing that it was initially at position z_0, at initial time $t=0$, is defined by the following mean-value over the stochastic force:

$$P(z,t)=\langle\delta[z-z(t)]\rangle_F=\int\mathcal{D}F\delta[z-z(t)]\mathcal{P}[F]\ , \tag{B.3}$$

we rewrite as:

$$P(z,t)=\int_{-\infty}^{+\infty}\frac{dk}{2\pi}\int\mathcal{D}Fe^{ik(z-z(t))}\mathcal{P}[F]\,. \tag{B.4}$$

Here, integral is performed over all random force configurations. Notice that this integral is a *path integral* in Quantum Mechanics language.

Now, by using expression (3.12) of position $z(t)$, we find that:

$$\begin{aligned}P(z,t) &= \int_{-\infty}^{+\infty}\frac{dk}{2\pi}e^{ik\left(z-\left(1-\frac{k}{m}I(t)\right)z_0\right)}\int\mathcal{D}Fe^{ikm^{-1}\int_0^t G(t-t')F(t')dt'}\times\\ &\exp\left\{-\frac{1}{4m\gamma_0k_BT}\int_0^t dt_1\int_0^t dt_2F(t_1)\delta(t_1-t_2)F(t_2)\right\}\ .\end{aligned} \tag{B.5}$$

To complete calculations, we use the following formula:

$$\begin{aligned}&\int\mathcal{D}Fe^{-\frac{1}{2}\int_0^t dt_1\int_0^t dt_2F(t_1)K(t_1-t_2)F(t_2)+i\int_0^t dt'L(t')F(t')}\\ &= \exp\left(-\frac{1}{2}\int_0^t dt_1\int_0^t dt_2L(t_1)K^{-1}(t_1-t_2)L(t_2)\right)\end{aligned} \tag{B.6}$$

Here, $K^{-1}(t_1-t_2)$ is the inverse of Kernel, $K(t_1-t_2)$, in distributions sense. In our case, $L(t')=km^{-1}G(t-t')$ and $K(t_1-t_2)=4(m\gamma_0k_BT)^{-1}\delta(t_1-t_2)$. Therefore, we obtain:

$$\begin{aligned}P(z,t) &= \int_{-\infty}^{+\infty}\frac{dk}{2\pi}e^{ik\left(z-\left(1-\frac{k}{m}I(t)\right)z_0\right)}\times\\ &\exp\left(-2k^2m\gamma_0k_BT\int_0^t dt_1G^2(t-t_1)\right)\ .\end{aligned} \tag{B.7}$$

The above integral is simply Gaussian, consequently, the final result is:

$$P(z,t)=\frac{1}{\sqrt{2\pi\sigma^2}}e^{-(z-\mu)^2/2\sigma^2}\ , \tag{B.8}$$

with $\mu=\langle z\rangle=z_0e^{-t/\tau}$ accounts for the centre of the Gaussian distribution, and $\sigma^2=k_BT\left(1-e^{-2t/\tau}\right)/k$, for variance.

This end the demonstration of the expected result.

Finally, we state that the equivalence is also true when the random force is distributed according to a Gaussian random force distribution, even for phenomena with memory.

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

Author contributions. Authors' contributions Abdelghani Daoudi, Mabrouk Benhamou and El Hassan El Kinani are the authors of this manuscript and contributed to it.

Acknowledgements. The authors thank the referees for their valuable comments that contributed to an improvement of scientific contents of our paper.

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

References

1. Alberts, B., Bray, D., Lewis, J., Raff, M., Roberts, K., & Watson, J. D. (1994). Molecular Biology of the Cell. Garland New York.
2. Benavides Parra, J. C. (2017). Brownian Motion of Colloidal Particles Located Near Different Types of Interfaces. PhD Thesis, Bretagne-Loire University, France.
3. Bendouch, A., Benhamou, M., & Kaidi, H. (2008). Brownian Dynamics of Nanoparticles Moving Near a Fluctuating Membrane. Electronic Journal of Theoretical Physics, 5(17), 211-226.
4. Bendouch, A., Kaidi, H., Bickel, T., & Benhamou, M. (2007). Aggregation of colloids near a fluid membrane. Journal of Statistical Mechanics: Theory and Experiment, 2007(01), P01016. https://doi.org/10.1088/1742-5468/2007/01/P01016
5. Benhamou, M. (2008). Organization of lipid molecules within biomembranes. Lipid Insight, 1, 3-11. https://doi.org/10.4137/LPI.S887
6. Bickel, T., Benhamou, M. & Kaidi, H. (2004). Statistical mechanics of a colloidal suspension in contact with a fluctuating membrane. Phys. Rev. E, 70, 051404-051411. https://doi.org/10.1103/PhysRevE.70.051404
7. Binder, K. (1983). Phase Transitions and Critical Phenomena, Vol. 8. edited by C. Domb and J. L. Lebowitz, Academic Press, London.
8. Bouchaud, J.-P., Georges, A. (1990). Anomalous diffusion in disordered media: Statistical Mechanisms, models and physical applications. Phy. Rep., 195, 127-293. https://doi.org/10.1016/0370-1573(90)90099-N
9. Canham, P. B. (1970). The minimum energy of bending as a possible explanation of the biconcave shape of the human red blood cell. J. Theoret. Biol., 26, 61-81. https://doi.org/10.1016/S0022-5193(70)80032-7
10. Dai, J., & Sheetz, M. P. (1997). Cell membrane mechanics. Methods in cell biology, 55, 157-171. https://doi.org/10.1016/S0091-679X(08)60407-0
11. Daoudi, A., & Benhamou, M. (2025). The study of fracional anomalous diffusion laws on the hypersphere. Gulf Journal of Mathematics, 19(2), 121-141. https://doi.org/10.56947/gjom.v19i2.2772
12. Diehl, H. W. (1986). Phase Transitions and Critical Phenomena, edited by C. Domb and J. L. Lebowitz, Vol. 10. Academic Press London.
13. Dietrich, S. (1988). Phase Transitions and Critical Phenomena, edited by Domb, C. & Lebowitz, J. L. Vol. 12. Academic Press, London.
14. Doi, M. (1986). M. and sf edwards, theory of polymer dynamics clarendon press. Oxford1988.
15. Edidin, M. (1997). Lipid microdomains in cell surface membranes. Curr. Opin. Struc. Biol, 7, 528-532. https://doi.org/10.1016/S0959-440X(97)80117-0
16. Einstein, A. (1905). On the motion of small particles suspended in liquids at rest required by the molecular-kinetic theory of heat. Annalen der physik, 17(549-560), 208. https://doi.org/10.1002/andp.19053220806
17. Evans, D. F. & Wennerstrom, H. (1999). The Colloidal Domain. Wiley, New-York.

18. Fradin, C., Abu-Arish, A., Granek, R. & Elbaum, M. (2003). Fluorescence correlation spectroscopy close to a fluctuating membrane, Biophysical Journal, 84, 2005-2020. https://doi.org/10.1016/S0006-3495(03)75009-7
19. Gradshteyn, I. S., & Ryzhik, I. M. (2014). Table of integrals, series, and products. Academic press.
20. H. Kaidi, H., Bickel, T. & Benhamou, M. (2005). Surface-mediated attraction between colloids. Europhys. Lett., 69, 15-21. https://doi.org/10.48550/arXiv.cond-mat/0410776
21. Helfrich, W. (1973). Elastic properties of lipid bilayers:theory and possible experiments. Zeitschrift für Naturforschung c, 28(11-12), 693-703. https://doi.org/10.1515/znc-1973-11-1209
22. Itzykson, C. & Drouffe, J. M. (1989). Statistical Field Theory:1 and 2. Cambridge University Press.
23. Kilbas, A. A., Saigo, M. & Saxena, R. K. (2004). Generalized Mittag-Leffler function and generalized fractional calculus operators. Integral Transforms and Special Functions, 15, 31-49. https://doi.org/10.1080/10652460310001600717
24. Kilbas, A. A. (2006). Theory and applications of fractional differential equations. North-Holland Mathematics Studies, 204.
25. Kubo, R., Toda, M. & Hashitsume, N. (2012). Statistical Physics II:Nonequilibrium Statistical Mechanics. Springer Science & Business Media.
26. Langevin, P. (1980). Sur la théorie du mouvement brownien. C.R. Acad. Sci. Paris, 146, 530-533.
27. Lipowsky, R., & Sackmann, E. (Eds.). (1995). Structure and dynamics of membranes: I. from cells to vesicles/II. generic and specific interactions. Elsevier.
28. Lmou, H., Elkhalloufy, K., Hilal, K., & Kajouni, A. (2024). Topological degree method for a new class of ϕ-Hilfer fractional differential Langevin equation. Gulf Journal of Mathematics, 17(2), 5-19. https://doi.org/10.56947/gjom.v17i2.2186
29. Wax, N. (1954). Noise and stochastic processes. New York: Dover Publication.
30. Pottier, N. (2003). Aging properties of an anomalously diffusing particule. Physica A: Statistical Mechanics and its Applications, 317(3-4), 371-382. https://doi.org/10.1016/S0378-4371(02)01361-49
31. Prabhakar, T. R. (1971). A singular integral equation with a generalized Mittag Leffler function in the kernel. Yokohama Mathematical Journal, 19(1), 7-15.
32. Safran, S. (2018). Statistical thermodynamics of surfaces, interfaces, and membranes. CRC Press.
33. Smoluchowski, M. V. (1916). Drei vortrage uber diffusion, brownsche bewegung und koagulation von kolloidteilchen. Zeitschrift fur Physik, 17, 557-585.
34. Vance, D. E., & Vance, J. E. (Eds.). (1996). Biochemistry of Lipids, Lipoproteins and Membranes (Vol. 31). Elsevier.
35. Wolfe, S. L. (1993). Molecular and cellular biology Wadsworth Publishing Company. Belmont, CaKfomia.
36. Zinn-Justin, J. (2021). Quantum field theory and critical phenomena (Vol. 171). Oxford university press.
37. Zwanzig, R. (2001). Nonequilibrium Statistical Mechanics. Oxford University Press.

[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
Email address: e.elkinani@umi.ac.ma

[2] Physics Department, Faculty of Sciences, P.O. Box 11201, Meknes, Morocco
Email address: m.benhamou@umi.ac.ma