A Novel Physical Mechanism to Model Brownian Yet Non-Gaussian Diffusion: Theory and Application

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Abstract: In the last years, a few experiments in the fields of biological and soft matter physics in colloidal suspensions have reported “normal diffusion” with a Laplacian probability distribution in the particle’s displacements (i.e., Brownian yet non-Gaussian diffusion). To model this behavior, different stochastic and microscopic models have been proposed, with the former introducing new random elements that incorporate our lack of information about the media and the latter describing a limited number of interesting physical scenarios. This incentivizes the search of a more thorough understanding of how the media interacts with itself and with the particle being diffused in Brownian yet non-Gaussian diffusion. For this reason, a comprehensive mathematical model to explain Brownian yet non-Gaussian diffusion that includes weak molecular interactions is proposed in this paper. Based on the theory of interfaces by De Gennes and Langevin dynamics, it is shown that long-range interactions in a weakly interacting fluid at shorter time scales leads to a Laplacian probability distribution in the radial particle’s displacements. Further, it is shown that a phase separation can explain a high diffusivity and causes this Laplacian distribution to evolve towards a Gaussian via a transition probability in the interval of time as it was observed in experiments. To verify these model predictions, the experimental data of the Brownian motion of colloidal beads on phospholipid bilayer by Wang et al. are used and compared with the results of the theory. This comparison suggests that the proposed model is able to explain qualitatively and quantitatively the Brownian yet non-Gaussian diffusion.

Keywords: Lennard-Jones potential; phase separation; Brownian motion; non-Gaussian; molecular interactions

1. Introduction

The dynamics of Brownian diffusion are frequently used for modeling stochastic motions to obtain information about the particle’s interaction with binding partners and the local environment [1,2]. The main characteristic of Brownian diffusion lies in the feature of random wiggling particle motion that generally produces a normal or Gaussian distribution in the particle density function, with mean \( \mu = 0 \) and variance \( \sigma^2 = 2D_t \), where \( D \) is interpreted as mass diffusivity or diffusion coefficient [3,4]. The Brownian diffusion model is very useful to analyze and study a variety of physical processes related to mechanisms of particle transport [5,6], thermal fluctuations [7–9], particle manipulation [10–12], and stellar dynamics [13,14]. Despite this Gaussian behavior being common in the displacement distribution for systems that exhibit Brownian motion, in the last years, efforts have been made to study a new type of Brownian diffusion. Like normal diffusion, it has a linear time dependence of the mean-square displacement (MSD), but is accompanied by a non-Gaussian displacement distribution, which has been identified as “anomalous yet Brownian” diffusion [15]. The Brownian yet non-Gaussian diffusion reported by Wang et al.
is based on the classical random walk, in which mean-square displacement is simply proportional to time, but instead, the displacement distribution has an exponential behavior with the decay length of the exponential being proportional to the square root of time. This concept was vastly used to propose a model based on anomalous, but Brownian diffusion to describe the nature of diffusivity memory, but not the memory in the direction of the particle’s trajectories. The model was coined as diffusing diffusivity, due to the random walk that the diffusivity experiences [16]. Chubinsky–Slater’s idea in [16] has also been useful to model the behavior of biological, soft, and active matter systems establishing a minimal model framework of diffusion processes with fluctuating diffusivity [17]. Further, in the realm of fluids, it has been shown that in some confinement conditions, density fluctuations might be relevant to Brownian yet non-Gaussian diffusion [18,19].

The literature presents different kind of stochastic models that explain mathematically the Brownian yet non-Gaussian diffusion process. This includes studies of the role of media heterogeneity by randomizing parameters that appear in diffusivity dynamical equations [20]; demonstrations on time-dependent diffusivity, induced by external non-thermal noise [21]; and interesting comparisons between non-Gaussian random diffusivity models [22].

Microscopic models have also been proposed, where they have rigorously, mathematically studied interesting physical scenarios such as the study of diffusion of ellipsoidal particles, active particles, diffusion of colloidal particles in fluctuating corrugated channels, and Brownian motion in arrays of planar convective rolls [23]; non-Gaussian diffusion in static disordered media via a quenched trap model, where the diffusivity is spatially correlated [24]; and the Hitchhiker model [25].

All the exposed models have in common that they reflect different levels of our lack of information/ignorance about the surroundings/media. The lack of information about the media is considered by introducing random elements to the models via random parameters, random diffusivity, noise, or by introducing additional physical assumptions (i.e., either about the environment or the Brownian particle (i.e., large particle being diffused in the fluid) that lead to study interesting, but more complex physical scenarios.

From a practical point of view, there is plenty of evidence that these models give correct predictions. Nevertheless, from a theoretical perspective, to the best of our knowledge, a simple physical mechanism that incorporates media information by establishing a link between weak molecular interactions, phase separation, and Brownian motion has not been studied. This fact motivates the study and verification of model calculations that consider long-range Brownian particle–fluid molecular interactions, which via a phase separation (in a compressible fluid) attempts to explain the Brownian yet non-Gaussian diffusion by providing accurate predictions (including the experimental observation of the transition to a Gaussian process as observed in [15]).

The rest of the paper is structured as follows: Section 2 presents the theoretical framework that explains how two phase-separated clusters (i.e., a group of phase-separated fluid molecules and Brownian particle is denoted by “cluster”) interact. Section 3 discusses relevant experimental information and how it compares to the proposed model. Section 4 provides experimental data and quantitatively evaluates the predictions of the model. Finally, Section 5 incorporates the conclusion.

2. Mathematical Model of Two Interacting Phase-Separated Clusters

To explain the results of recent experiments [15,26–28], a model that considers molecular interactions during Brownian motion is proposed. Motivated by [10], it is hypothesized here that due to interactions between molecules of the Brownian particle and active components of the biological fluid, the Brownian particle becomes weakly polarized. Then, it can be said that if the magnitude of the attractive energy produced by molecules of the Brownian particle is high enough to break weak interactions between components of the biological fluid, a phase separation occurs [29]. Note that if these interactions are too weak, no phase separation happens and the fluid is dispersed (see Figure 1). In this case, regular Brownian motion as described by Einstein takes place.
The case of interest in this manuscript is the one with a phase separation; therefore, a model is proposed for this case. This is characterized by a highly compressible liquid phase (i.e., weakly interacting liquid, behaving as a gaseous-like state) that is, on average, separated (however, mixed) of a nearly incompressible liquid phase. (i.e., gel-like characteristics) [30]. Further, since in the gaseous-like state particles can move more freely than in the gel-like state, equilibrium is reached faster [29]. To incorporate this fact in the model, a shorter time scale $\tau_e$, in which the system in the gaseous-like state relaxes to a local equilibrium (where no fluctuations occurs and local average of observables satisfy the ergodic hypothesis), where eventually at equilibrium, the $\tau_e$ scale is reached (there are two scales in time for this phase). For the gel-like state, it is assumed that in the $\tau_e$ scale, the system has not yet reached equilibrium; therefore, there is only one scale in this state.

It is mentioned here that for clarity purposes, we reproduce the derivation from De Gennes [31]. Before starting this derivation, we note that more information on the variables used in the derivation and in the rest of the paper can be found in Appendix A. Now, to model the weak, long-range, and pairwise interactions between molecules of the Brownian particle and fluid molecules (i.e., dipole–dipole/Van der Waals interactions) the attractive part (long-range) of a 6-12 Lennard-Jones potential $U(r_{ij})$ [32] is introduced, which is

$$U(r_{ij}) = -4\varepsilon \left(\frac{\sigma}{r_{ij}}\right)^6$$  

where $\varepsilon$ is the dispersion energy, $\sigma$ is the minimum distance a fluid molecule can be located with respect to a molecule at the interface (i.e., Van der Waals radius), and $r_{ij}$ is the distance between a molecule located in the Brownian particle and a fluid molecule.

Moreover, the Brownian particle is considered as a spherical interface interacting with the fluid, attracting fluid molecules. This model was already studied for a spherical shell interface in [31] and it was found that for the planar limit at $|R - r_m| \ll R$, with $R$ being the radius of the spherical shell and $r_m$ the distance from the center of the sphere, a $10^{-4}$ Lennard-Jones potential emerges; therefore, for a sphere, a 3-9 Lennard-Jones potential [31] would follow, where we are paying attention only to the cubic decaying part, as stated above. The limit we are interested in is the opposite limit, which is the long-range limit of $|R - r_m| \gg R$. In this same paper [31], the potential is shown to be a rapidly decaying function of $r_m^3$ ($V(r_m)$ change appreciably in scales of the order of $R \times 10^{-1}$), such that $|R - r_m| \ll R \sim |R - r_m| \gg R$. Then, expansion for each of those limits (i.e., planar and long-range limit) will have the same functional form, the same inverse cubic potential holds $V(z)$, which is from (1):

![Figure 1](image-url)
\[ V(z) = -\left(\frac{4\pi}{3} \rho_s \right) \frac{\rho_s}{z^3}, \]  

(2)

where \( z = |R - r_m| \) and \( \rho_s \) are the density at the interface. In equilibrium (i.e., where average of observables can be locally defined) at the \( \tau_c \) scale, Boltzmann–Gibbs distribution holds and, assuming that \( V(z) \ll k_B T \) (i.e., valid for relevant distances scales in Brownian experiments [15]), then the density profile \( \rho(z) \) of the fluid in the gaslike state is given by

\[ \frac{\rho(z)}{\rho_s - \rho_L} = \exp \left( -\frac{V(z)}{k_B T} \right) \approx 1 - \frac{V(z)}{k_B T}, \]  

(3)

\[ \frac{\rho(z) - \rho_L}{\rho_s - \rho_L} = \left( \frac{\rho_s}{z} \right)^3, \]  

(4)

Here, \( T \) is the temperature of the system, \( k_B \) the Boltzmann constant, and \( \rho_L \) is the density in the incompressible liquid phase. Further, as stated above, the appreciable change is in the order of \( R \times 10^{-1} \) and \( R \) dictates the validity of the approximation (3). As an example, if \( R \) is in the order of 50 nm, then the approximation performed to obtain Equation (4) and, therefore, Equation (4), is expected to hold for distances on the order of 0.1 \( \mu \text{m} \) up to infinity and the density is expected to change appreciably in the order of \( 10^{-7} \) m, (i.e., which means it can be assumed constant for smaller scales/subdomains such as \( 10^{-8} \) m or \( R \times 10^{-2} \) m, this is the scale of local equilibrium), which are the relevant experimental scales [15].

We are interested in seeing how Equation (4) generated by cluster 1 interacts with cluster 2 (in the end, we will see how this changes the displacement probability distribution). For these purposes, the relevant distance that we are interested in the behavior of the fluid is \( z' = z + R = \langle r' \rangle - R + R \). This is where the fluid interacts with the cross-section of a different cluster; then, \( z' = \langle r' \rangle \). This means that \( z \) can be replaced by \( \langle r' \rangle \) (i.e., mean relative distance measured from the center of a cluster). The \( \rho_L \) and \( \rho_s \) terms are there to satisfy the boundary conditions. When \( r \to \infty \), then \( \rho_L \) (i.e., density of the fluid in the gel-like phase) is recovered as one should expect. When \( \langle r' \rangle = R \), the density of the fluid at the interface (in the gaslike state) is recovered and is called \( \rho_s \). Note that in this case, \( \rho_s < \rho_L \); therefore, density increases up to infinity following Equation (4).

The relevant density differences in (4) are \( \bar{\rho} = \rho(\langle r' \rangle) - \rho_L \) and \( \rho_b = \rho_s - \rho_L \). Equation (4) is

\[ \bar{\rho} = \rho_b \left( \frac{\sigma}{\langle r' \rangle} \right)^3, \]  

(5)

Since there is radial symmetry, Equation (5) can be further simplified, given that

\[ \bar{\rho} = \frac{dN}{dV} = \frac{dN}{d\langle r' \rangle} \frac{1}{(4\pi \sigma^2 \langle r' \rangle^2)}; \]  

(6)

\[ \rho_b = \frac{1}{(4\pi \sigma^2 \langle r' \rangle^2)} \frac{dN}{d\langle r' \rangle} \bigg|_{\langle r' \rangle = R}; \]  

(7)

notice that \( \frac{dN}{d\langle r' \rangle} \bigg|_{\langle r' \rangle = R} = -\frac{1}{\sigma}. \) Therefore,

\[ \frac{dN}{d\langle r' \rangle} = -\left( \frac{1}{\langle r' \rangle} \right). \]  

(8)

Equation (8) was obtained by treating the fluid as an ideal gas. However, it is required in this model to consider molecular interactions in the gas (Van der Waals gas). This type of gas can be treated as an ideal gas with a modified number of particles \( ZN \), where \( Z \) is the compressibility factor [33] (note that \( Z \) is a ratio of volumes and it is a nondimensional quantity). \( Z < 1 \) means attractive interactions, \( Z = 1 \) means no interaction (ideal gas),
and \( Z > 1 \) means repulsive interactions between fluid molecules. Then, Equation (8) simply becomes

\[
\frac{d \langle ZN \rangle}{d \langle r' \rangle} = -\left( \frac{Z}{\langle r' \rangle} \right),
\]

(9)

It is this radial gradient of the number of particles that generates a radial attractive interaction between two clusters. As a comment, since the derivation of (9) was performed in the realm of classical statistical mechanics and local equilibrium was assumed, the quantity \( \langle r' \rangle \) was defined with angle brackets, since it represents an average and is a local observable in the \( \tau_e \) scale. Note that the number of particles gradient in (9) exists only in the radial direction. Further, the difference \( d \langle ZN \rangle \) is taken with respect to the number of particles the fluid would have in absence of the Lennard-Jones potential.

Moreover, the number of particles gradient in (9) do not generate any force acting on the same cluster that is producing it (self-interaction). Of course, due to its spherical symmetry, it can be deduced that this force is zero. However, it does generate a force acting on a different cluster (pairwise interaction). This force is derived as follows:

For the following, the thermodynamics for a classical ideal gas is used. This is justified since local equilibrium is assumed at the scales we are working with. At any given time, consider that cluster 1 is located a mean radial relative distance \( \langle r' \rangle \) from cluster 2. (In the following, it will be assumed that cluster 1 is in the reference frame of cluster 2). As it is done in the derivation of mean free path with ideal gases \([34,35]\), the cluster will be treated as a point particle with cross section of \( \pi R^2 \), where \( R \) is the radius of the sphere. Now, imagine a cylinder enclosing the cluster 1 cross-section oriented in the radial direction with the same cross-section as the cluster and height \( d \langle r' \rangle \) (see Figure 2). This cylinder will have a greater number of particles \( ZN_1 \) in its farthest half of cluster 2 (the fluid is not perturbed by cluster 2 for this half; then, the density remains unchanged) than in its closest half \( ZN_2 \) (see Figure 2b). This means that the force acting on cluster 1 will be simply due to the difference in the number of particles and the ideal gas law (Considering \( Z \)), and will be acting radially inwards towards cluster 2, as it can be seen from Figure 2a below:

![Figure 2](image_url)

\textbf{Figure 2.} (a) Pairwise interaction of 2 clusters via the radial force field in Equation (13). (b) Volume element that shows the gradient in the number of fluid particles (fluid particles in blue and cross-section of cluster 1 in purple) that generates a force acting on cluster 1.

Then, the magnitude of this force (i.e., interaction force) that is due to the local difference in the number of particles (analogous to two gases with different numbers of particles hitting a large wall/interface from each side (i.e., cluster 1 cross-section)) is given by

\[
F_n = \frac{Z(\langle N_2 - N_1 \rangle)}{d \langle r' \rangle} k_B T = \frac{k_B T}{\langle r' \rangle} \left| \frac{d \langle r' \rangle}{d \langle ZN \rangle} \right|,
\]

(10)
Now, using Equation (9),
\[ \frac{d\langle r' \rangle}{d(ZN)} = \frac{\langle r' \rangle}{Z}, \]  
(11)

Note that \( \langle r' \rangle \) is not the root-mean-squared displacement, but rather the root-mean-squared relative distance. Finally, using (10) and (11), the magnitude of the force is
\[ F_n = \frac{k_B T}{\langle r' \rangle}, \]  
(12)

Note that Equation (12) is dependent on \( Z \) and the quantity \( \frac{\langle r' \rangle}{Z} \) defines the expected range for which this force is relevant. This is to be determined experimentally (a critical displacement \( \langle r' \rangle \), where \( Z \) suffers a discontinuity, due to the phase separation assumed in this model) and it has been reported in some situations to be up to the order of a few \( \mu \text{m} \) [15,19]. Further, note that the gradient of number of particles in Equation (9); the density gradient; and therefore, the heterogeneity of this weakly interacting fluid is expected to be relevant up to the same scale of \( R \). That is, unlike an ideal incompressible fluid, which would be homogeneous in density at these scales. The force \( \vec{F} \) that corresponds to (12) is given by (considering the negative sign in (9))
\[ \vec{F} = -\frac{k_B T}{\langle r' \rangle} \hat{r}', \]  
(13)
where \( \hat{r}' \) is the unit vector in the direction of the relative distance between two clusters \( r' \). This force can be included in Newton’s second law in vector form and introducing the shorter time scale \( \tau \) previously mentioned. The Langevin approach is presented as follows:
\[ m\ddot{\vec{r}} = -a\dot{\vec{r}} - \frac{k_B T}{\langle r' \rangle} \hat{r}' + \epsilon(\tau), \]  
(14)
where all the derivatives are taken with respect to \( \tau \). Further, \( \vec{r} \) is the displacement vector of the cluster as measured from the origin. The first term to the right of the equals sign represents the viscous drag force generated by the fluid (with \( a \) being the drag coefficient), the second term is the density gradient force introduced in this model, and the third term \( \epsilon(\tau) \) is the white Gaussian noise [36] generally assumed in Brownian Motion. In the overdamped case (low-Reynolds regime), the inertial term (with \( m \) being the mass of the diffused particle) in the left-hand side of the equation is zero. Then, the simplified Newton’s equation only in the radial relative direction \( r' \) is
\[ 0 = -a\dot{r} - \frac{k_B T}{\langle r' \rangle} r + \epsilon(r', \tau), \]  
(15)

Note that since the force in (13) acts only in the radial direction, in the other two dimensions perpendicular to \( r' \) the motion will be a regular Brownian motion. Further, in these two other dimensions the particles distribution at local equilibrium is uniform, which means there exists spherical symmetry (i.e., the particle distribution translates to the random motion of a sphere in the radial direction). Additionally, since we are dealing with nonaccelerated frames, \( \vec{r} \) can be replaced by \( \hat{r} \) (i.e., displacement measured from the origin). In terms of the magnitude \( r \) (i.e., distance from the origin to a cluster), Equation (16) is
\[ 0 = -ar - \frac{k_B T}{\langle r' \rangle} sgn(r) + \epsilon(r, \tau), \]  
(16)

Note that the magnitude of the second term to the right is a constant since local equilibrium is assumed and \( \frac{\langle r' \rangle}{Z} \) is a constant observable (as a comment, this also means...
that for the proposed stochastic process white Gaussian noise can be used [36], since inside this local equilibrium domain, density is uniform). The process in (16) has already been studied in [37–40]. There are a couple of remarks here. First, the propagator (i.e., initial condition given by delta function centered at \(r_0\)) as solved in [38] by setting up the corresponding Fokker–Planck equation to (16) is given by

\[
P(r, \tau) = \frac{e^{-\frac{1}{4} \tau}}{2\sqrt{\pi \tau}} \exp\left(\frac{-|r| + |r_0|}{2} - \frac{(r - r_0)^2}{4\tau}\right) + \frac{\exp(-|r|)}{4} \left[1 + erf\left(\frac{\tau - |r| - |r_0|}{2\sqrt{\tau}}\right)\right],
\]

(17)

According to [39], a time-dependent diffusivity can be defined, and a modified form of the fluctuation–dissipation theorem holds, where temperature depends on time \(\tau\) (i.e., out of equilibrium process). Further, in this time scale, the process is instantaneously diffusive [39]. Then, when \(\tau \to t\), the fluctuation–dissipation theorem holds, where an average in time \(\tau\) is taken and the diffusivity and temperature are replaced by averages. This means the diffusivity \(D\) is given by

\[
D = \frac{k_B T}{6\pi \eta R},
\]

(18)

where \(D\) and \(T\) are average in the time scale \(\tau\), and observables in the time scale \(\tau_e\).

Since this process is diffusive, one has that the root-mean-squared radial displacement of a single cluster is \(\sqrt{2Dt}\) (there is only one degree of freedom; therefore, it is not dependent on dimension). Moreover, \(\tau\) has been replaced by \(t\). This is the long time limit and, for example, (as an estimation) if \(R \sim 10^{-9}\) m typically \(\tau_e \sim 100\ \tau\). (Note that the use of \(t\) in equations is for practical purposes as \(t\) denotes the experimentally measured time and \(\tau_e = t\), as defined above). Given that local equilibrium is assumed in the \(\tau_e\) scale, the process updates adiabatically. The mean relative displacement to a different cluster would require including a factor of \(\sqrt{2}\) as is shown in (19) [34]. Finally, the root-mean-squared relative distance traveled as measured from cluster 2 is required (therefore, a factor of \(\frac{1}{2}\) is also included in (19).

\[
\langle r' \rangle_Z = \sqrt{2} \sqrt{\frac{2D_{\text{eff}}}{2D}} t = \sqrt{\left(\frac{D}{Z^2}\right)} t,
\]

(19)

The stationary solution to the Fokker–Planck equation corresponding to (16) is the local equilibrium solution and is given by the Boltzmann–Gibbs distribution [38]. The argument of the Boltzmann–Gibbs distribution is the potential \(V(r)\) derived from the force in (13), which is

\[
V(r) = \frac{k_B T}{\sqrt{D_{\text{eff}} t}} r,
\]

(20)

where \(D_{\text{eff}} = \frac{D}{Z^2}\). Now, the Boltzmann–Gibbs distribution is

\[
p(\vec{r}) = Ae^{-\frac{V(r)}{k_B T}},
\]

(21)

where \(A\) is a constant. Note that the potential is linear in the absolute value of the position. The full stationary probability distribution in the radial direction is, therefore (the \(k_B T\) term cancels),

\[
p(\vec{r}, t) \propto e^{-\frac{r^2}{2D_{\text{eff}} t}},
\]

(22)

with \(r = |\vec{r}|\). The expression in (22) is valid from 0 up to \(t_c\), where, as mentioned at the beginning of this section, there is some \(\langle r' \rangle_c\) (with \(\langle r' \rangle_c = \sqrt{D_{\text{eff}} t_c}\). Note that \(D_{\text{eff}}\) suffers a discontinuity here) or, correspondingly, some \(t_c\) (should be determined experimentally and could be a very large value) at which the gaslike phase ends abruptly and the liquid phase starts. In general, in these types of phase separations, the compressibility factor \(Z\)
suffers a vast discontinuity [30]. It generally jumps from a value close to 1 to a very small value [30] that is reminiscent of a liquid. Of course, this indicates that interactions between molecules in the liquid phase are a lot stronger than in the gaslike phase [33]. Now, since \( Z \) has a very large decrease, \( D_{\text{eff}} \) has a very large increase. It (as specified in the assumptions of this model) can be realized that the magnitude of the force in (13) will be several orders of magnitudes less (high \( D_{\text{eff}} \)) than the viscous drag force term in (16). This means, for this regime, that the force in (16) can be safely ignored. After \( t_c \), the Langevin equation becomes

\[
\alpha \dot{\vec{r}} = \epsilon (t'),
\]

or the corresponding Fokker–Planck equation being the diffusion equation [41]

\[
\frac{\partial p(\vec{r},t')}{\partial t'} = D_{\text{effL}} \nabla^2 p(\vec{r},t'),
\]

which should be solved with (22) evaluated at \( t_c \) as initial condition and \( t' = t - t_c \). To account for the discontinuity of \( Z \), the effective diffusivity for this regime will be denoted by

\[
D_{\text{effL}} = \frac{D}{Z_{L}^{2}},
\]

and the effective diffusivity in the gaslike phase will be denoted by

\[
D_{\text{effG}} = \frac{D}{Z_{G}^{2}},
\]

Since \( D_{\text{effG}} < < D_{\text{effL}} \), Equation (22) can be seen as a delta function when evaluated at \( t_c \) and used as an initial condition to solve (24), meaning that after \( t_c \) the probability density will have some transition probability that will evolve rapidly towards a Gaussian, which is

\[
p(\vec{r},t') \sim \frac{1}{(4\pi D_{\text{effL}} t')^{2}} e^{-\frac{\vec{r}^2}{4D_{\text{effL}} t'}},
\]

From now on, the probability distribution in (22) will be denoted as \( p_G(\vec{r},t) \) (gaslike phase) and the full solution to (24) with the initial condition being \( p_G(\vec{r},t_c) \) will be denoted as \( p_L(\vec{r},t') \) (liquid phase).

The description above gives an accurate time evolution of the probability distribution of the position of the clusters for the two different regimes (the gaseous regime with \( D_{\text{effG}} \) and the liquid regime with \( D_{\text{effL}} \)). However, it does not tell you which probability distribution you should use at any given time (over a single scale over a time interval \( t \)) \( p_G(\vec{r},t) \) or \( p_L(\vec{r},t') \). This means that, at time \( t < t_c \), a cluster may be located at a position \( \vec{r} \) such that it is either in the gaslike phase or in the liquid phase. The same happens at \( t > t_c \). This means that the gaslike and liquid phases can be regarded as two possible states of the system, with no preference over one or the other. Of course, this should be accounted for when taking averages of observables, one should take the average over all possible states. Therefore, the mean squared displacement for this process will be given by

\[
\langle r(t)^2 \rangle = 0.5 \int_{-\infty}^{\infty} |\vec{r}|^2 p_G(\vec{r},t) d\vec{r} + \int_{-\infty}^{\infty} |\vec{r}|^2 p_L(\vec{r},t) d\vec{r},
\]

which the simplifying of gives

\[
\langle r(t)^2 \rangle = 0.5(6D_{\text{effG}}t + 6D_{\text{effL}}t),
\]

Rearranging terms,

\[
\langle r(t)^2 \rangle = 6\left[0.5\left(D_{\text{effG}} + D_{\text{effL}}\right)\right]t,
\]

where \( D_{\text{avg}} \) can be seen as an average diffusivity and is given by
\[ D_{\text{avg}} = 0.5 \left( D_{\text{effG}} + D_{\text{effL}} \right), \quad (31) \]

Equation (30) can be rewritten as
\[ \langle r(t)^2 \rangle = 6D_{\text{avg}} t, \quad (32) \]

Equation (32) leads to interpret the whole Brownian process discussed until now in two different regimes with diffusivities \( D_{\text{effG}} \) and \( D_{\text{effL}} \) as a single rescaled process, such that the standard deviation of the PDF is given by (32).

Finally, from (32) and the analysis up until before Equation (28), it can be inferred that the observed probability distribution of position \( r \) of the cluster before \( t_c \) will be given by the normalized Equation (22) and after \( t_c \) by the normalized solution to (24) with the initial condition of (22). Then, both probability distributions should be rescaled such that their mean squared displacement (i.e., standard deviation squared) at any given time is given by (32).

3. Initial Theoretical and Experimental Considerations

A few initial remarks concerning the proposed model and some relevant experimental observations are summarized here. In experiment [15], the Brownian motion of colloidal beads on phospholipid bilayer (DLPC) tubes (1-D) was studied. Lipid bilayers tend to have weak intermolecular interactions between “lipids tails” [42]. (London dispersion forces are the type of molecular interactions occurring between lipids). The weaker these intermolecular interactions, the more flexible the bilayers (i.e., membranes) and vice versa [42]. Therefore, (also motivated by the role of weak interactions in phase separation in different studies such as [29]), it is suggested here that the Brownian particle–fluid long-range interactions can lead to a phase separation and the whole machinery developed above can be applied.

First, note that what is being measured in the experiment of phospholipid bilayer tubes [15] (with no cholesterol, where Brownian yet non-Gaussian diffusion is reported) is \( D_{\text{avg}} \). Then, according to our model, we can write this quantity as
\[ D_{\text{avg}} = 0.5 \left( \frac{D}{Z_G^2} + \frac{D}{Z_L^2} \right), \quad (33) \]

Approximating the lipids tails in the gaslike phase as an ideal gas (i.e., almost no interaction), meaning \( Z_G \sim 1 \), then
\[ D_{\text{avg}} = 0.5 \left( D + D_{\text{effL}} \right), \quad (34) \]

As mentioned before, \( Z_L \) is a small number. This means \( D_{\text{avg}} \gg D \), which is what was found in the experiment [15] (i.e., unusually high diffusivity).

The Lennard-Jones potential introduced in this model causes a reduction in local pressure in the liquid (meaning the local pressure is less than the external pressure at which the experiment is being conducted). This means that determination of \( Z_L \) requires full knowledge of the effective local pressure experienced in the liquid at the phase separation point, which is not an easy task to estimate theoretically; therefore, a direct calculation of \( D_{\text{avg}} \) is not performed here. (However, it could in principle be done). In any case, the value of \( D_{\text{avg}} = 0.40 \mu m^2/s \) and \( D \) reported in this experiment [15] can be used to calculate the value of \( Z_L \) and check whether it is a reasonable value for a liquid at room temperature, low pressures, and far from the critical point. \( D \) is calculated by using Einstein’s relation:
\[ D = \frac{k_BT}{6\pi \eta_e R}, \quad (35) \]

where \( T \) is temperature, \( k_B \) is Boltzmann constant, \( R \) is the radius of the Brownian particle, and \( \eta_e \) is the extensional viscosity of the fluid. Extensional viscosity considers shear
viscosity and bulk viscosity, which should be the case for a compressible fluid [43,44]. For a Newtonian fluid, $\eta_e$ is given by [44]:

$$\eta_e = 3\eta,$$  \hspace{1cm} (36)

where $\eta$ is the shear viscosity of the fluid. Using (35) and (36),

$$D = \frac{k_B T}{18\pi \eta R},$$  \hspace{1cm} (37)

4. Application of the Model

In order to compare model predictions with experimental results, experimental data taken from Wang et al. [15] are employed. In this experiment, the temperature is reported to be $T = 22 \, ^\circ C$, the viscosity of the media $\eta$ is reported to be $\approx 100$ times higher than that of bulk water, and the radius of the Brownian particle is $R = 50 \, nm$. Therefore, with the conditions of this experiment, Equation (37) is used to find that $D \sim 0.014 \, \mu m^2 \times s^{-1}$. Further, using this result and Equations (25) and (34), it is calculated indirectly that $D_{eff} \sim 0.79 \, \mu m^2 \times s^{-1}$ and $Z_L \sim 0.13$ (note that $Z_L$ is a reasonable value for a liquid at room temperature and low pressures [33]). Further, note that $D_{avg} \gg D$, since $0.40 \gg 0.014$.

Regarding the temporal evolution of the probability distribution, in this experiment, $t_c \sim 4 \, s$. This means that up until $t = 4 \, s$, the probability density function (PDF) in the displacement $x$ will be given by the analogous 1D expression to (22) (i.e., Laplacian distribution) with a diffusivity of $D_{avg}$. Starting at $t = 4 \, s$, a numerical implementation of the proposed model in Mathematica shows that after $\sim 2 \, s$ (i.e., $t \sim 6 \, s$), the PDF (22) had transitioned towards a 1D Gaussian in the displacement $x$ (see Figure 3a,b below). This is precisely what was observed in this experiment [15].

![Figure 3](image-url)
A brief description of the figures and data is given here. The solid line curves of Figure 3a below were obtained from the proposed model. At \( t = 60 \text{ ms} \), \( t = 0.6 \text{ s} \), and \( t = 3 \text{ s} \), the corresponding 1D un-normalized Equation (22) was used. It was rescaled such that its mean square displacement is given by (32) 1D analogous (i.e., \( \langle r(t)^2 \rangle = 2D_{\text{avg}}t \)) with \( D_{\text{avg}} = 0.40 \mu\text{m}^2 \times \text{s}^{-1} \).

Furthermore, at \( t = 5.8 \text{ s} \), the differential Equation (24) in 1D with \( D_{\text{eff}}L \sim 0.79 \mu\text{m}^2 \times \text{s}^{-1} \) was solved numerically in Mathematica. The initial condition was taken to be the 1D un-normalized Equation (22) evaluated at time \( t_c = 4 \text{ s} \) (which is the critical time reported by Wang et al., where the Laplacian distribution starts changing towards a Gaussian) with diffusivity \( D_{\text{eff}} = 0.014 \mu\text{m}^2 \times \text{s}^{-1} \). The solution was rescaled such that \( \langle r(t)^2 \rangle = 2D_{\text{avg}}t \).

As a result, the transition probability \( p_L(x,t) \) at \( t = 5.8 \text{ s} \) is found to be

\[ p_L(x) \propto 0.5e^{35.55-0.45x} \left\{ \text{erfc}(5.96-0.038x) + e^{0.91x}\text{erfc}(5.96+0.038x) \right\}, \tag{38} \]

where \( x \) is the 1-D displacement of the bead and

\[ \text{erfc}(w) = 1 - \text{erf}(w), \tag{39} \]

It is interesting to note that (38) plotted in Figure 3a agrees very well with the experimental data points. With respect to the experimental data points, they were obtained from [15]. Linear regression analysis was performed to fit the experimental data points in Figure 3a. By fixing the lowest-order parameter, a calculation of the percentage error of the highest-order coefficient/parameter between the theoretical prediction and the experimental best fit was performed at the corresponding times. At \( t = 60 \text{ ms} \), the percentage error was found to be 0.28%; at \( t = 0.6 \text{ s} \), the percentage error was found to be 2.0%; at \( t = 3 \text{ s} \), the percentage error was found to be 3.4%; and at \( t = 5.8 \text{ s} \), the percentage error was found to be 3.5%.

From Figure 3b, it can be inferred that at some point before 200 d, the slope of the brown and gray lines changes. Note that this figure is plotted in a log–log scale, which means the graph shows the \( \log(\log(p(x,t))) \) in the y-axis vs. the \( \log(\frac{x}{5.8}) \) in the x-axis. This indicates exponential decay for the time passed after \( t_c = 4 \text{ s} \) of \( t = 1 \text{ s} \) and \( t = 1.5 \text{ s} \). At \( t = 2 \text{ s} \), the slope of the red line is constant, which indicates Gaussian behavior for all \( x \) up to \( x = 200 \text{ d} \).

From the figures above, it can be concluded that the theoretical model agrees with the experiment. Percentage error in the parameters do not exceed 3.5%. Further, besides the theoretical prediction of the emergence of a Laplacian distribution and an eventual Gaussian distribution in the particle displacements, it is remarkable that the transition probability at \( t = 5.8 \text{ s} \) and the transition time (which emerge from ideal scenarios) are an accurate representation of the experimental data.

Finally, as a second part of the experiment, the membranes of the fluid were filled with cholesterol. Everything else was held constant and no exponential distribution was observed. As it has been observed before, cholesterol provides rigidity to the membranes by strengthening molecular interactions between lipids [45]. Since not enough energy is provided to break those interactions by the interaction Brownian particle-fluid (via the Lennard-Jones potential proposed in this model), no phase separation occurs and one is in the regime of “regular Brownian motion”. Further, in this case, the diffusivity was observed to be \( D = 0.012 \mu\text{m}^2 \times \text{s}^{-1} \). This should be the case, since by using Einstein’s relation (37) and considering the 20% increase in viscosity due to cholesterol [15], the theoretical value of the diffusivity is exactly \( 0.012 \mu\text{m}^2 \times \text{s}^{-1} \).

5. Conclusions

A new model to explain Brownian yet non-Gaussian behavior is proposed, by including molecular interactions. Two regimes in a weakly interacting and highly compressible fluid are presented. Long-range interactions Brownian-particle fluid cause a phase separa-
tion for short distances. In the first regime, the fluid behaves similar to a real gas interacting with an interface with no shear viscosity. If only pairwise cluster interaction via the fluid is considered, a Laplace distribution on the displacements of a phase-separated cluster is derived. In the second regime, the fluid behaves as a liquid, but with very high diffusivity (i.e., due to low compressibility/high mean free path caused by the separation of fluid particles via Lennard-Jones potential). The Laplace distribution evolves starting at a time $t_c$ via a transition probability (38) rapidly into a Gaussian. (All the pdfs are rescaled, such that $\langle r(t)^2 \rangle$ is given by the 1D analogous of (32), since the cluster might be in either the gaslike or in the liquid phase at some point in time).

The experimental data obtained by Wang et al. are used to apply the model. It is shown that the theoretical model explains via a phase separation the physical cause for an unexpectedly high diffusivity. In addition, the lack of a phase separation when using cholesterol and the compressibility and shear deformation of the liquid (i.e., considering extensional viscosity) leads to a different and smaller value of the diffusivity that fully agrees with the experimentally reported value. Further, the pdfs for different times predicted by the model are compared with the experimental data and are found to be in excellent agreement. At $t_c = 4$ s, the pdf starts transitioning away from being exponential and, after approximately 2 s, is found to be Gaussian, as reported in the experiment.

Finally, this approach links theoretical concepts in the previous research of fluid behavior [44,46,47] and theory of interfaces [31] to Brownian motion, which indicate that there are strong motives to expand studies of highly compressible liquids (in colloidal suspensions) and their molecular interactions with a Brownian particle (i.e., different effective/interaction potentials to the one in (2) can be proposed to model different regimes at shorter time scales, where different types of diffusion processes may occur). Then, these effects can be incorporated into Langevin dynamics without the need of complicated mathematical modifications to noise terms.

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Appendix A

Table A1. Nomenclature used in this paper with SI units.

| Symbol | Description |
|--------|-------------|
| $\tau, \tau_e, \tau$ | time scale in which the gaseouslike state of the fluid relaxes towards equilibrium, equilibrium time scale (experimental scale), universal variable for time (s) |
| $\varepsilon$ | dispersion energy. Depth of Lennard-Jones potential well (J) |
| $\sigma$ | Van der Waals radius (m) |
Table A1. Cont.

| Symbol | Description |
|--------|-------------|
| \( r_{ij} \) | distance between a molecule of the Brownian particle and fluid molecule (m) |
| \( r'_m, z'_r, < r' > \) | classically measured distance between the center of a Brownian particle and fluid molecule, classically measured distance between the spherical interface of a Brownian particle and the center of a different Brownian particle, distance as measured from the center of a Brownian particle and the interface of a different Brownian particle (mean radial relative distance, m) |
| \( R \) | radius of particle being diffused (m) |
| \( z \) | distance as measured from the interface of Brownian particle (m) |
| \( U(r_{ij}) \) | Attractive term of 6-12 Lennard-Jones potential energy acting between two molecules (J) |
| \( V(z) \) | cubic part of a 3-9 Lennard-Jones potential energy (J) |
| \( \rho_s \) | number density of the fluid at the spherical interface of Brownian particle (m\(^{-3}\)) |
| \( \rho_L \) | number density of the fluid in the gel-like phase (m\(^{-3}\)) |
| \( \rho(z) \) | density profile of the fluid in the gaseouslike phase (m\(^{-3}\)) |
| \( k_B T \) | Boltzmann constant and temperature of the system (J \( \times \) K\(^{-1}\)) (K) |
| \( \bar{\rho} \) | number density profile of the fluid measured with respect to \( \rho_L \) (m\(^{-3}\)) |
| \( \rho_b \) | number density of the fluid at the spherical interface of a Brownian particle measured with respect to \( \rho_L \) (m\(^{-3}\)) |
| \( N \) | number of fluid molecules inside a cluster (nondimensional) |
| \( V \) | cylindrical volume enclosed by the cross-section of the cluster, with height \( d(r_{fi}) \) (m\(^3\)) |
| \( Z \) | compressibility factor, measures deviation from ideal gas law in the fluid (nondimensional) |

Table A2. Nomenclature used in this paper.

| Symbol | Description |
|--------|-------------|
| \( ZN_1 \) | modified number of fluid molecules (considers fluid molecules interactions) from one side of the cross-section of the cluster (nondimensional) |
| \( ZN_2 \) | modified number of fluid molecules (considers fluid molecules interactions) from opposite side of the cross-section of the cluster (nondimensional) |
| Symbol | Description |
|--------|-------------|
| $\frac{dN}{d\langle r \rangle}$ | number density of the fluid/gradient of number of fluid molecules in the radial direction ($m^{-1}$) |
| $\frac{d(ZN)}{d\langle r \rangle}$ | gradient of modified number of fluid molecules in the radial direction (considers fluid molecules interactions) ($m^{-1}$) |
| $r', r, \vec{r}$ | radial relative distance between two clusters, radial distance/distance as measured from origin (laboratory frame), radial relative displacement, displacement as measured from origin. All of them random variables (m) |
| $F_n, \vec{F}$ | magnitude of interaction force between two clusters of molecules, vector form of the interaction between two clusters of molecules (N) |
| $\epsilon(\tau)$ | white Gaussian noise (N) |
| $m$ | mass of the Brownian particle (kg) |
| $\alpha$ | resistive constant ($N \times s \times m^{-1}$) |
| $\eta$ | shear viscosity of the fluid at equilibrium (in both phases) (P) |
| $D$ | diffusivity of the fluid at equilibrium ($m^2 \times s^{-1}$) |
| $r_0$ | initial position of diffusing particles (m) |
| $P(), p()$ | denotes probability density functions and/or propagators (nondimensional) |
| $D_{eff}$ | reduced diffusivity due to molecular interactions in the phase-separated fluid. ($m^2 \times s^{-1}$) |
| $V(\vec{r})$ | potential energy acting on the clusters ensemble at equilibrium (J) |
| $Z_L, Z_G$ | compressibility factor in the gel-like phase, compressibility factor in the gaseous-like phase of the phase-separated fluid. (non-dimensional) |
| $D_{effL}, D_{effG}$ | modified diffusivity in the gel-like phase due to fluid molecules interactions, modified diffusivity in the gaseous-like state due to fluid molecules interactions. ($m^2 \times s^{-1}$) |
| $p_G(\vec{r}, t)$ | probability density function for the gaseous-like state (nondimensional) |
| $p_L(\vec{r}, t)$ | probability density function for the gel-like state (nondimensional) |
| $t_c$ | experimentally measured critical time at which $p_G(\vec{r}, t)$ changes towards $p_L(\vec{r}, t)$ (s) |
| $\langle r \rangle_c$ | experimentally measured critical mean radial distance $p_G(\vec{r}, t)$ changes towards $p_L(\vec{r}, t)$ (m) |
| $t'$ | time measured with respect to $t_c$ (s) |
| $\eta_e$ | extensional viscosity (P) |
| $D_{avg}$ | average of $D_{effL}$ and $D_{effG}$ ($m^2 \times s^{-1}$) |
Table A2. Cont.

| Symbol     | Description                                                                 |
|------------|-----------------------------------------------------------------------------|
| $\langle r(t)^2 \rangle$ | mean squared displacement for the whole process (m$^2$)                     |
| $x$        | 1D experimental displacement of a diffusing bead (m)                        |
| $d$        | diameter of diffusing bead (m)                                              |

References

1. Koo, P.K.; Mochrie, S.G. Systems-level approach to uncovering diffusive states and their transitions from single-particle trajectories. *Phys. Rev. E* 2016, 94, 52412. [CrossRef] [PubMed]
2. Gillespie, D.T.; Seitaridou, E. *Simple Brownian Diffusion: An Introduction to the Standard Theoretical Models*; Oxford University Press: Oxford, UK, 2013.
3. Feynman, R.P.; Leighton, R.B.; Sands, M. *The Feynman Lectures on Physics: The New Millennium Edition: Mainly Mechanics, Radiation, and Heat*; Basic Books: New York, NY, USA, 2011; Volume 1.
4. Liu, D.L. Particle deposition onto enclosure surfaces. In *Developments in Surface Contamination and Cleaning*; Elsevier: Amsterdam, The Netherlands, 2010; pp. 1–56.
5. Malý, P.; Lüttig, J.; Turkin, A.; Dostál, J.; Lambert, C.; Brixner, T. From wavelike to sub-diffusive motion: Exciton dynamics and interaction in squaraine copolymers of varying length. *Chem. Sci.* 2020, 11, 456–466. [CrossRef] [PubMed]
6. Sposini, V.; Metzler, R.; Oshanin, G. Single-trajectory spectral analysis of scaled Brownian motion. *New J. Phys.* 2019, 21, 73043. [CrossRef]
7. Narumi, T.; Hidaka, Y. Slow diffusive structure in Nikolaevskii turbulence. *Phys. Rev. E* 2020, 101, 22202. [CrossRef] [PubMed]
8. Chakraborty, S.; Kumar, H.; Dasgupta, C.; Maiti, P.K. Confined water: Structure, dynamics, and thermodynamics. *Accounts Chem. Res.* 2017, 50, 2139–2146. [CrossRef] [PubMed]
9. Kanazawa, K.; Sano, T.G.; Cairoli, A.; Baule, A. Loopy Lévy flights enhance tracer diffusion in active suspensions. *Nature* 2020, 579, 364–367. [CrossRef]
10. Ribeiro, H.; Ferreira, W.; Potiguar, F.Q. Trapping and sorting of active matter in a periodic background potential. *Phys. Rev. E* 2020, 101, 32126. [CrossRef]
11. Hanasaki, I.; Shoji, T.; Tsuibo, Y. Regular assembly of polymer nanoparticles by optical trapping enhanced with a random array of Si needles for reconfigurable photonic crystals in liquid. *ACS Appl. Nano Mater.* 2019, 2, 7637–7643. [CrossRef]
12. Rudnizky, S.; Khamis, H.; Malik, O.; Melamed, P.; Kaplan, A. The base pair-scale diffusion of nucleosomes modulates binding of transcription factors. *Proc. Natl. Acad. Sci. USA* 2019, 116, 12161–12166. [CrossRef]
13. El-Zant, A.A.; Freundlich, J.; Combes, F.; Halle, A. The effect of fluctuating fuzzy axion haloes on stellar dynamics: A stochastic model. *Mon. Not. R. Astron. Soc.* 2020, 492, 877–894. [CrossRef]
14. Cincotta, P.M.; Giordano, C.M.; Marti, J.G.; Beauge, C. On the chaotic diffusion in multidimensional Hamiltonian systems. *Celest. Mech. Dyn. Astron.* 2018, 130, 1–23. [CrossRef]
15. Wang, B.; Anthony, S.M.; Bae, S.C.; Granick, S. Anomalous yet brownian. *Proc. Natl. Acad. Sci. USA* 2009, 106, 15160–15164. [CrossRef] [PubMed]
16. Chubykinsky, M.V.; Slater, G.W. Diffusing diffusivity: A model for anomalous, yet Brownian, diffusion. *Phys. Rev. Lett.* 2014, 113, 98302. [CrossRef]
17. Chechkin, A.V.; Seno, F.; Metzler, R.; Sokolov, I.M. Brownian yet non-Gaussian diffusion: From superstatisitcs to subordination of diffusing diffusivities. *Phys. Rev. X* 2017, 7, 21002. [CrossRef]
18. Nygård, K. Local structure and density fluctuations in confined fluids. *Curr. Opin. Colloid Interface Sci.* 2016, 22, 30–34. [CrossRef]
19. Wang, B.; Kuo, J.; Bae, S.C.; Granick, S. When Brownian diffusion is not Gaussian. *Nat. Mater.* 2012, 11, 481–485. [CrossRef]
20. Slezak, J.; Burov, S. From diffusion in compartmentalized media to non-Gaussian random walks. *Sci. Rep.* 2021, 11, 1–18.
21. Bialas, K.; Luczka, J.; Hänggi, P.; Spiechowicz, J. Colossal Brownian yet non-Gaussian diffusion induced by nonequilibrium noise. *Phys. Rev. E* 2020, 102, 42121. [CrossRef]
22. Sposini, V.; Chechkin, A.V.; Seno, F.; Pagnini, G.; Metzler, R. Random diffusivity from stochastic equations: Comparison of two models for Brownian yet non-Gaussian diffusion. *New J. Phys.* 2018, 20, 43044. [CrossRef]
23. Yin, Q.; Li, Y.; Marcheson, F.; Nayak, S.; Ghosh, P.K. Non-Gaussian normal diffusion in low dimensional systems. *Front. Phys.* 2021, 16, 1–14. [CrossRef]
24. Luo, L.; Yi, M. Non-Gaussian diffusion in static disordered media. *Phys. Rev. E* 2018, 97, 42122. [CrossRef] [PubMed]
25. Hidalgo-Soria, M.; Barkai, E. Hitchhiker model for Laplace diffusion processes. *Phys. Rev. E* 2020, 102, 12109. [CrossRef] [PubMed]
26. Toyoda, T.; Head, D.A.; Schmidt, C.F.; Mizuno, D. Non-Gaussian athermal fluctuations in active gels. *Soft Matter* 2011, 7, 3234–3239. [CrossRef]
27. Weeks, E.R.; Crocker, J.C.; Levitt, A.C.; Schofield, A.; Weitz, D.A. Three-dimensional direct imaging of structural relaxation near the colloidal glass transition. *Science* 2000, 287, 627–631. [CrossRef]

28. Eaves, J.D.; Reichman, D.R. Spatial dimension and the dynamics of supercooled liquids. *Proc. Natl. Acad. Sci. USA* 2009, 106, 15171–15175. [CrossRef]

29. Sahli, L.; Renard, D.; Solé-Jamault, V.; Giuliani, A.; Boire, A. Role of protein conformation and weak interactions on γ-gliadin liquid-liquid phase separation. *Sci. Rep.* 2019, 9, 13391.

30. Alberti, S.; Gladfelter, A.; Mittag, T. Considerations and challenges in studying liquid-liquid phase separation and biomolecular condensates. *Cell* 2019, 176, 419–434. [CrossRef] [PubMed]

31. De Gennes, P.G. Some effects of long range forces on interfacial phenomena. *J. Phys. Lett.* 1981, 42, 377–379. [CrossRef]

32. Hernández, E.; MW Cole y M. Boninsegni. *Phys. Rev. B* 2003, 68, 1254181.

33. Wilhelm, E.; Letcher, T. *Volume Properties: Liquids, Solutions and Vapours*; Royal Society of Chemistry: London, UK, 2014.

34. Atkins, P.; De Paula, J. *Physical Chemistry for the Life Sciences*; Oxford University Press: Oxford, UK, 2011.

35. Chang, R. *Physical Chemistry for the Biosciences*; University Science Books: Sausalito, CA, USA, 2005.

36. Vamoș, C.; Crăciun, M. Separation of components from a scale mixture of Gaussian white noises. *Phys. Rev. E* 2010, 81, 51125. [CrossRef] [PubMed]

37. De Gennes, P.G. Brownian motion with dry friction. *J. Stat. Phys.* 2005, 119, 953–962. [CrossRef]

38. Touche, H.; Van der Straeten, E.; Just, W. Brownian motion with dry friction: Fokker–Planck approach. *J. Phys. A Math. Theor.* 2010, 43, 445002. [CrossRef]

39. Kawarada, A.; Hayakawa, H. Non-Gaussian velocity distribution function in a vibrating granular bed. *J. Phys. Soc. Jpn.* 2004, 73, 2037–2040. [CrossRef]

40. Murayama, Y.; Sano, M. Transition from Gaussian to non-Gaussian velocity distribution functions in a vibrated granular bed. *J. Phys. Soc. Jpn.* 1998, 67, 1826–1829. [CrossRef]

41. Bian, X.; Kim, C.; Karniadakis, G.E. 111 years of Brownian motion. *Soft Matter* 2016, 12, 6331–6346. [CrossRef]

42. Marrink, S. Berger O, Tieleman P, Jahnig F. Adhesion forces of lipids in a phospholipid membrane studied by molecular dynamics simulations. *Biophys. J.* 1998, 74, 931–943. [CrossRef]

43. Mezger, T. The rheology handbook. In *The Rheology Handbook*; Vincentz Network: Hannover, Germany, 2020.

44. Chiang, Y.W.; Shimoyama, Y.; Feigenson, G.W.; Freed, J.H. Dynamic molecular structure of DPPC-DLPC-cholesterol ternary lipid system by spin-label electron spin resonance. *Biophys. J.* 2004, 87, 2483–2496. [CrossRef]

45. Straube, A.V.; Kowalik, B.G.; Netz, R.R.; Höfling, F. Rapid onset of molecular friction in liquids bridging between the atomistic and hydrodynamic pictures. *Commun. Phys.* 2020, 3, 1–11. [CrossRef]

46. Stephan, S.; Thol, M.; Vrabec, J.; Hasse, H. Thermophysical properties of the Lennard-Jones fluid: Database and data assessment. *J. Chem. Inf. Model.* 2019, 59, 4248–4265. [CrossRef]