Non-Gaussian information of heterogeneity in soft matter

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received 20 March 2020; accepted in final form 29 June 2020
published online 29 July 2020

PACS 82.70.Dd - Physical chemistry and chemical physics: Colloids
PACS 64.70.pv - Equations of state, phase equilibria, and phase transitions: Colloids

Abstract - Heterogeneity in dynamics in the form of non-Gaussian molecular displacement distributions appears ubiquitously in soft matter. We address the quantification of such heterogeneity using an information-theoretic measure of the distance between the actual displacement distribution and its nearest Gaussian estimation. We explore the usefulness of this measure in two generic scenarios of random walkers in heterogeneous media. We show that our proposed measure leads to a better quantification of non-Gaussianity than the conventional ones based on moment ratios.

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The usual laws of Fickian diffusion have dominated the world of molecular transport for more than a century [1–3]. Fickian diffusion models the evolving probability distribution of the molecular displacement in a medium as a Gaussian when there is an extreme separation of time scales between the molecular solute and the atomistic solvent particles [1–4]. Since the relaxation of atomistic solvent particles is by many magnitudes faster than that of the larger solute particles, the solute dynamics is treated as a molecular dynamics with effective fluctuations due to constant thermal kicks by the bath particles. However, this is not a priori valid locally in systems with rough energy landscapes or under external potential where the microscopic motion undergoes metastability [5–7], converting energy into information [8–13].

In the presence of such microscopic heterogeneity, the molecular displacement distributions generically deviate from Gaussianity [6,14–35]. This is observed ubiquitously in soft matter [15,17,18,20,22,23,25–27,32]: from bio-molecules to glassy liquids. However, the dynamics is essentially bounded by the central limit theorem and thus the distribution reverts to a Gaussian form at timescales essentially bounded by the central limit theorem and thus thus not a priori valid locally in systems with rough energy landscapes or under external potential where the microscopic motion undergoes metastability [5–7], converting energy into information [8–13].

Also, in the presence of domains, increasing the effective polydispersity can give rise to heterogeneity in dynamics where each of the molecular hops from one domain to another corresponds to unique time scales while the intra-domain movements mostly mimic the dynamics in bulk [36]. Similar heterogeneity is seen in glassy liquids due to intermittency caused by local metastability, where motion in the vicinity of a local cage becomes arrested, and becomes diffusive again when it gets out of the cage overcoming the free energy barrier [37–39]. Thus the coexistence of competing relaxation processes leads to dynamic heterogeneity in soft matter [37–39]. This necessarily asks for validation and generalisation of Einstein-Stokes in many physical systems where relaxation processes undergo heterogeneity.

One phenomenological approach is to understand the heterogeneous diffusion in terms of an effective single particle dynamics with a diffusion spectrum, \( P(D) \), instead of a unique diffusion coefficient \( D \) [23]. Non-Gaussian probability distributions of particle displacements are thus modeled as an ensemble of diffusive processes, \( G(x,t;D) \), given by \( P_{\text{ng}}(x,t) \sim \int dD \, P(D) \, G(x,t;D) \) [23,24]. Here the weighted distribution, \( P(D) \), captures the dynamical fluctuations in molecular diffusion. Such a framework of heterogeneous diffusion has been quite successful in explaining some natural processes which earlier seemed anomalous [23,24,27–29]. From here, one ends with a surprising conclusion, that is the linearity of the mean squared displacements no longer ensures the Gaussianity of the underlying probability distribution function [23]. Thus, a system with a diffusivity spectrum, \( P(D) \), can yield \( P_{\text{ng}}(x,t) \sim \exp(-\frac{x^2}{4t}) \), leading to a MSD linear in time: \( \langle x^2 \rangle = D_{\text{eff}}t = t \int dDP(D)D \). The effective

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distribution of particle displacements $P_{\text{ng}}(x, t)$ remains non-Gaussian. Thus the actual distribution of particle displacements contains information about the heterogeneity which is masked if one approximates the diffusion by the Gaussian approximation, $P_{\text{eff}}(x, t) \approx P_{\text{ng}}^G(x, t) \sim \frac{1}{\sqrt{4\pi Dt}} e^{-x^2/4Dt}$.

In this letter, we introduce a new information-theoretic quantification to tackle the immense challenge of extracting the heterogeneity in molecular diffusion. We detect such heterogeneity in the form of non-Gaussian information, quantified in terms of the relative entropy, more efficiently than conventional approaches. We encode this in terms of entropic non-Gaussianity given by the difference between the Shannon entropy of the closest Gaussian estimate of the given function and the test non-Gaussian distribution. In the process, we separate the non-trivial information from the expected increase in the Shannon information of the Gaussian counterpart that increases naturally with elapsed time due to diffusion. We explicitly show that this new quantification leads to more efficient detection of the true non-Gaussian information than what the standard non-Gaussian parameter captures. We exemplify this in two generalised cases where the existence of dynamic heterogeneity is well known and is known to appear due to the intrinsic structural heterogeneity of the medium.

The conventional approach of quantifying non-Gaussianity in a distribution considers deviation from moment relationships which hold for a Gaussian distribution. The standard non-Gaussian parameter [40,41] is defined as

$$\alpha_2 = \begin{cases} \frac{\langle x^4 \rangle}{\langle x^2 \rangle^2} - 1 \quad \text{(in 1D)}, \\ \frac{\langle x^4 \rangle}{\langle x^2 \rangle^2} - 1 \quad \text{(in 3D)}. \end{cases}$$

This relies on the fact that for a 1D Gaussian, $\langle x^4 \rangle = 3\langle x^2 \rangle^2$, and for a 3D Gaussian, $\langle x^4 \rangle = \frac{5}{2}\langle x^2 \rangle^2$. However, the reliance of this quantity on the fourth moment necessarily limits the amount of information it captures. Quantifications involving higher moments can also be defined [40], but there is no sensible way to integrate the information in these separate quantities to form a unified picture of non-Gaussianity.

Non-Gaussianity from relative entropy: We now define our new information theoretic quantification to capture the similarity between the given non-Gaussian probability distribution, $P_{\text{ng}}$, and its closest Gaussian distribution, $P_{\text{ng}}^G$, which is obtained by minimizing the Kullback-Leibler (KL) divergence [42] between $P_{\text{ng}}$ and all Gaussian distributions $P^G$. To be specific, it could be easily shown that

$$D_{\text{KL}}(P_{\text{ng}} || P^G) = D_{\text{KL}}(P_{\text{ng}} || P_{\text{ng}}^G) + D_{\text{KL}}(P_{\text{ng}}^G || P^G),$$

where $P_{\text{ng}}^G$ is the Gaussian distribution that has the first and second moments equal to those of the $P_{\text{ng}}$ itself [43]. Since $D_{\text{KL}}(P_{\text{ng}} || P^G) \geq 0$, $D_{\text{KL}}(P_{\text{ng}} || P^G)$ in eq. (2) is minimum for $P^G = P_{\text{ng}}^G$. Thus, the KL-divergence–based quantification of non-Gaussianity of $P_{\text{ng}}$ reads as

$$\Delta S_{\text{ng}} = D_{\text{KL}}(P_{\text{ng}} || P_{\text{ng}}^G) = \int dx P_{\text{ng}}(x) \ln \left( \frac{P_{\text{ng}}^G(x)}{P_{\text{ng}}(x)} \right) = S_{\text{ng}}^G - S_{\text{ng}},$$

where $S_{\text{ng}} = S(P_{\text{ng}})$, $S_{\text{ng}}^G = S(P_{\text{ng}}^G)$, $S(P(x)) = \int dx P(x) \ln(P(x))$ being the the Shannon entropy [44].

Evidently, for a Gaussian distribution, $P_{\text{G}}$, $\Delta S_{\text{ng}} = D_{\text{KL}}(P_{\text{ng}} || P_{\text{G}}^G) = 0$. This quantification has earlier been proposed in analysing non-Gaussianity in quantum optical states as Negentropy [45].

Non-Gaussian walker with fluctuating diffusivities: We first consider a typical case of molecular heterogeneity adopting the phenomenological picture discussed in refs. [23,24] when the single particle dynamics is estimated as a convolution of diffusive processes with the distribution of diffusivities, $P(D)$ instead of a single diffusion coefficient:

$$P_{\text{ng}}(x, t) = \int dDG(x, t; D)P(D)$$

where the normal diffusive process be given by

$$G(x, t; D) = \frac{1}{\sqrt{2\pi Dt}} e^{-x^2/2Dt}.$$ The pecularity of this kind of diffusion is that it leads to the linear mean-squared displacements as in normal diffusion [23]. But in contrast to normal diffusion, it has a diffusion spectrum to represent its single particle dynamics. Using the concavity of the logarithmic function, $\ln(\int dDG(x, t; D)P(D)) \geq \int dDP(D) \ln(G(x, t; D))$ [47] and the expression of the entropy of $G(x, t; D)$, $-\int_C dG(x, t; D) \ln(G(x, t; D)) = \frac{1}{2}(1 + \ln(2\pi Dt))$, we obtain

$$S_{\text{ng}}(t) = \langle D \rangle \left\{ \frac{1}{D} + \frac{1}{2}\left(1 - \ln(2\pi) + \ln(Dt)\right) \right\} - F_1(D),$$

where $F_1(D)$ is a strictly positive quantity. It must be noted that $F_1(D)$ is a non-linear function of $P(D)$ and may not have a closed form, in general.

On the other hand, the closest Gaussian approximation of $P_{\text{ng}}(x, t)$, $P_{\text{ng}}^G(x, t)$, is given by

$$P_{\text{ng}}^G(x, t) = \frac{1}{\sqrt{2\pi(Dt)}} e^{-\frac{x^2}{2(Dt)}}.$$
leading to
\[ S_{ng}^D(t) = -\int_C dx P_{ng}^D(x,t) \ln[P_{ng}^D(x,t)] \]
\[ = \frac{1}{2} (1 + \ln(2\pi Dt)) \]
\[ \geq \frac{1}{2} (1 + \ln(2\pi) + \langle \ln(Dt) \rangle) \]
\[ = \frac{1}{2} (1 + \ln(2\pi) + \langle \ln(Dt) \rangle) + F_2(D), \tag{7} \]
where \( F_2(D) = \frac{1}{2} \ln(Dt) - \frac{1}{2} \langle \ln(Dt) \rangle \geq 0 \) and depends only on the form of \( P(D) \). Putting the results from eq. (5) and eq. (7) in eq. (3) we get
\[ \Delta S_{ng} = F_2(D) + F_1(D) + 1 - \langle D \rangle \left( \frac{1}{D} \right) \]
\[ = F_1(D) + F_2(D) - \langle D \rangle \left( \frac{1}{D} \right) - 1 \geq 0, \tag{8} \]
as by Jensen's inequality [47] \( \frac{1}{2} \geq \frac{\langle \ln(Dt) \rangle}{\ln(Dt)} \) for any bona fide distribution \( P(D) \). The positivity of \( \Delta S_{ng} \) is ensured by the positivity of KL divergence.

The undetermined constants, \( F_1(D) \) and \( F_2(D) \), are strictly positive definite quantities and appear in order to obtain equality, making use of the concavity of the logarithm. The exact analytical realisation of \( F_1 \) and \( F_2 \) is difficult due to its non-linear dependence on the functional form of the diffusion spectrum that is specific to the system of interest\(^3\).

On the other hand, for such a form of \( P_{ng}(x,t) \) (eq. (4)), the conventional quantifier of non-Gaussianity (eq. (1)) could be written as
\[ \alpha_2 = \frac{\langle D^2 \rangle - \langle D \rangle^2}{\langle D \rangle^2} = \frac{\Delta D^2}{\langle D \rangle^2}, \tag{9} \]
where \( \Delta D^2 = \langle D^2 \rangle - \langle D \rangle^2 \). One can check that for \( P(D) \sim \delta(D - D_{eff}) \), we get \( \alpha_2 = 0 \).

As an example, we now consider a typical class of heterogeneity in diffusion that is consistent with both the models of non-Gaussian yet Fickian random walk [23] and diffusing diffusivity [24] where the heterogeneity is represented by a diffusion spectrum \( P(D;\lambda) \). We introduce a parameter \( \lambda \) that controls the heterogeneity in the system. Thus, the form of displacement distribution becomes a superstatistics of \( D \) as it contains a diffusive propagator \( (G(x,t;D)) \) whose diffusion coefficient is sampled from the diffusion spectrum, \( P(D) \) following eq. (4) [48,49]. We limit our analysis to a form of \( P(D;\lambda) \) for which \( \frac{\partial}{\partial D} P(D;\lambda) \) remains a constant [23]:
\[ P(D;\lambda) = \lambda e^{-\lambda(D-1)} , \quad \text{for } D > 1. \tag{10} \]

Thus, the displacement distribution for such a random walker after a time \( t \) is given by \( P_{ng}(x,t;\lambda) = \int_0^\infty \frac{P(D;\lambda)}{\sqrt{4\pi Dt}} e^{-\frac{x^2}{4Dt}}. \) The closest Gaussian approximation to this distribution at time \( t \) is the one with the same variance as \( P_{ng}(x,t;\lambda) \), viz. \( 2(Dt) = 2(1 + \lambda^{-1}) \). We denote this distribution by \( P_{ng}^L(x,t;\lambda) \). For the \( P(D;\lambda) \) given in eq. (10), we can calculate a closed form for \( P_{ng}(x,t;\lambda) \):
\[ P_{ng}(x,t;\lambda) = \frac{\sqrt{\lambda}e^{\lambda x}}{4\sqrt{\pi}t} \left( e^{-\frac{|x|}{\sqrt{\lambda}}} \text{Erfc}\left( \sqrt{\lambda} - \frac{|x|}{\sqrt{4t}} \right) + e^{\frac{|x|}{\sqrt{\lambda}}} \text{Erfc}\left( \sqrt{\lambda} + \frac{|x|}{\sqrt{4t}} \right) \right), \tag{11} \]
where \( \text{Erfc}(x) \) is the complementary Error function. We use \( P_{ng}(x,t;\lambda) \) as in eq. (11) to obtain the non-Gaussian parameter:
\[ \alpha_2 = \frac{\lambda^{-2}}{\lambda^{-2} + 2\lambda^{-1} + 1}. \tag{12} \]

Using the form of \( P(x,t;\lambda) \) derived above in eq. (11), we compute \( \Delta S_{ng} \) by numerical integration with the suitable construction of instantaneous \( P_{ng}^L(x,t,\lambda) \) that has the same first two moments equal to the one of \( P_{ng}(x,t;\lambda) \), following eq. (3).

We now compare the conventional measure of non-Gaussianity with our measure based on the KL divergence, see fig. 1(a). The two measures, \( \alpha_2 \) and \( \Delta S_{ng} \), show a non-linear correlation. We see that although \( \alpha_2 \) saturates for high values of \( \lambda^{-1} = \langle D \rangle - 1 \), \( \Delta S_{ng} \) does not. We thus see that \( \Delta S_{ng} \) represents more information about the non-Gaussianity than \( \alpha_2 \), and can distinguish between cases where \( \alpha_2 \) might not differ appreciably.

The diffusions considered so far can all be expressed as superpositions of Gaussian diffusions with a distribution of diffusion constants, \( P(D) \). For this case, it is easy to see that the measure of non-Gaussianity \( \Delta S_{ng} \) does not change with time. This is because the time-dependent parts of both \( S_{ng} \) and \( S_{ng}^L \) are equal and equal to \( \frac{1}{2} \ln(t) \), where \( d \) is the number of dimensions.

**Non-Gaussian walker in supercooled liquid:** In order to validate this non-linear dependence of the two quantifications of non-Gaussianity, we now switch to a more realistic model of heterogeneous system where we look at the transient development of heterogeneity and how its disappearance. The model was proposed by Langer and Mukhopadhyay [50] in the context of the diffusion of tracer particles in a supercooled liquid, with distinct
domains which can be glassy or non-glassy. When in a glassy domain, the particle does not move at all, and only moves once the walls of the domain, which diffuse at a rate $1$, cross the position of the walker, thus leaving it in a non-glassy (or mobile) domain. The waiting time of the walker, assuming a distribution of glassy domain sizes $W(\rho) \sim \rho^2 \exp(-\rho^2)$, comes out to be $\psi_G(t) = 2e^{-2\sqrt{t}}$. In a mobile region, the particle is free to diffuse, and it diffuses with a diffusion constant $\Delta$, which is assumed to be larger than $1$. It now diffuses until it crosses a domain wall into a glassy domain. The distribution of waiting times in a mobile domain is $\psi_M(t) = \Delta e^{-\Delta t}$.

If the walker begins in an unfrozen domain, and assuming spherical symmetry (thus integrating over the angles), one can use the theory of continuous-time random walks to find that [50]

$$P_{ng}(k, u) = \frac{1}{u} \frac{1 - \psi_G(u) + u/\Delta}{1 - \psi_G(u) + k^2/2 + u/\Delta}.$$  \hspace{1cm} (13)

which is the Fourier and Laplace transformed form of the three-dimensional spherically symmetric probability distribution $P_{ng}(r, t)$ of the position of the particle at time $t$, and $\psi_G(u)$ is the Laplace transform of the waiting time distribution in glassy domains. The above equation can be numerically inverted in Fourier and Laplace space to find $P_{ng}(r, t)$.

It was shown in [50] that the distribution for very short ($\ll \Delta$) and very long ($\gg \Delta$) times is Gaussian, but with different variances, $\Delta t$ and $2t$, respectively. The crossover period in between is where the non-Gaussianity, measured in [50] by the quantity $\alpha_2 = \frac{3\langle r^4 \rangle}{5\langle r^2 \rangle^2} - 1$, reaches a peak at times of the order of $\Delta^{-1}$. Figure 2 shows that our measure of non-Gaussianity, $\Delta S_{ng}$, captures these basic properties as well.

Figure 1(b) shows a plot of $\alpha_2$ vs. $\Delta S_{ng}$ for $\Delta = 3$. We see significant differences in the dynamic behavior between $\alpha_2$ and $\Delta S_{ng}$. Time moves counter-clockwise along the loop. For low and high times (the lower left-hand corner, near the origin), the behavior is mostly identical when the dynamics is essentially a Gaussian, and both $\alpha_2$ and $\Delta S_{ng}$ are small. In the intermittent regime, however, heterogeneity is captured differently in $\alpha_2$ and $\Delta S_{ng}$ as is clearly seen in fig. 1(b).

We now show the time-dependent behavior of $\Delta S_{ng}$ for the same system in fig. 2 for $\Delta = 3.0$ that corresponds to the case in fig. 1(b). We see $\Delta S_{ng}$ has small values at $t \to 0$ and when is $t$ sufficiently larger than the timescale associated with the heterogeneity. $\Delta S_{ng}$ has a peak near $t \approx \Delta^{-1}$. The behavior is qualitatively similar to the behavior of $\alpha_2$ reported in glassy systems [39,50–54]. However, as seen from fig. 1(b), the two quantities show maximum deviation near the peak. This is because for intermediate times, the heterogeneous nature of the medium strongly affects the displacement distribution $P_{ng}$.

Fig. 1: Comparing the information theoretic quantification ($\Delta S$) with the conventional measure ($\alpha_2$) for the case of (a) Fickian yet non-Gaussian random walk, whose dynamics is governed by eq. (10) and (b) a walker in a super-cooled liquid following ref. [50].

Fig. 2: (a) Time-dependent behavior of $\Delta S_{ng}$ for a walker in a super-cooled liquid following ref. [50].
At \( t \gg \tau_S \), \( \Delta S_{\text{ng}} = D_{KL}(P_{\text{ng}} || P_{\text{G}}) \approx D_{KL}(P_{\text{ng}} || P_{\text{eff}}) \sim \ln(G(x,t;D_L)) \rightarrow 0 \) as \( P_{\text{ng}}(x,t;D_L) \rightarrow P_{\text{G}}(x,t;D_L) = G(x,t;D_L) \), where \( D_L \) is the long time diffusion coefficient. This happens at \( t \rightarrow 0 \) and \( t \gg \tau_S \) when \( P_{\text{ng}} \) has a shape close to Gaussian.

The potential of information theory based frameworks in understanding thermodynamic and physical transitions has been demonstrated recently [55–58]. As we have shown, the relative entropy based construction not only captures the essential factors of the non-Gaussianity, but also provides higher order information coming from higher order moments. We have shown that this information is most relevant when the heterogeneity in the system is large. For the specific model, [50] this occurs at \( t \approx \Delta^{-1} \).

Our analysis adds a new, sophisticated tool for gaining insights into heterogeneity via the role of non-Gaussianity in physical systems. The information extracted via the newly proposed relative entropy based quantification shows that the non-Gaussianity inferred by moment ratios like \( \alpha_2 \) is not adequate in handling heterogeneous situations, as we have shown with the study of two generic models of soft-matter systems, namely a non-Gaussian walker with fluctuating diffusivities, and a walker in a super-cooled liquid. The relative entropy integrates information from the whole distribution of walker displacements and thus provides a more complete picture than looking at the first few moments. Our framework also simplifies the extraction of non-trivial information from non-Gaussian distributions, even from experimental set up, bypassing the difficulties in estimating the diffusion spectrum [49]. Using confocal microscopy, it is possible to obtain the displacement distributions in real space experiments [42]. From the moments of such displacement distribution, construction of the closest Gaussian estimate is possible following our framework. The quantification of non-Gaussianity simply comes from the entropic distance between the given distribution and the constructed Gaussian counterpart. Moreover, our study also has potential to explore microscopic heterogeneity in terms of entropy or disorder.

In brief, we extract physical information in dynamic heterogeneity when molecular motion undergoes heterogeneity in diffusion. The heterogeneity appears as a competition between intrinsic disorder due to diffusion and order arising out of spatial localization of molecular mobilities. Such interplay has been realized as a non-monotonic non-Gaussianity in the particle displacement distributions in a host of systems. In this work, we gain insights into these complex transport processes via an information-theoretic description. In recent times, various other information theoretic notions have also been explored in understanding complex physical systems [13,56,59–61]. Here, in contrast, we provide a simple mathematical formalism that directly encodes the dynamic heterogeneity in terms of non-Gaussian information. It would be interesting to see how this new development finds its relevance in more realistic complex situations where microscopic dynamics experience heterogeneity due to external field [62], activity [63], local heating [64], background potential [65] or spatially varying potential [66]. The application of this inter-disciplinary tool is not limited to soft matter, and we believe that it will continue to surprise scientists from different domains where microscopic heterogeneity governs the complex transport processes and molecular metastability subjected to complex energy landscape is of key interest.

We acknowledge discussions with P. CHAUDHURI, S. GHOSH, C. DASGUPTA and M. S. SHELL. We also thank V. VAIBHAV for sharing data. This research was supported in part by the International Centre for Theoretical Sciences (ICTS) during a visit for participating in the program - Entropy, Information and Order in Soft Matter (Code: ICTS/eosm2018/08).

**Appendix: evaluation of \( F_1(D) \) and \( F_2(D) \) for a typical case.** – Let us consider the underlying distribution of the local diffusive processes is a double hump (delta) function, i.e., \( P(D) = \frac{1}{2}(\delta(D - D_l) + \delta(D - D_u)) \), where \( D_u, D_l > 0 \) and \( D_l > D_u \). This leads to the non-Gaussian distribution function \( P_{\text{ng}}(x,t) = \int dD \ G(x,t : D) \ P(D) = \frac{1}{2}(G(x,t;D_l)+G(x,t;D_u)) \). A straightforward calculation yields

\[
S_{\text{ng}}(x,t) = - \int dx P_{\text{ng}}(x,t) \ln(P_{\text{ng}}(x,t))
\]

\[
= \frac{1}{2} \ln(2) - \frac{1}{2} \int dx G(x,t;D_l) \ln(G(x,t;D_l)) - \frac{1}{2} \int dx G(x,t;D_u) \ln(G(x,t;D_u)) - \frac{1}{2} \int dx G(x,t;D_l) \ln \left( 1 + \frac{G(x,t;D_u)}{G(x,t;D_l)} \right) - \frac{1}{2} \int dx G(x,t;D_u) \ln \left( 1 + \frac{G(x,t;D_l)}{G(x,t;D_u)} \right) = \frac{1}{2} \ln(2) + \frac{1}{2} \left( 1 + \ln(2\pi) + \ln(D_l t) + \ln(D_u t) \right) - \frac{1}{2} \left( 2 + \sum_{m=2}^{\infty} \frac{(-1)^{m-1}}{m} \int dx G^{m}(x,t;D_u) G^{m-1}(x,t;D_l) \right) + \sum_{n=2}^{\infty} \frac{(-1)^{n-1}}{n} \int dx G^{m}(x,t;D_l) G^{m-1}(x,t;D_u) = \frac{(D_l + D_u)^2}{4D_l D_u} + \frac{1}{2} \left( -1 + \ln(2\pi) + \ln(D_l t) + \ln(D_u t) \right)
\]
\[
-\frac{1}{2} \left( \sum_{m=2}^{\infty} \frac{(-1)^{m-1}}{m} \int dx \frac{G^m(x; t; D_u)}{G^{m-1}(x; t; D_l)} \right.
\]
\[
+ \sum_{n=2}^{\infty} \frac{(-1)^{n-1}}{n} \int dx \frac{G^n(x; t; D_l)}{G^{n-1}(x; t; D_u)} \right)
\]
\[
- \ln(2) - \frac{(D_l + D_u)^2}{4D_l D_u} \right)
\]
\[
= \langle D \rangle \left( \frac{1}{D} \right) + \frac{1}{2} \left( 1 + \ln(2\pi) + \langle \ln(D(t)) \rangle \right) - F_1(D).
\]
\[
\text{(A.1)}
\]

Similarly, with this distribution of diffusive processes, the closest Gaussian of the non-Gaussian distribution
\[
P_{\text{ng}}^\delta(x, t) = \frac{1}{2\pi(D) t} e^{-\frac{x^2}{2(D) t}}
\]
leading to \( \langle D \rangle = (D_l + D_u)/2 \). A straightforward calculation further yields
\[
S_{\text{ng}}(t) = -\int dx \ P_{\text{ng}}^\delta(x, t) \ln(P_{\text{ng}}^\delta(x, t))
\]
\[
= \left\{ \frac{1}{2} \left( 1 + \ln(2\pi) + \ln\left(\frac{(D_l + D_u) t}{2}\right) \right) \right\}
\]
\[
= \left\{ \frac{1}{2} \left( 1 + \ln(2\pi) \right) \right\}
\]
\[
+ \left\{ \frac{1}{2} \ln\left((D_l) t \left(1 + \frac{D_u}{D_l}\right)\right) \right\}
\]
\[
+ \left\{ \frac{1}{2} \ln\left((D_u) t \left(1 + \frac{D_l}{D_u}\right)\right) \right\}
\]
\[
= \left\{ \frac{1}{2} \left( 1 + \ln(2\pi) \right) \right\}
\]
\[
- \frac{1}{2} \left\{ \ln\left((D_l) t \left(1 + \frac{D_u}{D_l}\right)\right) \right\}
\]
\[
- \frac{1}{2} \left\{ \ln\left((D_u) t \left(1 + \frac{D_l}{D_u}\right)\right) \right\}
\]
\[
+ \left\{ \frac{1}{4} \sum_{m=1}^{\infty} \frac{(-1)^{m-1} D_l^m}{m D_l^{m-1}} \right\}
\]
\[
+ \left\{ \frac{1}{4} \sum_{n=1}^{\infty} \frac{(-1)^{n-1} D_u^n}{n D_u^{n-1}} \right\}
\]
\[
= \left\{ \frac{1}{2} \left( 1 + \ln(2\pi) \right) \right\} + \langle \ln(D(t)) \rangle + F_2(D).
\]
\[
\text{(A.3)}
\]

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