Skewness of steady-state current fluctuations in nonequilibrium systems

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A skewness of the probability for instantaneous current fluctuations, in a nonequilibrium steady state, is observed experimentally in a dusty plasma for the first time. This skewness is attributed to the spatial asymmetry, which is imminent to the nonequilibrium systems due to the external hydrodynamic gradient. Using the modern framework of the Large Deviation theory, we extend the Onsager-Machlup ansatz for equilibrium fluctuations to systems with a preferred spatial direction, and provide a novel Modulated Gaussian probability distribution, which is tested by simulations. This new probability distribution is also of potential interest for other statistical disciplines. Connections with the principles of Statistical Mechanics, due to Boltzmann and Gibbs, are discussed as well.

Keywords: large deviation; probability distribution; asymmetry; current; nonequilibrium; fluctuation

I. INTRODUCTION

Recently a non-Gaussian structure of fluctuations was reported for the electric charge density and currents in quantum and nanoscale systems [1–3]. In particular, the skew asymmetry of the fluctuations probability distribution was found not only for the nonequilibrium current [3], but also for the charge density of an equilibrium system in the presence of an external magnetic field. This discovery contradicts the original theory of fluctuations due to Onsager and Machlup [4, 5], who predicted a Gaussian probability distribution of the equilibrium fluctuations.

In this paper we provide the first evidence of the skew fluctuations for a manifestly macroscopic system in a Non-Equilibrium Steady State (NESS), using laboratory experiments. We study a hydrodynamic shear flow of a 2-Dimensional (2D) dusty plasma, a complex noble gas plasma with massive charged dust particles [6–9]. The fluctuating quantity will be the instantaneous value of a current, which persists in the NESS system due to an externally applied constant gradient. In our study, the current is the xy-component of the pressure tensor P_{xy} or, in other words, the viscous flux of the x-component of the linear momentum in the y-direction. The momen-

tum flow is maintained by the shear rate, a transverse gradient of the streaming velocity.

To illustrate the skewness of a distribution, we sketch in Fig. 1 three probability densities as functions of a variable J: (a) is symmetric about zero; (b) has the same shape as (a), but is displaced from the origin without skewness; (c) is both shifted from the origin and skewed. We will find that the probability distributions of instantaneous current fluctuations in a NESS are not only shifted, with respect to the equilibrium case, but also skew, as sketched in Fig. 1(c).

The skewness of a distribution is characterized by the third probability moment. For a sample of M measurements, the skewness is calculated from

$$\frac{1}{M\sigma^3} \sum_{i=1}^{M} (J_i - \bar{J})^3, \tag{1}$$

where J_i, $\bar{J}$ and σ are, respectively, i-th value of the measurements, the sample mean and its standard deviation.

In the presence of a hydrodynamic gradient, there is *a preferred spatial direction* of the flow, which reduces the space symmetry of the system with respect to that of its equilibrium state. This leads to a bias of the current fluctuations, so that their probability distribution is skew in the direction of the flux. Hence the fluctuations, which enhance the flow, are favored over the opposite ones. While being subtle, this bias remained unnoticed by earlier theories and experiments, which were mainly concentrated on the near equilibrium regimes. Nonetheless, it becomes quite evident in far from equilibrium sys-

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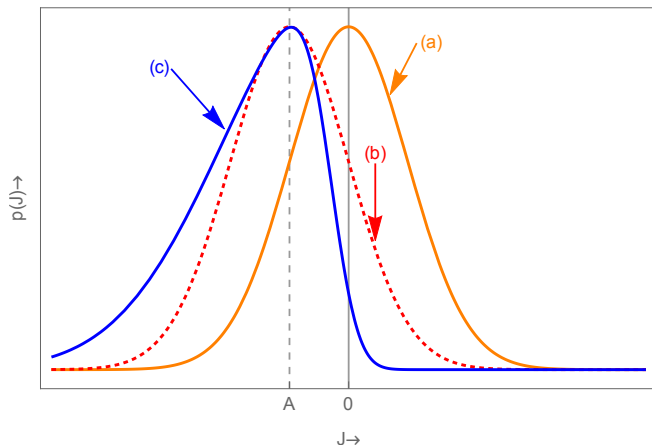[‡] chun-shang-wong@uiowa.edu
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FIG. 1. Illustration of probability distributions p, as a function of J, with and without skewness: (a) a Gaussian distribution symmetric about the origin with $p(J) = p(-J)$; (b) a Gaussian distribution, which is symmetric about $J = A$ with $p(A + J) = p(A - J)$ and satisfies the Gallavotti-Cohen relation Eq. (2); (c) an asymmetric Skew-normal distribution [10], which is skew to the left. For the curve c) there is no constant B, such that $p(B + J) = p(B - J)$ for any J. Its negative skewness implies that $p(A - \Delta A) > p(A + \Delta A)$ for any $\Delta A > 0$.

tems, where both the strength of the macroscopic current and the skewness fluctuations increase substantially.

As already mentioned, a similar observation was made not only for a NESS current. In [3] fluctuations of the charge distribution was found skew for an equilibrium system, subject to an external magnetic field. There it was argued that the presence of the magnetic field reduces the symmetry of the system and alters statistical properties of the microscopic stochastic noise. Consequently, the Gaussian noise, considered by Onsager and Machlup in their dynamical theory of fluctuations, may not be adequate to describe systems with a preferred spatial direction.

To make a progress, we disregard the issues of fluctuations dynamics, which still needs a substantial revision. Using a suitable extension of the Onsager-Machlup original ansatz, we developed a novel Modulated Gaussian (MG) distribution to account for the skewness of fluctuations. This new MG distribution is a natural model of the time-independent probability density for fluctuations in systems with a preferred spatial direction. A special case of the MG is the normal distribution, used in [4, 5], thus the earlier theory of Onsager and Machlup is consistent with ours.

We expect that the MG will describe the fluctuations of instantaneous currents in a variety of physical systems. While our theory concludes with the prescription that the Modulated Gaussian (MG) describes the instantaneous fluctuations, its parameters have only a statistical meaning. Their physical interpretation might not be possible, until the original stochastic dynamics of Onsager

and Machlup is properly generalized for asymmetric systems.

Note that the *temporal* asymmetry, predicted for the trajectories of the fluctuations in the NESS by the Macroscopic Fluctuation Theory (MFT) [11, 12], differs from the bias of the *time-independent* probabilities of the NESS fluctuations, studied here. The MFT uses the time irreversibility of the NESS, which generalizes the time reversibility of an equilibrium system in the Onsager-Machlup theory of fluctuations. This approach, among other things, predicts various properties for the evolution of the NESS current fluctuations. Nonetheless, it alone might not be able to provide a model for their probability distribution, analogous to the Gaussian model in the Onsager-Machlup theory. The MG we proposed fills in this gap between the present theories of equilibrium and the NESS fluctuations of currents.

The topic of this paper is also different from fluctuation relations, such as the Galavotti-Cohen fluctuation theorem [13–18]. These relations deal with a distribution $p(J)$, which is asymmetric in a way that

$$p(J)/p(-J) = \exp\{-\kappa A J\}, \qquad (2)$$

where κ and A are constants.

For example, this relation is satisfied by a Gaussian (b) centered at A in Fig. 1 (e.g. [19]). In other words, the Gallavotti-Cohen fluctuation theorem Eq. (2) can be satisfied by a distribution with zero skewness [20, 21]. The class of such functions $p(J)$ is actually not limited to the Gaussian family.

The fluctuation relations do not apply to *instantaneous* currents, since they describe distributions that have been averaged over a significant time interval. However, one can use the MG expression to model the probability density of these *time-averaged* currents. In [22, 23], their distribution was characterized by the cumulants. The time-averaged currents should, in principle, have the same kind of skew bias, which implies a non-vanishing third cumulant. This might lead to a connection between the MG parameters and the results of MFT. Nonetheless, this idea is not pursued in the present study.

Our laboratory evidence of the skew asymmetry is supported by our Molecular Dynamics (MD) simulations. These computations were carried out for a 2D system, which imitates our experiments, with particles interacting through the Debye-Hückel (DH) potential. To demonstrate a broader extent of our theory, also 3D simulations of a fluid were conducted, using the Weeks-Chandler-Andersen potential (WCA, [24]) [25].

In particular, we consider further a system with N particles of equal mass m. The current P_{xy}, caused by an externally applied shear rate γ, is[26]:

$$P_{xy} = L^{-D} \sum_{i=1}^{N} \left(\frac{p_{xi} p_{yi}}{m} + F_{xi} y_i \right). \qquad (3)$$

Here L^D is the area or volume of the system, depending on the number of its physical dimensions D; for the

i-th particle, y_i, p_{xi}, p_{yi}, F_{xi} are, respectively, the y coordinate, the x and y components of the *peculiar* [27] linear momentum, and the force acting in the x direction due to the interactions with all other particles.

Finally the MG distribution, derived in this manuscript, in principle, could also be relevant for other disciplines, which use the Large Deviation (LD) theory [28]. The LD theory studies deviations of an average over a large number of random variables, from its most likely value. Our formulation of the MG relies solely on this approach [28]. Thus we regard the current of interest as a randomly fluctuating quantity, which satisfies the principles of the LD theory. Therefore the MG probability distribution is not limited to its applications in physics.

II. NON-GAUSSIANITY OF THE CURRENT FLUCTUATIONS

The experimental results, discussed in this section, were previously published in [29]. There, we observed a shear flow, driven by laser manipulations in a monolayer of a dusty argon plasma. The motion of dust particles was confined to a plane and tracked using video cameras, which allowed us to study properties of this system at a level of detail, similar to that of MD simulations. Below we provide a further analysis of the data, acquired earlier in [29]. We computed long time series of P_{xy} for three separate regions of the flow with the shear rate $\gamma = 3.4\,s^{-1}$ and average particles number $\langle N \rangle = 58$. This allows to neglect the time correlations in the collected sample of measurements.

The histogram for P_{xy} in Fig. 2, obtained from our experiments after subtracting the sample average $\bar{P}_{xy}$ to emphasize its skew asymmetry, reveals a skewness, which we attribute to the presence of the applied shear. The bias of the probability density towards the negative values is small, which is possibly a reason that it remained unnoticed so far by experimenters.

To confirm the statistical significance of our experimental result we applied the statistical framework of hypothesis testing [30, 31] as well as the bootstrap techniques [32]. These showed, that the probability of error in the assessment of skewness was less than 1%.

The MG distribution, which will be derived in Sec. III, accounts for the *skewness* (skw$\{P_{xy}\}$) and the *excess kurtosis* (krt$\{P_{xy}\}$) [33] of the sample, with an accuracy up to the 5th significant digit. It fits the experimental data better than the Gaussian, with its error δ_{err}^{MG} being three times smaller than δ_{err}^{G} for the Gaussian, as reported in Fig. 2 [34].

Our MD simulations show that the asymmetry of $p(P_{xy})$ increases with the magnitude of the shear rate. In Fig. 3 the observed skewness, indicated by the cross symbols, becomes progressively negative with increasing γ [35]. Indeed, comparing panels a) and b) of Fig. 4, one observes that the Gaussian model is quite inaccurate for the larger shear rate of $\gamma = 7.681\,s^{-1}$ due to the notable
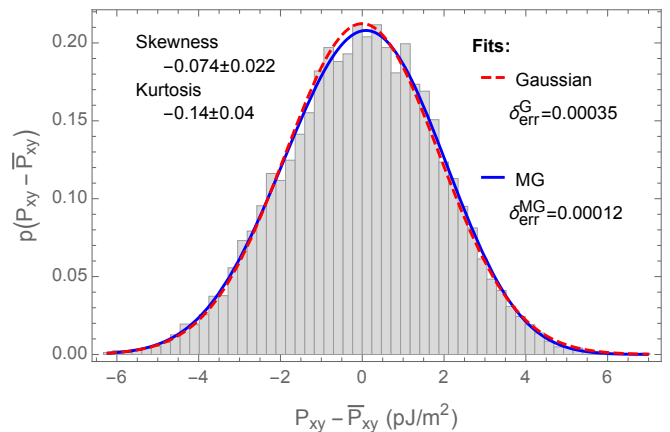

FIG. 2. Histogram for $p(P_{xy} - \bar{P}_{xy})$ from our experimental measurements. The skewness of the distribution can be detected by looking at the peak of the Gaussian fit at the origin, which is shifted slightly to the left from the maximum of the probability density. The inequality $\bar{P}_{xy} < \tilde{P}_{xy}$ agrees with the observed negative skewness.

skew asymmetry of the underlying probability distribution. To neglect the effects of time correlations, we made long pauses between consecutive measurements of P_{xy}.
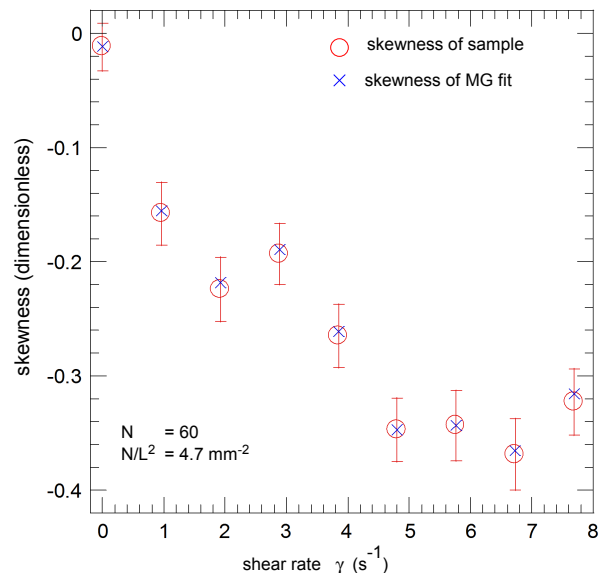

FIG. 3. Skewness of a time series $P_{xy}(t)$ as a function of γ for our 2D DH plasma simulations in SI units. The circles refer to the skewness computed from Eq. (1), with the error bar estimated by the bootstrap [32]. The crosses stand for the skewness of the MG fit.

The MG model agrees very well with the histogram data of our simulations in Fig. 4. The fitting error δ_{err} is reduced more than three-fold at a low shear rate of $0.961\,s^{-1}$ by using the MG instead of Gaussian model, and it is reduced twelve-fold at a higher shear rate of
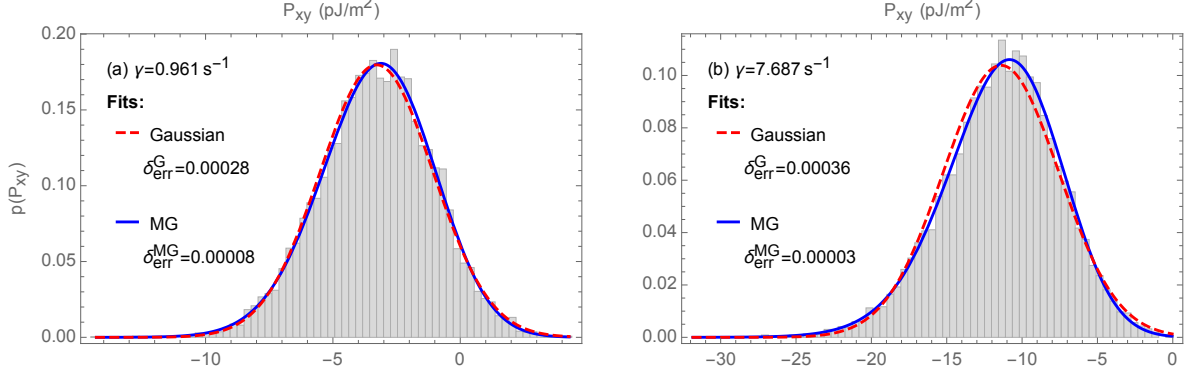
FIG. 4. Histograms for the probability density $p(P_{xy})$ from our 2D DH plasma simulations in SI units ($N = 60$ and $N/L^D = 4.7\,mm^{-2}$): (a) a low shear rate, (b) a high shear rate. The fitting error of the MG $\delta_{\mathrm{err}}^{\mathrm{MG}}$ is of the same order at the low and high shear rates, while the fitting error of the Gaussian $\delta_{\mathrm{err}}^{\mathrm{G}}$ notably grows for the larger γ, which indicates the increase of non-Gaussianity with the shear rate.

$7.587\,s^{-1}$. While, of course, the Gaussian cannot account for the skewness of the distribution, the MG does it quite well, as follows from the close agreement between the skewness observed in P_{xy} data and the skewness of their MG fit in Fig. 3.

The skew asymmetry of $p(P_{xy})$ is a characteristic property of the current fluctuations in a NESS. The skewness of their probability distribution has the same sign as the average $\bar{P}_{xy}$ and the most likely value $\tilde{P}_{xy}$, as can be seen in Fig. 4. When approaching equilibrium conditions, the bias of $p(P_{xy})$ gradually disappears. It vanishes completely only at the equilibrium point, $\gamma = 0$.

The skew asymmetry of the fluctuations appears in the third order probability moment and, consequently, is more subtle than the phenomenon of the fluctuations themselves, which are of the second order. This implies, that when the size of the system approaches a macroscopic limit, while the variance of the current probability distribution decreases, the skewness decays even faster. However the asymmetry of the distribution still persists.

III. MODULATED GAUSSIAN DISTRIBUTION

To account for the deviations from the Gaussian structure of fluctuations, which was suggested in the theory of Onsager and Machlup, we repeat their initial ansatz, Eq. (2-11) in [4]. Namely, we are looking for a probability density function of a fluctuating variable x in the form:

$$p(x) = \exp\{\frac{S(x)}{k_B}\}, \qquad (4)$$

where k_B is Boltzmann's constant and S is some function of x.

The theoretical justification of the treatment that follows below is provided in the context of LD theory and can be found in the Appendix with other details. Here we only mention that Onsager and Machlup interpreted

$S(x)$ as the entropy of the macroscopic state x. Since we are going to extend their idea to NESS, S can not be connected with the equilibrium entropy. In the modern framework of the LD theory [28], $S(x)$ is related to the *LD function*.

As in [4] we proceed expanding the function $S(x)$ from Eq. (4) in a power series about the most likely (macroscopic) value $\tilde{x}$, which is the global maximum of $S(x)$, up to a prescribed order n. Denoting $S_i = d^i S(x)/dx^i|_{x=\tilde{x}}$, we obtain:

$$\frac{S(x)}{k_B} \simeq \frac{S(\tilde{x})}{k_B} + \sum_{i=1}^{n} \frac{S_i}{i!k_B}(x - \tilde{x})^i$$
$$\stackrel{\mathrm{def}}{=} -\frac{\tilde{S}}{k_B} + \frac{\Delta S(\Delta x)}{k_B}, \text{ respectively,} \qquad (5)$$

which defines a constant $\tilde{S}$ and a fluctuation cost function $\Delta S(\Delta x)$ of the deviation $\Delta x = x - \tilde{x}$.

Since $S(x)$ is expanded in Eq. (5) about its global maximum $\tilde{x}$, we have $S_1 = 0$. Due to the symmetry, which does not favor any direction of fluctuations in equilibrium systems considered by Onsager and Machlup, it follows that $\Delta S(x - \tilde{x}) = \Delta S(-x + \tilde{x})$. Hence, using a trivial substitution $y = x - \tilde{x}$, one can easily find that in such systems $S_3 = 0$.

Summarizing the above arguments, the approximation order $n = 2$ in Eq. (5), as chosen by Onsager and Machlup [4], is actually accurate up to the 4th order. Absence of the third order term S_3, as it occurs in a system without a preferred spatial direction, leads to an approximate Gaussian structure of fluctuations.

Naturally, to account for the skewness we have to use a higher order of approximation, retaining only the prop-

erty $S_1 = 0$:

$$\Delta S(\Delta x) = \frac{S_2}{2}\Delta x^2 + \sum_{i=3}^{n} \frac{S_i}{i!}\Delta x^i$$

$$= \frac{S_2}{2}\Delta x^2 \left\{ 1 + 2\sum_{i=3}^{n} \frac{S_i}{i!S_2}\Delta x^{i-2} \right\}$$

$$= \frac{S_2}{2}\Delta x^2 \Sigma_n, \qquad (6)$$

where we call the expression between the curly braces a modulating factor Σ_n, to which the MG distribution owes its name.

Then Eq. (4), together with Eqs. (5 and 6), give:

$$p(x) = \exp\{-\frac{\tilde{S}}{k_B} + \frac{\Delta S(\Delta x)}{k_B}\}$$

$$= \exp\{-\frac{\tilde{S}}{k_B} + \frac{S_2\Sigma_n}{2k_B}\Delta x^2\}, \qquad (7)$$

where the normalization of the total probability requires that

$$\exp\{\frac{\tilde{S}}{k_B}\} \simeq \int_{-\infty}^{\infty} dx \exp\{\frac{\Delta S(x - \tilde{x})}{k_B}\}.$$

For $p(x)$ to be integrable in Eq. (7), n has to be restricted to even integers. When $n = 2$, $i.e.$ $\Sigma_n \equiv 1$, Eq. (7) turns into a Gaussian distribution. Taking the next admissible order of approximation $n = 4$, we replace the three parameters S_i ($i = 2, 3, 4$) in Σ_4 by another set of three parameters: scale $\Pi > 0$, asymmetry A and non-Gaussianity $B \geq 0$, respectively. These parameters acquire a clear statistical interpretation, when used in Eq. (7):

$$p(x) \propto \exp\{\frac{\Delta S(x)}{k_B}\} = \exp\left\{ -\frac{\Delta x^2}{2\Pi^2}\left[1 - 2\sqrt{2/3}AB\frac{\Delta x}{\Pi} + B^2\frac{\Delta x^2}{\Pi^2}\right] \right\}. \qquad (8)$$

Equation (8) is the Modulated Gaussian distribution. The dimensionless constant B controls the level of non-Gaussianity [36]. Furthermore, A causes the asymmetry of $p(P_{xy})$, which is skew to the left (right), when $A < 0$ ($A > 0$), respectively. The coefficient $2\sqrt{2/3}$ of the term with A in Eq. (8) was chosen to make $\Delta S(x)$ a non-concave function of x for $-1 \leq A \leq 1$ [37]. Violation of the non-concavity condition would admit more sophisticated shapes of the probability density because of additional critical points [38]. Unless there is a physical argument for these special points, when fitting a statistical sample, numerical artifacts may emerge due to the approximate nature of Eq. (8). To avoid this, the desired MG expression should be restricted to non-concave solutions.

The superior accuracy of MG over the Gaussian approximation was already demonstrated in Sec. II. The above theoretical arguments, though, miss a proper generalization of the stochastic dynamics suggested by Onsager and Machlup, which would lead to the skew time-independent probability distribution of fluctuations.

IV. CONCLUSION

Our experiments and numerical simulations demonstrate the skewness of the probability distribution for instantaneous fluctuations of the viscous shear current in a NESS. We attribute this property to the asymmetry of

our system in the presence of a *preferred spatial direction of the flow*. By the same argument, the skewness of instantaneous current fluctuations, as well as that of their time averages, should also be expected in similar situations, *e.g.* for a heat flux in the presence of a constant temperature gradient.

The skewness of NESS current fluctuations notably increases for large externally applied forces, as observed in our study and similar experiments [1–3]. Therefore a non-Gaussian probability distribution is especially important to describe fluctuations far from equilibrium or equilibrium systems subject to a large external potential fields.

The new MG probability distribution accurately describes the probability of fluctuations and, in particular, their skew asymmetry. This model is justified here using the LD theory. It extends the Onsager-Machlup original idea, by considering higher order terms in a power series expansion of the LD function. We verified its accuracy experimentally and numerically.

The skew asymmetry of the fluctuations probabilities poses a new constraint on the LD function of the current fluctuations. Since this function must manifest this asymmetry, it should consist not only of a quadratic form, which leads to the normal distribution, but involve some asymmetric contributions. This is consistent with particular results found for some simple mathematical models of the NESS [39, 40] and prior experimental observations in quantum and nano scale systems [1–3].

In Appendix the probability of current fluctuations is also characterized by their entropy cost in the Boltzmann approach to Statistical Physics [41]. The entropy cost was identified as the decrease of the LD function produced by a fluctuation from the most likely state of the system. This approach may be further related with that of a fluctuation free energy *cf* [31].

A possible direction for future developments is the *dynamical theory* of current fluctuations [21]. A suitable correction of the Langevin equation, suggested by Onsager and Machlup for equilibrium fluctuations [4, 5], is a viable approach to deal with this problem. There are various modifications of the Langevin equation, which produce a skew time-independent probability distributions, *e.g.* a non-Gaussian noise. Thus a proper dynamical representation, which would account for the preferred spatial direction and the time irreversibility, addressed in the MFT approach, remains yet to be found.

Appendix A: The Modulated Gaussian and the Large Deviation theory

In this Appendix we provide the theoretical details of Eq. (4) for $x = P_{xy}$. Considering each term of the summation operator in Eq. (3) as a random variable, we assume that the spatial average P_{xy} satisfies the LD principle. From this assumption it follows (*cf.* [28]) that:

1. There exists a non-negative rate function, which is also called the LD function:

$$I(P_{xy}) \overset{\text{def}}{=} - \lim_{N \to \infty} [S(P_{xy})/(Nk_B)], \qquad (A1)$$

for the exponential decay of the probability density

$$p(P_{xy}) \sim \exp[-NI(P_{xy})]. \qquad (A2)$$

2. This rate function has a global minimum at the most likely value of $P_{xy} = \tilde{P}_{xy}$, which satisfies:

$$I(\tilde{P}_{xy}) = 0. \qquad (A3)$$

Since by definition $\Delta S(\Delta P_{xy})$ in Eq. (5) is zero for the most likely value $\tilde{P}_{xy}$, Eqs. (A1, A3 and 5) suggest:

$$I(\tilde{P}_{xy}) = \lim_{N \to \infty} \frac{\tilde{S}}{Nk_B} = 0$$
$$I(P_{xy}) = - \lim_{N \to \infty} \frac{\Delta S(\Delta P_{xy})}{Nk_B}. \qquad (A4)$$

Moreover, invoking Eq. (A2) of the LD principle, for a finite N we pose that $S_i \approx -Nd^i I(P_{xy})/dP_{xy}^i|_{P_{xy} = \tilde{P}_{xy}}$. Then it follows, that the first derivative S_1 vanishes and that the second derivative S_2 is positive [42], because $I(\tilde{P}_{xy})$ is the global minimum of the rate function by

definition. This allows us to express $\Delta S(\Delta P_{xy})$ as done in Eq. (6).

These developments can be readily connected with Statistical Mechanics. According to the Boltzmann principle, given a measure of system's microstates $w(P_{xy})$, for a given value of P_{xy}, and the total number of accessible microstates $W = \int_{-\infty}^{\infty} w(P_{xy})dP_{xy}$ under the specified macroscopic constraints of temperature, shear rate *etc.*, the time-independent probability density of a steady state $p(P_{xy})$ and the Boltzmann entropy $S_B(P_{xy})$ are given by:

$$p(P_{xy}) = \frac{w(P_{xy})}{W}$$
$$S_B(P_{xy}) = k_B \ln w(P_{xy}). \qquad (A5)$$

Due to the existing controversies on the entropy concept for nonequilibrium systems, we have to note that the Boltzmann entropy can be *always defined* for a steady state using its time-invariant probability density via Eq. (A5) [41]. In particular, this approach neither relies on, nor verifies thermodynamic relations for equilibrium systems or their widely criticized formal expansions. Equations (A5) may be regarded merely as the "frequentist" interpretation of the probability.

Using the notion of a total entropy $S_{\text{tot}} = k_B \ln W$ after [41], we deduce from Eq. (A5) that:

$$k_B \ln p(P_{xy}) = k_B \ln w(P_{xy}) - k_B \ln W$$
$$= S_B(P_{xy}) - S_B(\tilde{P}_{xy}) + S_B(\tilde{P}_{xy}) - S_{\text{tot}}$$
$$= \Delta S_B(P_{xy}) + S_B(\tilde{P}_{xy}) - S_{\text{tot}}, \quad (A6)$$

where in the second equality we added and subtracted $S_B(\tilde{P}_{xy})$, to introduce the Boltzmann entropy difference $\Delta S_B(P_{xy}) = S_B(P_{xy}) - S_B(\tilde{P}_{xy})$.

Comparing Eq. (7) with Eq. (A6), one sees that

$$-k_B \ln p(\tilde{P}_{xy}) = \tilde{S} = S_{\text{tot}} - S_B(\tilde{P}_{xy})$$
$$\Delta S(P_{xy} - \tilde{P}_{xy}) = \Delta S_B(P_{xy}), \qquad (A7)$$

because $\Delta S_B(P_{xy})$ and $\Delta S(P_{xy} - \tilde{P}_{xy})$ are both zero at $P_{xy} = \tilde{P}_{xy}$ by definition.

Equation (A7) provides the physical interpretation of $\Delta S(\Delta P_{xy})$ as well as of the constant $\tilde{S}$. The most likely value of $P_{xy} = \tilde{P}_{xy}$ maximizes the Boltzmann entropy (*cf.* Eq. (A5)). Consistently, $\Delta S(\Delta P_{xy}) \leq 0$ is the *entropy cost* of a fluctuation $P_{xy} = \tilde{P}_{xy} + \Delta P_{xy}$ [43]. The constant $\tilde{S}$ is the remaining total entropy, after subtracting the Boltzmann entropy of the most likely macrostate. It is opportune to note, that one may restate the derivation of MG distribution, using the principle of maximum Boltzmann entropy, instead of the global minimum of the LD function, as done above.

Finally, the Gibbs entropy, given by a functional $S_G[p]$, for the distribution Eq. (7) is

$$S_G[p(\cdot)] = -k_B \int_{-\infty}^{\infty} dP_{xy} p(P_{xy}) \ln p(P_{xy})$$
$$= -k_B \langle \ln p(P_{xy}) \rangle = \tilde{S} - \langle \Delta S(P_{xy}) \rangle, \quad (A8)$$

which provides $\tilde{S}$, up to a constant term $\langle \Delta S(P_{xy}) \rangle$.

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[25] See Supplementary Materials at [link to be inserted by publisher] for the technical details of our numerical models.

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[33] *I.e.* the third and fourth order moments of the probability distribution, respectively.

[34] See Supplementary Materials at [link to be inserted by publisher] for the method we use to quantify the fitting errors δ_{err}^{G} and δ_{err}^{MG}.

[35] See Supplementary Materials at [link to be inserted by publisher] for the results of the WCA fluid simulations analogous to those of Fig. 3.

[36] $p(P_{xy})$ is Gaussian when $B = 0$.

[37] To check this, one needs to solve a quadratic inequality $d^2\Delta S(x)/dx^2 \geq 0$ for x.

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