Asymmetry of steady state current fluctuations in nonequilibrium systems

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(Dated: August 20, 2015)

Abstract

For systems in nonequilibrium steady states, a novel modulated Gaussian probability distribution is derived to incorporate a new phenomenon of biased current fluctuations, discovered by recent laboratory experiments and confirmed by molecular dynamics simulations. Our results consistently extend Onsager-Machlup fluctuation theory for systems in thermal equilibrium. Connections with the principles of Statistical Mechanics due to Boltzmann and Gibbs are discussed. At last, the modulated Gaussian distribution is of potential interest for other statistical disciplines, which make use of the Large Deviation theory.

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

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I. INTRODUCTION

In this letter the fluctuation theory of currents, originally proposed by Onsager and Machlup for systems in thermal equilibrium Ref. [9], is extended to the nonequilibrium Steady States (SS). We suggest a novel Modulated Gaussian (MG) probability distribution, which supersedes the normal distribution, used to describe the fluctuations of thermodynamic variables in equilibrium. This new model features an asymmetry of the current fluctuations, a property inherent to the nonequilibrium regime. MG includes the Gaussian distribution as a special case so that the earlier theory is consistent with ours. Furthermore, in Sec. V connections between our formalism and the principles of statistical mechanics due to Boltzmann and Gibbs are discussed.

Our research was motivated by the evidence of non-Gaussianity of current fluctuations in a nonequilibrium SS, found in recent laboratory experiments Ref. [3] on a shear flow in a 2-Dimensional (2D) dusty plasma, similar to that considered in Ref. [5]. The new probability distribution, which is based on the Large Deviation (LD) approach (Ref. [12]), allows to account for this newly observed phenomenon. Our theoretical results are confirmed by Molecular Dynamics (MD) simulations of a shear flow for a Debye-Hückel (DH) plasma.

The asymmetry of nonequilibrium SS current fluctuations, studied in this paper, is due to a probability bias, namely due to the higher probability for the current to deviate from its most likely value towards larger magnitudes, rather than to smaller ones. Naturally, equilibrium systems are free of such a bias, since their most likely magnitude of a current is zero.

Finally, we would like to remark, that the MG probability distribution, which is derived in Sec. III without any specific physical assumptions, relies solely on the LD theory. Therefore, whenever the latter holds, the MG should also be applicable, in principle, to random variables outside the field of Statistical Mechanics.

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1 The temporal asymmetry predicted for trajectories of fluctuations onset and decay by the so called macroscopic fluctuation theory Ref. [2] is different from the bias of the time-independent nonequilibrium SS probabilities, studied here.
II. COMPUTATIONAL MODEL

In our numerical experiments we studied an iso-thermal MD model of a 2D plasma system. We will concentrate on computations performed with \( N = 100 \) particles of a unit mass \( m \), interacting through the DH potential \( \Phi_{DH} \), shifted and truncated at a cutoff \( r_c \):

\[
\Phi_{DH}(r) = \begin{cases} 
\epsilon \lambda \left[ r^{-1} \exp(-\frac{r}{\lambda}) - r_c^{-1} \exp(-\frac{r}{\lambda}) \right], & \text{if } r < r_c \\
0, & \text{if } r \geq r_c 
\end{cases},
\]

where \( r \) is the distance between two interacting particles, while the energy constant \( \epsilon \) and the Debye screening length \( \lambda \) are chosen as the basis of a reduced units system.

A constant shear rate and temperature of the plasma were maintained by the SLLOD equations of motion coupled to the Nosé-Hoover (NH) thermostat in \( D = 2 \) dimensions (Ref. [4]):

\[
\begin{align*}
\dot{q}_i &= \frac{p_i}{m} + \gamma q_{iy} \mathbf{X} \\
\dot{p}_i &= F_i(q_i) - \gamma p_{iy} \mathbf{X} - \alpha_{NH} p_i \\
\dot{\alpha}_{NH} &= \theta^{-2} \left( \sum_{i=1}^{N} \frac{p_i^2}{D N k_B T m} - 1 \right).
\end{align*}
\]

Here \( \mathbf{X} \) is a unit vector along the Cartesian x-coordinate axis, \( k_B \) is the Boltzmann constant, \( \gamma = \frac{du_x}{dy} \) is the applied shear rate, \( i.e. \) the gradient of the streaming velocity \( u = (u_x, 0) \); while \( q_i, p_i = m(q_i - u) \) and \( F_i \) are, respectively, the \( i \)-th particle position \((q_{iy} \) being its y-coordinate\), peculiar momentum and force on the \( i \)-th particle, resulting from the interactions with all the other particles; finally, the NH thermostat is characterized by its temperature \( T \), the relaxation time \( \theta \) and the time-dependent coupling \( \alpha_{NH} \). The flux of x-momentum along the y-coordinate \( P \) generated by Eq. [2] in the primary simulation cell of the linear dimension \( L \), is

\[
P_{xy} = L^{-D} \sum_{i=1}^{N} \left( \frac{p_{ix} p_{iy}}{m} + F_{ix} y_i \right).
\]

\[I.e.\] the xy-component of the pressure tensor
In all our simulations $k_B T$ is set equal to $1 \epsilon$, whereas $\theta$ was adopted as the unit of time. The equations of motion were integrated by the classical 4th-order Runge-Kutta algorithm (Ref. [7]), with the time step $0.001 \theta$ and the potential cutoff $r_c = 3 \lambda$. The linear size of primary cell $L$ was $8.86 \lambda$.

III. MODULATED GAUSSIAN DISTRIBUTION

In this section we present a derivation of the MG distribution for the flux Eq. (3). We begin with a probability density function of the form:

$$p(P_{xy}) = \exp\left\{\frac{S(P_{xy})}{k_B}\right\}.$$  

(4)

The function $S(P_{xy})$, which depends implicitly on $N$, will be interpreted in Sec. V. Considering each term of the summation operator in Eq. (3) as a random variable, we assume, that their spatial average $P_{xy}$ obeys the LD theory. That is, there exists a rate function

$$I(P_{xy}) = - \lim_{N \to \infty} \left\{ \frac{S(P_{xy})}{Nk_B} \right\},$$

(5)

which has a global minimum

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

(6)

at the most likely value of $P_{xy} = \tilde{P}_{xy}$ (cf. Ref. [12]).

Expanding $S(P_{xy})$ in Eq. (4) about $\tilde{P}_{xy}$ in a power series up to a prescribed finite order $n$, we obtain:

$$S(P_{xy}) \approx \frac{S(\tilde{P}_{xy})}{k_B} + \sum_{i=1}^{n} \frac{S^{(i)}(\tilde{P}_{xy})}{i!k_B} (P_{xy} - \tilde{P}_{xy})^i$$

$$\overset{\text{def}}{=} - \tilde{S} + \Delta S(\Delta P_{xy})$$

(7)

which defines (\overset{\text{def}}{=}) a constant $\tilde{S}$ and a cost function $\Delta S(\Delta P_{xy})$ of the deviation $\Delta P_{xy} = P_{xy} - \tilde{P}_{xy}$, which will be considered from a physical perspective in Sec. V.

From now on, the values of the derivatives $S^{(i)}(\tilde{P}_{xy})$ will be denoted by $S_i$, for brevity. Since the cost function is zero for $P_{xy} = \tilde{P}_{xy}$, Eqs. (5 6) suggest to pose:
\[
\lim_{N \to \infty} \frac{\tilde{S}}{N k_B} = 0
\]

\[
I(P_{xy}) \approx -\lim_{N \to \infty} \frac{\Delta S(\Delta P_{xy})}{N k_B}.
\] (8)

As \( p(\tilde{P}_{xy}) \) is the global maximum of probability density, it follows that the derivative \( S_1 \) vanishes. For a finite \( N \) Eq. (4) then becomes:

\[
p(P_{xy}) \approx \exp\left\{ -\frac{\tilde{S}}{k_B} + \frac{\Delta S(\Delta P_{xy})}{k_B} \right\} = \exp\left\{ -\frac{\tilde{S}}{k_B} + \sum_{i=2}^{n} \frac{S_i}{i! k_B} (\Delta P_{xy})^i \right\},
\] (9)

where the normalization of the total probability requires that

\[
\exp\left\{ \frac{\tilde{S}}{k_B} \right\} = \int_{-\infty}^{\infty} dP_{xy} \Delta S(P_{xy} - \tilde{P}_{xy}).
\]

For \( n = 2 \), Eq. (9) turns into a Gaussian distribution. One may recognize that, this case was treated by Onsager and Machlup in Ref. [9, 10] for the equilibrium state \( (\tilde{P}_{xy} = 0) \). Therefore to account for non-Gaussian phenomena, one must consider higher order terms of the cost function, which can be arranged as follows:

\[
\Delta S(\Delta P_{xy}) = \frac{S_2}{2 k_B} (\Delta P_{xy})^2 + \sum_{i=3}^{n} \frac{S_i}{i! k_B} (\Delta P_{xy})^i
\]

\[
= \frac{S_2}{2 k_B} (\Delta P_{xy})^2 \left\{ 1 + 2 \sum_{i=3}^{n} \frac{S_i}{i! S_2} (\Delta P_{xy})^{i-2} \right\},
\] (10)

where the term in the curly braces \( \Sigma_n = 1 + 2 \sum_{i=3}^{n} \frac{S_i}{i! S_2} \Delta P_{xy}^{i-2} \) is a modulating factor, to which the MG distribution owes its name.

The original Gaussian is recovered, when \( \Sigma_n \equiv 1 \). For \( p(P_{xy}) \) to be integrable in Eq. (9), the order \( n \) has to be restricted to even integers. Using the lowest order non-Gaussian approximation, \( i.e. \) \( n = 4 \), we replace the derivatives \( S_i \) by three parameters of scale \( \Pi \), of asymmetry \( A \), and of non-Gaussianity \( B \), which allow a more convenient interpretation, in Eq. (9):

\[\text{Especially, current fluctuations about equilibrium are considered in Ref. [9].}\]
\[ p(P_{xy}) \propto \exp\{\Delta S(\Delta P_{xy})\} = \exp\left\{ \frac{S_2(\Delta P_{xy})^2}{2k_B} + \Delta P_{xy} \frac{\Delta P_{xy}}{\Pi} + B^2 \frac{(\Delta P_{xy})^2}{\Pi^2} \right\} \]

\[ = \exp\left\{ -\frac{(\Delta P_{xy})^2}{2\Pi^2} (1 - 2\sqrt{2/3}AB + B^2 \frac{\Delta P_{xy}}{\Pi} + B^2 \frac{(\Delta P_{xy})^2}{\Pi^2}) \right\}. \tag{11} \]

The dimensionless non-negative constant \( B \geq 0 \) controls the level of non-Gaussianity, thus \( p(P_{xy}) \) is exactly Gaussian for \( B = 0 \) with the scale parameter \( \Pi \). \( A \) determines the asymmetry of \( p(P_{xy}) \), which is skew to the left (right), \textit{i.e.} has a negative (positive) skewness, when \( A < 0 \) (\( A > 0 \)), respectively. The numerical factor \( 2\sqrt{2/3} \) in Eq. (11), as a coefficient of the term with \( A \), was chosen to make \( \Delta S(\Delta P_{xy}) \) a non-concave function of \( P_{xy} \), \textit{i.e.} \( \frac{d^2 \Delta S(\Delta P_{xy})}{dP_{xy}^2} \geq 0 \), when \( -1 \leq A \leq 1 \). Violation of the non-concavity condition would admit special artifacts of the probability distribution, \textit{e.g.} a local maximum of its density. For experimental or numerical observations, such flexibility may result in a data over-fitting, and hence should be suppressed by using constrained optimization techniques.

IV. NON-GAUSSIANITY OF CURRENT FLUCTUATIONS

Fig. 1 illustrates probability density estimates of the momentum current \( P_{xy} \), at low and high shear rates, for our computational experiments, including the fits of the Gaussian and modulated Gaussian distributions (Eq. (11)). For the low value of \( \gamma = 0.2\theta^{-1} \) an excellent agreement is observed between the shape of histograms and the parametric models (indistinguishable on the graph for the lower shear rate). However, the Gaussian model renders an unsatisfactory result in the case of a larger value \( \gamma = 1.0\theta^{-1} \), where the superior performance of the MG distribution is evident.

Most obviously, the Gaussian model fails, when the fluctuations are asymmetric about the mode of the probability distribution \( \hat{P}_{xy} \), as can be quantified by the skewness statistics. Increasing the departure from equilibrium augments the non-Gaussianity of \( p(P_{xy}) \), as confirmed by the plots of the skewness and excess kurtosis, which are both zero for the normal distribution, \textit{vs.} the magnitude of the shear rate in Fig. 2. This occurs, because in

\[ \text{4} \text{ Everywhere we adopt the Freedman-Diaconis rule (Ref. [6]) to calculate the bin width of traditional histograms and the band width of smooth histograms with the Gaussian kernel (Ref. [11]).} \]

\[ \text{5} \text{ Because the normalizing constant of the MG can be evaluated only numerically, to obtain the corresponding Maximum Likelihood Estimator (MLE) of the MG model, we used the Wolfram Mathematica}^{\text{®}} \text{ implementation of the interior point method with the constraint of non-concavity, mentioned at the end of Sec. [11].} \]
FIG. 1. Probability density of $P_{xy}$: (a) for a low shear rate in the left panel; (b) for a high shear rate in the right panel. For the low shear rate the Gaussian and the MG are indistinguishable.

In nonequilibrium the probability of a positive deviation $\Delta P_{xy} > 0$ from the most likely value $\tilde{P}_{xy}$ becomes different from the probability of the opposite deviation $-\Delta P_{xy}$. Fig. 2 also demonstrates the efficiency of the modulated Gaussian to fit high-order statistics.

FIG. 2. Statistics of skewness and excess kurtosis as a function of the shear rate for our observations of $P_{xy}$ and the corresponding fits by the MG.

For a positive shear rate, which maintains a negative flux $P_{xy}$ (cf. Fig. 1), we observe $p(\Delta P_{xy} > 0) < p(-\Delta P_{xy})$. The MG distribution accounts for this bias of probabilities and fits very well the 3rd and 4th order statistics (skewness and kurtosis, respectively). We
conjecture, that the asymmetry of the distribution $p(P_{xy})$ is a characteristic property of the current fluctuations about a nonequilibrium SS. It is small, though, in the near-equilibrium SS regime and vanishes only in the limit $\gamma \to 0$.

V. CONNECTION WITH CLASSICAL STATISTICAL MECHANICS

The results obtained in Sec. III can be readily connected with the Boltzmann and Gibbs principles of statistical mechanics. According to the Boltzmann principle, given a measure of system microstates $w(P_{xy})$, for a given value of $P_{xy}$, and the total measure of the accessible microstates $W = \int_{-\infty}^{\infty} w(P_{xy})dP_{xy}$ under the specified macroscopic constraints of temperature, volume, shear rate etc., the SS probability density $p(P_{xy})$ and the Boltzmann entropy $S_B(P_{xy})$ are given by:

$$
p(P_{xy}) = \frac{w(P_{xy})}{W},
S_B(P_{xy}) = k_B \ln w(P_{xy}), \quad (12)
$$

respectively.

Using the notion of total entropy $S_{tot} = k_B \ln W$ after Ref. [1], we deduce from Eq. (12) that:

$$
k_B \ln p(P_{xy}) = k_B \ln w(P_{xy}) - k_B \ln W = S_B(P_{xy}) - S_B(\tilde{P}_{xy}) + S_B(\tilde{P}_{xy}) - S_{tot}
= \Delta S_B(P_{xy}) + S_B(\tilde{P}_{xy}) - S_{tot}, \quad (13)
$$

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. (9) with Eq. (13), one sees that

$$
\tilde{S} = S_{tot} - S_B(\tilde{P}_{xy})
\Delta S(P_{xy} - \tilde{P}_{xy}) = \Delta S_B(P_{xy}), \quad (14)
$$

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.
Hence Eq. (14) provides the interpretation of the cost function $\Delta S(\Delta P_{xy})$ and the constant $\tilde{S}$, introduced in Sec. III, in terms of Boltzmann entropy and the total entropy. The most likely value of $P_{xy} = \bar{P}_{xy}$ maximizes the Boltzmann entropy (cf. Eq. (12)). Consistently, $\Delta S(\Delta P_{xy}) \leq 0$ is the (Boltzmann) entropy cost of a fluctuation $P_{xy} = \bar{P}_{xy} + \Delta P_{xy}$ \footnote{One can also define the (free) energy cost of a fluctuation $\Delta F = \Delta S(P_{xy})T$, cf. Ref. [8].}. The constant $\tilde{S}$ is the remaining total entropy, after subtracting the entropy cost of the most likely macrostate.

Finally, the Gibbs entropy, given by a functional $S_G[p]$, for the distribution Eq. (9) is

$$S_G[p(\cdot)] = -k_B \int_{-\infty}^{\infty} dP_{xy} p(P_{xy}) \ln p(P_{xy}) = -k_B \langle \ln p(P_{xy}) \rangle$$

$$= \langle \tilde{S} - \Delta S(P_{xy}) \rangle = \tilde{S} - \langle \Delta S(P_{xy}) \rangle$$

$$= S_{tot} - S_B(\bar{P}_{xy}) - \langle \Delta S(P_{xy}) \rangle,$$

which extracts $\tilde{S}$, up to a constant term $\langle \Delta S(P_{xy}) \rangle$, from the SS probability density $p(P_{xy})$.

VI. CONCLUSION

In Sec. III we used the 4th order modulating factor ($\Sigma_4$) to derive our modulated Gaussian probability distribution. In principle, the same procedure can be applied to obtain the next order approximation, using $\Sigma_6$. However, the formalism becomes then much more complicated. The performance of the forth order factor was demonstrated in Sec. IV and proved to be sufficient to describe the asymmetry of fluctuations and the decay of their probability distribution tails. Although we presented our data only for the simulations with $N = 100$ particles, we checked that the modulated Gaussian performs equally well for smaller and larger values $N$ as well.

An important consequence of the asymmetric fluctuations about the SS is the discrepancy between the current average and its most likely value $\langle P_{xy} \rangle \neq \bar{P}_{xy}$, which are equal in equilibrium. Since we do not provide the dynamics of fluctuations, like the Langevin equation suggested by Onsager and Machlup for equilibrium, it is an open question, how this probability bias emerges. There are various ways to modify the Langevin equation to produce
asymmetric time-independent distribution, e.g. non-Gaussian noise or non-linear deterministic term. Physically, this could be a consequence of the arrow of time in a nonequilibrium system, which admits a bias. Yet a dynamical theory remains to be found.

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