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# The entropy and fluctuation theorems of inertial particles in turbulence

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We study Lagrangian particles, with Stokes numbers,  $St = 0.5, 3,$  and  $6,$  transported by homogeneous and isotropic turbulent flows. Based on direct numerical simulations with point-like inertial particles, we identify stochastic equations describing the multi-scale cascade process. We show that the Markov property is valid for a finite step size larger than a  $St$ -dependent Einstein-Markov memory length. The formalism allows estimation of the entropy of the particles' Lagrangian trajectories. Integral, as well as detailed fluctuation theorems are fulfilled. Entropy consuming trajectories are related to specific local accelerations of the particles and may be seen as reverse cascade processes.

The physics of particles submerged in fluids has played a central role in the development of statistical mechanics, and in our current understanding of out-of-equilibrium systems. Brownian motion set the path for the study of diffusion and random processes. More recently, colloidal particles were used in the first experiments to verify fluctuation relations such as the Jarzynski equality [1], which links the statistics of fluctuating quantities in a non-equilibrium process with equilibrium quantities. Colloidal particles were also used to verify the thermodynamic cost of information processing [2] proposed by Landauer. But what fundamental statistical relations are satisfied by particles that interact with a complex and out-of-equilibrium turbulent flow? Fluctuation theorems for such a problem, in which a physical system is coupled with (and driven by) another out-of-equilibrium system, would open applications in other areas such as soft and active matter. But even a simple point-wise passive particle in a turbulent flow already provides a challenge in which puzzling phenomena can arise [3, 4].

Inertial particles are inclusions in the flow which are denser or lighter than the fluid and have a size smaller or larger than the smallest relevant flow scale (the scale of the smallest eddies, also called the flow dissipative scale). Such particles are carried by the fluid, but they also have their own inertia, and therefore can combine both dynamics. In the limit of point-wise particles with negligible inertia, the particles become Lagrangian tracers, which perfectly follow the fluid elements. In all these multi-phase systems there are striking phenomena, and the mechanisms that explain how turbulence affects the motion of the particles are not completely clear. As an example, turbulence can both enhance or hinder the settling velocity of inertial particles [5]. For heavy particles, an initially homogeneous distribution of particles may, after interacting with a turbulent flow, regroup into clusters forming dense areas and voids, in a phe-

nomenon called preferential concentration where turbulence somehow unmixes the particles [6]. It is only in the last decades when sufficient time and spatial resolution have been achieved in experiments and numerical studies to allow analysis of these phenomena. Frequently new data has been in contradiction with theoretical models, and previous knowledge on fluid-particle interactions had to be reconsidered even in simplified cases [7]. Furthermore, there are many open questions concerning inhomogeneous flows [8], finite-size [9, 10] and non-spherical particles [11], among others.

Recent techniques have also allowed a better understanding of the (single-phase) turbulence cascade as a stochastic process. Thermodynamically speaking we can understand turbulence, and especially its energy cascade, as a process leading a fluid under specific conditions and parameters (the Reynolds number) from a non-equilibrium state into another non-equilibrium state, by the combination of energy injection and dissipation. Within this context, the stochastic approach to the description of turbulence has recently been linked to stochastic thermodynamics [12, 13] (also called stochastic energetics [14]), developed for many different physical systems [15–18]. Experimental studies of stochastic thermodynamics mainly focus on nanoscale or quantum systems, or when dealing with classical systems, on biological systems [19, 20], which are assumed to be well off the thermodynamic limit so that the probabilistic nature of balance relations becomes clearer. In an Eulerian description of turbulent flows, the use of the Fokker-Planck equation has allowed to define the Shannon entropy of individual “cascade trajectories” (here meant in the Eulerian sense), and corresponding to a sequence of velocity correlations at decreasing scales, with either entropy consumption or production. These entropy values follow a rigorous law of non-equilibrium stochastic thermodynamics, namely, the integral fluctuation theorem (IFT)

which can be expressed as  $\langle e^{-\Delta S_{tot}} \rangle = 1$ , where  $\Delta S_{tot}$  is the total entropy variation in each cascade trajectory, cf. [21].

Based on the general interest in the relation between Eulerian and Lagrangian properties of turbulence the question of the validity of such theorems for Lagrangian and inertial particles arises, which would have multiple consequences. First, particle trajectories have a clear physical meaning: each particle follows a path according to the fluid dynamics and its own inertia. The distinction between these particles is quantified by the Stokes number  $St$  (the ratio of the particle relaxation time to the fluid dissipation time), with the limit of small  $St$  corresponding to tracers. Second, for particles with inertia the description corresponds to a non-trivially coupled system, as these particles do not randomly sample the flow topology [6].

To this end, in this letter we verify if the stochastic formalism can be applied to the dynamics of dense sub-Kolmogorov particles coupled to a turbulent flow, using pseudo-spectral direct numerical simulations (DNSs) with a simple point particle model [22]. We also consider whether the IFT holds for particles with different Stokes numbers, and study the properties of the Lagrangian trajectories with positive and negative entropy variations. The turbulent velocity field of three-dimensional homogeneous and isotropic turbulence (HIT) is obtained from the DNSs by solving the incompressible Navier-Stokes equation. A large-scale external mechanical forcing is given by a superposition of modes with slowly evolving random phases, following standard practices for its temporal integration and de-aliasing procedures. An adequate spatial resolution of the smallest scales, i.e.,  $\kappa\eta \gtrsim 1$  is chosen [23]. Here,  $\eta$  is the Kolmogorov or dissipation length scale,  $\eta = (\nu^3/\varepsilon)^{1/4}$  (where  $\varepsilon$  is the kinetic energy dissipation rate, and  $\nu$  the kinematic viscosity of the fluid), and  $\kappa = N/3$  the maximum resolved wavenumber in Fourier space (with  $N = 512$  the linear spatial resolution in each direction). The DNSs have a Reynolds number based on the Taylor microscale of  $Re_\lambda \approx 240$  (see [6] for details). Inertial particles were modeled using the Maxey-Riley-Gatignol equation in the limit of point heavy particles, which for a particle with velocity  $\mathbf{v}$  in the position  $\mathbf{x}_p$  submerged in a flow with velocity  $\mathbf{u}$ , reads

$$\dot{\mathbf{v}}(t) = [\mathbf{u}(\mathbf{x}_p, t) - \mathbf{v}(t)]/\tau_p, \quad (1)$$

where  $\tau_p$  is the particle Stokes time. There is no particle-particle or particle-fluid interaction (i.e., we use a one-way coupling approximation). As mentioned above, the particles have one dimensionless number of interest: the Stokes number  $St = \tau_p/\tau_\eta$  (where  $\tau_\eta = (\nu/\varepsilon)^{1/2}$  is the Kolmogorov time). We integrate three sets of  $1.3 \times 10^5$  particles, respectively with  $St = 0.5, 3$ , and  $6$ .

To characterize HIT it is customary to study the statistics of Eulerian velocity increments  $u_r = [\mathbf{u}(\mathbf{x} + \mathbf{r}) - \mathbf{u}(\mathbf{x})] \cdot \mathbf{r}/|\mathbf{r}|$  for scales  $r = |\mathbf{r}|$ . However,

two-point statistics of velocity increments do not fully characterize small-scale turbulence [24], and many attempts at dealing with multi-point statistics have been considered. One way to do this is to use the Friedrich-Peinke approach [25], in which the stochastic dynamics of velocity increments  $u_r$  are considered as they go through the cascade from large to small scales  $r$ . A central assumption is that the evolution of the stochastic variable  $u_r$  possesses a Markov process “evolving” in  $r$ . Previous studies showed that  $u_r$  can be considered as Markovian to a reasonable approximation [25–27], at least down to a scale close to the Taylor scale [27, 28]. Furthermore,  $u_r$  satisfies a diffusion process [29]

$$-\partial_r u_r = D^{(1)}(u_r, r) + [D^{(2)}(u_r, r)]^{1/2} \Gamma(r), \quad (2)$$

where the noise  $\Gamma(r)$  is zero-mean, white-noise Gaussian with a variance of 2 and rapidly decaying correlations, such that  $\delta$ -correlation in scale can be assumed as  $\langle \Gamma(r)\Gamma(r') \rangle = 2\delta(r - r')$ . The drift and diffusion coefficients  $D^{(1,2)}(u_r, r)$  can be estimated from experimental data by an optimization procedure, based on approximating the solution of the corresponding Fokker-Planck equation using the short time propagator [30] as proposed in [13, 21, 31–33]. A linear dependence on the value of the increment for the drift, and a quadratic dependence for the diffusion, was found cf. [13, 24, 27, 32, 34]. From the drift and diffusion coefficients, the entropy can then be defined as will be detailed below. This entropy allows to verify the validity of the IFT.

Here we focus instead on the statistics of the inertial particles in the turbulent flow, integrated for sufficiently long times to reach their steady state. To apply the previously described approach to particles trajectories, we study  $\mathbf{v}(t)$  (the velocity of the particle at time  $t$ ). Similar conclusions as the ones shown below are obtained by studying the fluid velocity at the particle position  $\mathbf{u}(\mathbf{x}_p, t)$ , or even the slip velocity  $\mathbf{u}(\mathbf{x}_p, t) - \mathbf{v}(t)$ . Building now velocity increments in time component wise ( $\mathbf{e}_i$  corresponds to the unitary vector in the direction  $x, y$ , or  $z$  specified in the figures)

$$u_\tau = [\mathbf{v}(t + \tau) - \mathbf{v}(t)] \cdot \mathbf{e}_i, \quad (3)$$

we define for every trajectory a “cascade trajectory” (or “sequence”)  $[u(\cdot)] = \{u_T, \dots, u_{\tau_f}\}$  for different time-separations or time scales  $\tau$ , from the initial time scale  $T$  to the final time scale  $\tau_f$  with  $T > \tau_f$ . The notation  $[u(\cdot)]$  indicates the entire path through the hierarchy of time scales instead of a distinct value  $u_\tau$ ;  $T \approx 120\tau_\eta$  and  $\tau_f \approx 0.2\tau_\eta$  were chosen as references.

To test the IFT for the particles, we consider whether the  $u_\tau$  increments satisfy a Langevin equation as in Eq. (2), but in terms of the time increment  $\tau$  instead of spatial scales  $r$ , with equivalent  $D^{(1)}$  and  $D^{(2)}$  coefficients. But before using the DNS data to estimate the probability density function (PDF) of  $u_\tau$  and  $\tau$ ,  $p(u_\tau, \tau)$ , and of estimating these coefficients, we must check the

validity of the Markovian approximation. Indeed, as mentioned above, a central assumption of the Friedrich-Peinke approach is that the evolution of the stochastic variables possess a Markov process. While this was shown to be valid for the Eulerian turbulent velocity, it is an open question for Lagrangian particles' velocities.

In Fig. 1 a qualitative validation of the Markov property based on the alignment of the single conditioned  $p(u_{\tau_2}|u_{\tau_1})$  and double conditioned  $p(u_{\tau_2}|u_{\tau_1}, u_{\tau_0})$  PDFs of datasets of increments for a chosen set of three different time scales  $\tau_0 > \tau_1 > \tau_2$  is shown as contour plots. Each scale is separated by  $\Delta\tau \approx 13\tau_\eta$  (the Einstein-Markov length for  $St = 3$  as described below, and in the discussion accompanying Fig. 2). For Markovian processes the relation  $p(u_{\tau_2}|u_{\tau_1}) = p(u_{\tau_2}|\dots, u_{\tau_1}, u_{\tau_0})$  holds. For finite datasets,

$$p(u_{\tau_2}|u_{\tau_1}) = p(u_{\tau_2}|u_{\tau_1}, u_{\tau_0}), \quad (4)$$

is commonly assumed to be a sufficient condition. The close alignment between the PDFs (black and red solid lines in Fig. 1) confirms the validity of the Markov property. Note that while Fig. 1 shows results for  $St = 3$ , all our datasets give similar results.

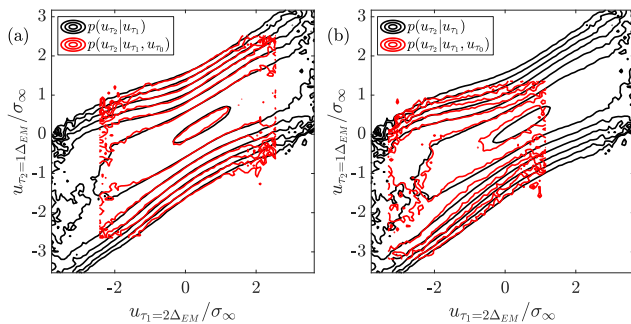


FIG. 1. Visualization of Markov properties at  $St = 3$ . Contour plots showing single conditioned (black solid lines) and double conditioned PDFs (red solid lines) of velocity increments for three different time scales  $\tau_0 > \tau_1 > \tau_2$ , each separated by  $\Delta\tau$ . The conditioned value for the large increment was chosen here as  $u_{\tau_0} = 0$  (left) and  $u_{\tau_0} = -2$  (right). We use the normalization of the increments in [27] with  $\sigma_\infty = \sqrt{2}\sigma$ , where  $\sigma$  is the data standard deviation.

In Fig. 2 the Markovian approximation is checked systematically for different values of  $\Delta\tau$ , using the Wilcoxon test, which is a quantitative and parameter-free test that determines the Einstein-Markov coherence length  $\Delta_{EM}$  (i.e., the smallest time-scale for which the Markovian assumption can be considered valid). This test is a reliable procedure to validate equation Eq. (4) [27, 28]. Based on this analysis, we set  $\Delta_{EM} \approx 10, 13$ , and  $16\tau_\eta$  for  $St = 0.5, 3$ , and  $6$  respectively (see Fig. 2 (a)). Accordingly, the Markovian approximation is valid for time-scales larger than or equal to this  $St$ -dependent critical time separation, and the complexity of the dynamics of inertial particles in turbulent flows can be treated as a Markov

process. Furthermore, we see that  $\Delta_{EM}$  is the same (at fixed  $St$ ) for all velocity components (see Fig. 2 (b)). The  $D^{(1,2)}$  coefficients can be empirically estimated from the data (see Supplemental Material [35] for a detailed presentation of the coefficients associated with all Stokes numbers considered).

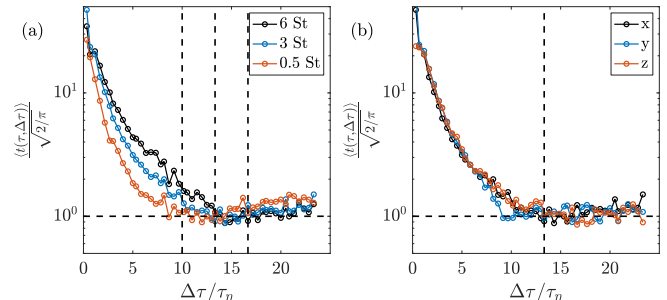


FIG. 2. Wilcoxon test for the particles' trajectories: expectation value  $t(\tau, \Delta\tau)$  (see [27, 28] for details; note that the normalized expectation parameter is supposed to be close to 1 if the Markovian assumption is valid) for the  $x$  component of the particles velocity, for all Stokes numbers studied here and as function of  $\Delta\tau$  (left). Same test but for all three velocity components at  $St = 3$  (right).

In the spirit of non-equilibrium stochastic thermodynamics [36] it is possible to associate with every trajectory  $[u(\cdot)]$  a total entropy variation [12–14, 18, 36] given by the sum of two terms  $\Delta S_{tot}[u(\cdot)] = \Delta S_{sys}[u(\cdot)] + \Delta S_{med}[u(\cdot)]$ . A thermodynamic interpretation of this quantity based on the relation between heat, work, and inner energy, can be given [14, 18, 36].  $\Delta S_{sys}$  is the change in the system entropy caused by changes in the particle trajectory. It is simply the logarithmic ratio of the probabilities of the stochastic process, so that its variation is

$$\Delta S_{sys}[u(\cdot)] = -\ln\left(\frac{p(u_{\tau_f}, \tau_f)}{p(u_T, T)}\right). \quad (5)$$

The other term, the entropy exchanged with the surrounding medium  $\Delta S_{med}$  throughout the process from the initial to the final time scale, measures the irreversibility of the trajectories:

$$\Delta S_{med}[u(\cdot)] = \int_T^{\tau_f} \left[ \partial_\tau u_\tau \frac{D^{(1)} - \partial_{u_\tau} D^{(2)}/2}{D^{(2)}} \right] d\tau. \quad (6)$$

Figure 3 (a) shows the empirical average  $\langle e^{-\Delta S_{tot}} \rangle_N$  as a function of the number  $N$  of trajectory sequences  $[u(\cdot)]$ . See Supplemental Material [35] for the illustration of  $p(\Delta S_{tot})$  for  $St = 0.5, 3$  and  $6$ . The dashed line in the figure 3 (a) corresponds to the integral fluctuation theorem (IFT)

$$\langle e^{-\Delta S_{tot}} \rangle = \int e^{-\Delta S_{tot}} p(\Delta S_{tot}) d\Delta S_{tot} = 1, \quad (7)$$

which is a fundamental entropy law for non-equilibrium systems [18, 36]. We find that the three Stokes numbers are in agreement with the IFT. This is not a trivial result, as inertial particles (i.e., for sufficiently large  $St$ ) sample preferentially some specific regions of the flow [6]. The remarkable observation here is that they do it always in agreement with this theorem. We performed a similar analysis for the flow velocity at the particle position  $\mathbf{u}(\mathbf{x}_p, t)$ , and the slip velocity  $\mathbf{u}(\mathbf{x}_p, t) - \mathbf{v}(t)$  (not shown here), and they also satisfy the IFT (with, interestingly, an Einstein-Markov time-scale that decreases with increasing  $St$ , unlike the behavior observed for  $\mathbf{v}(t)$ ). Figure 3 (b) shows that together with the IFT also the detailed fluctuation theorem (DFT) holds, which is expressed as

$$\ln \left( \frac{p(\Delta S_{tot})}{p(-\Delta S_{tot})} \right) \propto \Delta S_{tot}. \quad (8)$$

Thus, in addition to the IFT, the DFT expresses the balance (explicit exponential symmetry constraint) between entropy-consuming ( $\Delta S_{tot} < 0$ ) and entropy-producing ( $\Delta S_{tot} > 0$ ) trajectories.

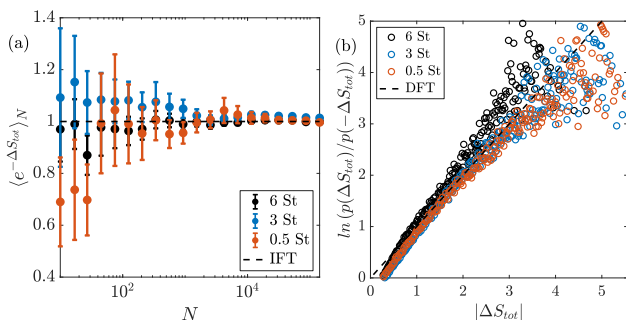


FIG. 3. Left: Average  $\langle e^{-\Delta S_{tot}} \rangle_N$  as a function of the sample size  $N$  of  $[u(\cdot)]$  trajectories. The arrow indicates the direction of the path through the hierarchy of time scales  $\tau$ , from the initial time scale  $T$  to the final time scale  $\tau_f$ . Right: According to the integral fluctuation theorem, the empirical average has to converge to a value of 1 (indicated by the horizontal dashed line). Test of the detailed fluctuation theorem. The dashed line represents a linear behavior  $p(\Delta S_{tot}) = p(-\Delta S_{tot}) e^{\Delta S_{tot}}$ .

Finally we ask if the entropy values of the trajectories are also related to some flow structure therefore we study the velocity increment trajectories conditioned on a specific total entropy variation. Figure 4 (a) and (c) show that the behavior of the total entropy variation corresponds to distinct trajectories. Entropy-consuming trajectories are characterized by an increase in the averaged absolute values of the increments  $\langle |u_\tau| \rangle_{\Delta S_{tot}}$  with decreasing time scale  $\tau$ , while trajectories marked by entropy-production smoothly decrease their absolute increments with decreasing  $\tau$ . This is further highlighted by Fig.4 (b) and (d). In this three-dimensional representation the absolute velocity increment at initial and final

scale,  $T, \tau$ , conditioned on the entropy are shown.

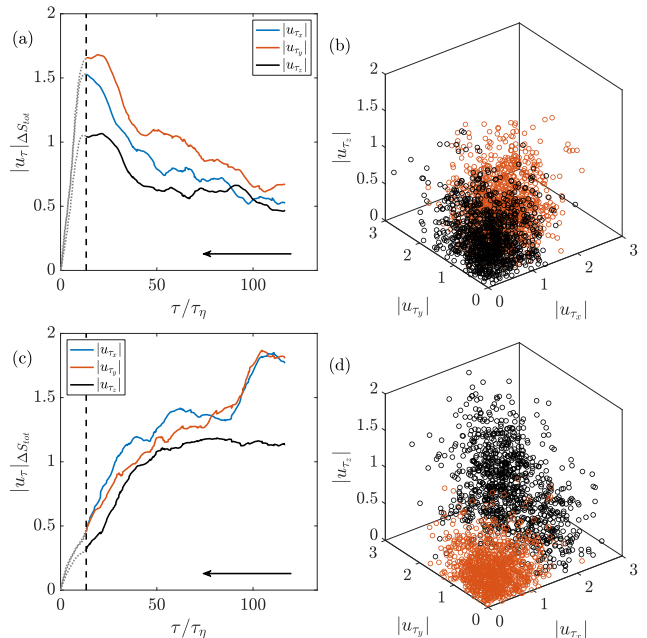


FIG. 4. Mean absolute velocity increment trajectories conditioned on a specific total entropy variation  $\langle |u_\tau| \rangle_{\Delta S_{tot}}$ ,  $\Delta S_{tot} = -2$  (top) and  $\Delta S_{tot} = 3$  (bottom). Left: individual representation of the components  $x, y$  and  $z$ . The vertical dashed line marks the Einstein-Markov coherence length  $\Delta_{EM}$ . The dissipation region is indicated by dotted lines. Right: The circles mark the increment on the initial (black) and final (red) scale in a three dimensional scatter plot.

In conclusion, we studied the dynamics of inertial particles in turbulent flows via their Lagrangian trajectories by means of entropy and fluctuation theorems. Sequences of velocity increments of such particles are Markovian, with Einstein-Markov coherence length that increases with  $St$ . This opens an interesting interpretation of the  $St$  number in terms of the Markov memory of the particles' trajectories, and is compatible with the picture that particles with more inertia filter fast and small scale fluctuations of the carrying flow: for particles with larger  $St$  (and thus larger particle response times) the Markovianization of trajectories by the turbulence takes place at longer times. This can help to quantify this parameter in cases where the particles' inertia is not clearly defined, like active, finite-size, or even non-spherical particles.

All cases studied here fulfill both the IFT and DFT in a strict sense. Previous observations of irreversibility of particles in turbulence considered a Jarzynski-like equality for the particles' energetics [37], but the slope was not one as expected for a DFT. The results presented here provide fluctuations theorems which can shed light on asymmetries in particle trajectories as those reported before in [37], and as shown here for velocity increments depending on their entropy evolution. A connection can be made between entropy-increasing and -decreasing



trajectories with losing- or gaining-velocity fluctuations with decreasing time scale, pointing to possible relations with inverse or direct energy transfer events. Interestingly, our study also shows that particle trajectories are out-of-equilibrium while keeping ergodicity. This remains a first study on the topic, but it can be adapted to situations where particles are in other conditions, particularly in environmental situations, and with direct consequences for the modeling of systems in which particles are coupled to out-of-equilibrium extended systems.

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