Experimental verification of Arcsine laws in mesoscopic non-equilibrium and active systems

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A large number of processes in the mesoscopic world occur out of equilibrium, where the time course of the system evolution becomes immensely important – they being driven principally by dissipative effects. Non-equilibrium steady states (NESS) represent a crucial category in such systems – which are widely observed in biological domains – especially in chemical kinetics in cellular processes [1], and molecular motors [2]. In this study, we employ a model NESS stochastic system which comprises of a colloidal microparticle, optically trapped in a viscous fluid and externally driven by a temporally correlated colored noise, and show that the work done on the system and the work dissipated by it – both follow the three Lévy arcsine laws. These statistics remain unchanged even in the presence of a perturbation generated by a microbubble at close proximity to the trapped particle. We confirm our experimental findings with theoretical simulations of the systems. Our work provides an interesting insight into the NESS statistics of the meso-regime, where stochastic fluctuations play a pivotal role.

In 1940, Paul Lévy introduced three laws that described the long-term statistics of a one-dimensional Wiener process, often used to describe the trajectory of Brownian motion [3]. For the simplest case of Brownian motion, the particle’s position coordinate starts from zero. It has an equally probable incremental step in either direction, which does not depend on its past trajectory – a key signature of a Markovian process. Also, the increments have a zero-centered Gaussian nature where the variance of the distribution increases linearly with time. For such a process, three variables can be defined: a) the ratio of the last time when the variable had changed its sign to the total time: $T_{last} = \sup\{t \in [0,1] : W(t) = 0\}$, b) the ratio of the last time when the variable had changed its sign to the global maximum to the total time: $T_{max} : W(T_{max}) = \sup\{W(t), t \in [0,1]\}$. These variables are scaled from 0 to 1, and they can be shown to have a normalized probability density function, $PDF(T_{\pm}/T_{last}/T_{max}) = \frac{1}{\pi\sqrt{T_{\pm}(1-T_{\pm})}}$, as long as the root variable follows a Wiener process. It follows that the cumulative distribution function (CDF) of these three variables follow the arcsine law given by:

$$CDF(T) = \int_0^T PDF(T')dT' = \frac{2}{\pi} \arcsin(\sqrt{T}) \quad (1)$$

for $T = T_{\pm}/T_{last}/T_{max} \in [0,1]$.

A large number of processes in the natural sciences follow the properties of the Wiener process, and consequently, the arcsine laws. Notably, operations in the microscopic regime have low degrees of freedom, where fluctuations can dominate the system’s thermodynamic properties. These fluctuations are modeled successfully using the arcsine distributions. In recent times, the electron current in cold quantum dots [4], the net number of steps of a molecular motor [5], position fluctuation of a particle in a periodic potential [6], and the work done on the system in micrometer-sized stochastic Carnot engines [7], have been shown to follow the arcsine distribution. Apart from non-equilibrium systems, entirely disparate systems including the quantum state of a dressed photon in a fiber probe [8], the fluctuation of stock prices [9], waiting time distributions of human dynamics [10], leads in competitive sports like football or basketball [11] as well as various random walk models [12], are examples that follow arcsine distributions. These laws can even be modified to include the behavior of fractional Brownian motion [13], non-Markovian processes [14], and processes that display anomalous diffusion [15].

At mesoscopic length scales, the thermal fluctuations in the system play an important role since the energy of the system is in the order of $k_BT$. Very often, these systems which include Brownian motors [16] and engines [17], operate far from equilibrium by deriving energy from the external fluctuations, whereas their dissipation is dictated by the rate of entropy generated [18]. An interesting class of processes is those that are non-equilibrium, but in steady state – since, with the sophisticated micromanipulation techniques enabled by optical trapping, these can be studied in great detail even experimentally. However, apart from an experimental study of the work done in a Brownian Carnot engine [7], there exists a lacuna towards determining and validating arcsine properties of stochastic variables in mesoscopic systems, both periodic and non-periodic, experimentally. Indeed, microscopic engines developed around Carnot and Stirling cycles [17], as well as Brownian ratchets and motors [16], or even the simple manoeuvre of dragging a particle to and fro at a constant velocity [19] – all represent experimentally demonstrable periodic systems. In the last example, the free energy may not change over many pe-
periods – since, though it contains both steady state and transient effects – the particle may always relax back to equilibrium. However, for non-periodic systems that remain in a steady state out of equilibrium, the physics of operation is entirely different, with \((\Delta F \neq 0)\).

On another note, interest regarding systems driven with colored noise \(19, 20\) has been growing in the scientific community, primarily due to the fact that these serve as a tunable model in understanding real-world biological systems at the microscopic level – especially where phenomena such as non-Gaussian discharge statistics \(21\), active dissipative fluctuations \(22\), and flows \(23\) may become prominent. There is an obvious requirement to study the long-term statistics of stochastic micro-currents that drive these systems in a steady state, since any deviations from the arc sine laws will capture “aging” \(15\) and “non-markovian” \(13\) behavior that determine the efficiency of these micro-engines. For example, if a fluctuating current does not satisfy the Markov properties of a typical diffusing Wiener process, it will deviate from the arc sine behavior. Thus, it is possible to infer about underlying processes from the long-term statistics of these fluctuating currents, without scanning the system holistically.

In this paper, we experimentally demonstrate that both the work done by the colored noise and work dissipated by the micro-system in the process, follow Lévy arc sine statistics in a model NESS system. In our study, we trap a micron-sized polystyrene probe particle in water in a harmonic potential generated by optical tweezers, and drive it externally with a colored noise generated by the Ornstein-Uhlenbeck (OU) process. The work done in such a process changes the effective free energy of the system irreversibly. To maintain the temperature of the system, which is out of equilibrium with its surroundings, work needs to be done continuously by an externally added noise. A small component of that work is dissipated as heat, mainly due to viscous effects of the fluid where the probe is embedded. However, the presence of a microbubble in the fluid medium causes the generation of flow fields, so that the effective velocity of a trapped mesoscopic probe in the close vicinity of the bubble gets modified, along with the spatial diffusion coefficient profile. We observe that even in this case, the Lévy arc sine statistics are not modified. We finally conclude that the arc sine statistics of the fluctuations of an optically confined Brownian probe in a Newtonian fluid remain unaffected with change in amplitude and time constant of an externally added noise – as long as the observation period is much longer than the time constant of the added noise – and in the presence of flow fields provided the flow velocity the probe experiences is constant. (See Supplementary Information).

We can describe the dynamics of our system with the overdamped Langevin equation as:

\[
\dot{x}(t) = -\frac{x(t) - \lambda(t)}{\tau_d} + \sqrt{2D\xi(t)}
\]

(2)

\[
\dot{\lambda}(t) = -\frac{\lambda(t)}{\tau_0} + \sqrt{2A\zeta(t)}
\]

(3)

In Equation (2), \(D\) is the diffusion constant and \(\tau_d = \gamma/k\) is the relaxation time of the harmonic trap, where \(k\) is the trap stiffness and \(\gamma\) is the drag coefficient related by the Stokes-Einstein relation as \(D = k_B T\). In Equation (3), \(\lambda(t)\) is the Ornstein-Uhlenbeck noise that drives the center of the harmonic trap, with a time constant \(\tau_0\), and amplitude proportional to \(A\). \(\zeta(t)\) and \(\xi(t)\) are zero-centered unit variance delta correlated white noise with no mutual correlations, \(\langle \zeta(t)\xi(t') \rangle = 0\). The OU noise has an exponentially decaying correlation \(\langle \lambda(t)\lambda(t') \rangle = A\tau_0 \exp(-|t-t'|/\tau_0)\) and a power spectrum that falls off as \(f^{-\alpha}\), \(\alpha \approx 2\). In presence of the microbubble the system can be modelled with a suitable addition of the flow field \(u_d\), \(\dot{x} \rightarrow \dot{x} - u_d\) and by values of \(\tau_d\) and \(D\) that get scaled as a function of the spatial distance from the microbubble. (see Ref. 23).

The incremental work in each small step can be calculated as \(\Delta W = dW = (\partial U(x(t), x_0(t))/\partial x_0(t)) \ast dx_0(t)\), where \(x_0(t)\) is the parameter varying in the non equilibrium process – the center of the trap in our case. On the other hand the work done in small dissipative steps \(\Delta d\) is due to the viscous drag on the probe embedded in the bath. We use a tightly focused laser beam to form a harmonic trapping potential that varies with time, given by \(U(x(t), \lambda(t)) = k[x(t) - \lambda(t)]/2\), and trap a spherical probe particle (polystyrene) of diameter \(3\) \(\mu\)m, where the trap stiffness is \(k = 19.7 \pm 0.1\) pN/\(\mu\)m. We modulate the trap-center with an Ornstein Uhlenbeck noise given by \(\lambda(t)\) which is generated by Eq 3 with its time constant for exponentially decaying correlation \(\tau_0 = 2.5\) ms, and an amplitude equal to \(A = 0.3 \times (0.6 \times 10^{-6})^2\) m\(^2\)/s. The effective operating temperature of the system is approximatively 588 K. For the next stage of the experiment, we form a microbubble of diameter \(21 \pm 0.6\) \(\mu\)m (see Methods) using another laser \(24\) such that its surface remains \(d = 10\) \(\mu\)m away from the mean position of the center of the probe particle. We sample the displacement of the colloidal particle at a rate of 10 kHz for 100 seconds. We repeat these experiments for varying \(A, \tau_0, d\), and discuss the results in the Supplementary Information. The cumulative work done on the system and work dissipated by the system is given by,

\[
W = \int -k[x(t) - \lambda(t)] \ast d\lambda(t)
\]

(4)

\[
W_d = \int \gamma \lambda(t) \ast dx(t)
\]

(5)

where the period of integration should be chosen such that it is greater than \(\tau_0\). The symbol ‘\(\ast\)’ represents the Stratonovich product for a stochastic variable. (Note that for two stochastic variables \(\alpha_t\) and \(\beta_t\),...
FIG. 1. Work done on the system and work dissipated by the system represented schematically, as obtained from experimental data. (a) The work dissipated by the probe over time plotted in units of $k_B T$ after subtracting the mean value of dissipating current. Values of the Lévy variables $T_+, T_{\text{last}}, T_{\text{max}}$ are displayed for this window in red, green, blue color coding, respectively. (b) The fraction of time the stochastic current $W_d$ (in red) lies above the average $\langle W_d \rangle$ over many cycles. (c) The cumulative work done ($W$) and work dissipated ($W_d$) by the engine in units of $k_B T$ are plotted over time, which are positive and negative, respectively. (d) The probability distributions of the work done and work dissipated per unit sampling time $\tilde{W}$ and $\tilde{W}_d$ plotted on different axial scales. (e) Schematics of the process, where the trap center is modulated by an Ornstein Uhlenbeck process of a constant amplitude to proceed from an equilibrium to a non-equilibrium steady state (ESS to NESS).

FIG. 2. Statistics of work dissipated as obtained from simulation. The results for the three arcsine laws for $T_+$ (in red), $T_{\text{last}}$ (in green) and $T_{\text{max}}$ (in blue) from a simulation of the time series, where the CDF and PDF are plotted in (a) and (b).

the Stratonovich product is given by $\int_0^\tau f(\alpha_t) \ast d\beta_t = \sum_t f(\alpha_t) + f(\alpha_t + \Delta t) \overline{\langle \beta_t + \Delta t - \beta_t \rangle}$, where $t$ runs from 0 to $\tau - \Delta t$.

Results: The position fluctuations for the probe lies in the order of tens of nanometers. The external source of colored noise, whose diffusion constant ($A = 0.3 \times (0.6 \times 10^{-6})^2 m^2/s$) is of the order of the diffusion constant of water, increases the variance of the probe fluctuations and drives it out of equilibrium to an elevated temperature of 588 K. However the temperature of the water (environment), surrounding our driven micro-colloidal system does not increase more than 1K, as energy dissipated by the viscous forces as well as heat absorbed from the laser [25] is negligible compared to the work done on the system. We show the results from a section of the experimental data as an example in Fig. 1. Note that we
FIG. 3. Action of a microbubble in close vicinity of the optically trapped probe. (a) Schematic of the experimental probe particle-microbubble system. The microbubble is grown on a pre-existing absorbing pattern/trail in the sample chamber (see Methods). The different dimensions of the bubble and . (b) Histogram of the position fluctuation – clearly it develops a skew in the presence of the flow field generated by the microbubble.

FIG. 4. Arcsine laws tabulated for all the cases. The work dissipated by the stochastic engine as well as the work done in our experiments are used to plot the three arcsine laws for $T_+$, $T_{last}$ and $T_{max}$ in red, green and blue colours, respectively.

consider the work done on the system as positive, while the work dissipated by the system is negative. Fig. (a) shows the fluctuating current for dissipated work after the average fluctuation for all the segments is subtracted from the time series. Further, Fig. (b), shows that the total time the time series of $W_d$ lies above the average current $\langle W_d \rangle$, is 0.02 s. Similarly, we calculate from this section of data, that the last instant of time when the stochastic current was positive is 0.81 s, and the time instant when it reached the global maximum within this section of the data is 0.04 s. Although the work done and dissipated are stochastic in short times, over a long time, they accumulate cumulatively. We plot the histogram of the work done (dissipated) in small steps $\tilde{W}_d$ in Fig. (d). The work on the micro-system is in the order of $k_B T$ – out of which approximately 0.1% is dissipated into the fluid due to friction. We represent the process schematically in Fig. (e). The stochastic currents of work done (dissipated) on (by) the system follow the properties of a Wiener process – they start at zero, their small increments are independent of the past (Markov) and are Gaussian with its variance increasing linearly. The time series of the position variable has embedded correlation because of the correlated noise, and due to the effect of the trap – so that those variables do not follow the Wiener properties. Next, we simulate the stochastic trajectories of a probe perturbed by an OU process and verify all the results obtained in the experiment. The cu-
mutative density function work dissipated (done) by (on) such a system follows an arcsine distribution (for all the three variables), which we show in Fig. 2(a). Similarly, in Fig. 2(b), we demonstrate an example that the PDF of the stochastic current follows $\frac{1}{\pi \sqrt{T(1-T)}}$.

When we perform the experiment in the presence of a microbubble as we display in Fig. 3(a), an active flow field is generated, which constricts and skews the motion of the probe in one direction – which we demonstrate in Fig. 3(b), where the Gaussian nature of the position probability distribution function is clearly observed to be skewed in the presence of the flow generated by the bubble. Note that the work done depends on the probe’s velocity through the medium, which now has an additive term due to the flow induced by the microbubble. However, since the probe’s fluctuation is minimal compared to its distance from the surface of the microbubble, we can assume the strength of the flow field to unchanged, so that the arcsine nature of the statistics also remains undisturbed.

While obtaining the statistics, we divide the data into segments of 1 s ($10^4$ points) each, which is much greater than the time constant of the underlying process, $\tau_d (\approx 2.5 \text{ ms})$. Note that this is important towards obtaining robust statistics. When more data points are sampled, a better fit is obtained for the arcsine laws. In Fig. 3 we tabulate all the experimental plots and show that the CDF of $T_+$, $T_{last}$, and $T_{max}$ obtained from both the work done and work dissipated by the system follow Lévy arcsine statistics.

**Conclusion:** We unambiguously demonstrate that the stochastic currents associated with a Markov process in a non-equilibrium steady-state system in the presence of viscous forces obey Lévy arcsine statistics, leading to experimentally measurable variables which also demonstrate the same statistics. Thus, we observe that – even in the presence of colored noise, temporally correlated perturbations, or active flow-fields – the statistical characteristics of the work exchange between the system and its surroundings are not modified – indeed they all follow arcsine laws. Our method can be extended to other systems in non-equilibrium, or even to study transient phenomena in physics. Most importantly, any deviations from arcsin laws can indicate non-Markovian behavior or anomalous diffusion in the underlying processes that generate the stochastic current, thus providing useful insight into the system dynamics before probing it in detail.

**Methods**

**Particle trapping and manipulation:** We perform all experiments on a double-distilled aqueous dispersion of spherical polystyrene particles (Sigma-Aldrich LB30) having a diameter 3 µm. We build a custom sample chamber of thickness 100 µm with double-sided sticky tape between two coverslips and mount the sample chamber on a motorized stage. A Gaussian beam (1064 nm) tightly focused with high numerical aperture oil immersion objective (100x, NA=1.3) of a standard oil immersion objective an inverted microscope (Olympus IX71) traps the particle at a height 15 µm from the lower surface of the sample chamber, to mitigate surface forces. The laser passes through an acoustooptic modulator (BRIMROSE-AOM), and the first-order beam traps and modulates the particle perpendicular to the beam’s direction with input noise generated through the Ornstein-Uhlenbeck process. We focus a second co-propagating beam of wavelength 785 nm and measure the backscattered intensity by position-sensitive photodetectors (PDA100A2) to sample the one-directional trajectory of the probe at a spatio-temporal resolution of 1 nm - 10 kHz. Additionally, we use the autocorrelation of the time series of a trapped particle and the noise to calibrate the fluctuation of the probe from volts (measured by the photodiode) to nm.

**Generation of microbubble:** For the experiments involving the microbubble, we employ a coverslip patterned by a polyoxometalate material [24], absorbing at 1064 nm as the bottom surface of the sample chamber while keeping everything else unchanged. We use a second laser operating at a 1064 nm wavelength to generate a microbubble of diameter 21 µm – the remains constant for unchanging laser power [24] throughout the time-scale of the experiment. We trap the particle at the same height as the radius of the bubble and calibrate the length scale of our setup with the company manufactured integrated software, which we also verify from the size of the probe.

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I. SUPPLEMENTARY INFORMATION

A. Calculating effective temperature

When white noise is added to a system, the effective temperature can be easily calculated in by adding the variance of the noise to the data; however this assumption breaks down when the added noise has a finite temporal correlation, given by, $\lambda(t)\lambda(t') = \sigma^2 T(t-t')$. The effective temperature is related by $\frac{2k_B T}{\gamma} + \frac{\sigma^2}{\gamma^2} = \frac{2k_B T_{\text{eff}}}{\gamma}$. However, the equality holds only for the case of delta correlated noise. We can calculate the effective temperatures using the relation $\text{Var}(x') = \frac{k_B T_{\text{add}}}{k}$ such that we get $\frac{2\text{Var}(x)}{\tau_d} + \frac{\text{Var}(\lambda)}{\tau_0} = \frac{2k_B T_{\text{add}}}{\tau_d}$, where $T_{\text{add}}$ is the additive temperature. The effective temperature $(T_{\text{eff}})$ is calculated directly from the variance of the position fluctuation. In Fig. 1 we show that the effective temperature as well as the additive temperatures increase with increasing amplitude of the added Ornstein-Uhlenbeck noise. In all the cases we note that $T_{\text{add}} > T_{\text{add}} > T$. $T_{\text{eff}}$ will be equal to $T_{\text{add}}$ iff, the added noise is temporally delta correlated [27].

![FIG. 1. The additive temperatures ($T_{\text{add}}$) and the effective temperatures ($T_{\text{eff}}$) for the non equilibrium steady state (NESS) system plotted along with the standard errors for varying amplitude of the input OU noise.](image-url)
B. Accuracy

To obtain the arcsine statistics, we must integrate much more data points than those in a single decay cycle of the temporally correlated modulation. Naturally, using a larger number of data points improve the accuracy of the statistics. We define error by the absolute deviation of the obtained statistic from the arcsine distribution, divided by the number of points used. Fig. 2 shows in the logarithmic plot that the error decreases in a power law with the increasing number of data points, where the exponent $\approx -0.9$.

C. Ornstein Uhlenbeck noise.

Fig. 3 shows that the OU process generates a colored noise that has an exponentially decaying correlation and a power spectrum that fits a power law. We can use the autocorrelation property of the added noise to calibrate its amplitude. In Fig. 3(a) we plot the autocorrelation for $\lambda(t)$ where $\tau_0 = 25$ ms. In Fig. 3(b), we plot the power spectrum of the noise in a logarithmic plot. By fitting the data points with $1/f^\alpha$, we get $\alpha \approx 1.8$, which confirms that it is colored noise.

D. Mean Square displacement

The main criterion for any variable to follow a Wiener process (and therefore the Lévy arcsine statistics) is that its variance should increase linearly with time – which implies that the MSD of the variable should increase linearly with increasing time lags. To demonstrate this, we plot the MSD of the variable in Fig. 4 which is required to characterize the work done by the external modulation as well as the work dissipated for both the cases where the microbubble is present and absent. By fitting the MSD with a power-law, we observe the exponent is approximately equal to 1 for all the cases, which is a necessary condition to be followed.

E. Variation of amplitude of the added noise

All our results are demonstrated for the case where the amplitude of the added noise is given by $A = 0.3 \times (0.6 \times 10^{-6})^2 m^2/s$. Changing the amplitude of the input noise changes the heat dissipated by the system and the magnitude of the work done associated with the process. With increasing amplitude, the production rate of entropy also increases linearly, so does the system’s effective temperature. However, the arcsine nature of the concerned variables, $W$ and $W_d$ remains intact. In Fig. 5(a) & (b) and (d) & (e), we plot the arcsine statistics for two different amplitudes $A_1 = 0.1 \times (0.6 \times 10^{-6})^2 m^2/s$ and $A_2 = 0.2 \times (0.6 \times 10^{-6})^2 m^2/s$. In Fig. 5(c) & (f), we show that the effective spread of $x-\lambda$ is in tens of nanometers, which increases with increasing amplitude of the added noise. It has been demonstrated in Ref [23] that increase in the entropy production rate or the work dissipated is not infinite but has an upper-bound.

F. Change of the distance from microbubble

The ambient flow field changes with changing distance from the microbubble – as a result, the effective velocity of our trapped colloidal particle also gets modified. The flow field skews the symmetry of the position distribution of the trapped particle. In Ref [23], we observe how the flow-field and the current increase when we trap the particle at the close proximity of the bubble. However, in Fig. 6 we show that the arcsine laws for $W_d$ do not get modified when the distance changes because the properties of the Wiener process are still applicable. In our case, we have reported calculations for the center of the probe $10 \mu m$ away from the surface in the main manuscript. In Fig. 6(a) and (b), we show results for distances of $5 \mu m$ and $15 \mu m$, respectively. In the presence of the microbubble, the motion of the probe gets spatially constrained. Further away from the bubble, the dissipated work (see Fig. 6(c)), as well as entropy production, increases to an upper bound, which is predicted by thermodynamic uncertainty relations.
FIG. 3. Characteristics of the added Ornstein-Unhelbeck noise. (a) The autocorrelation of the added noise which we fit with the theoretical exponential decay function with amplitude $A\tau_0$ and decay constant $\tau_0$. (b) The logarithmic plot of the power spectral density of the noise in the Fourier domain. The amplitude falls off as $1/f^\alpha$, where $\alpha$ is an exponent denoting the characteristics of the noise.

FIG. 4. The Mean squared displacement (MSD) plotted for short time scales. (a) and (b) show the MSD of the $W_d$ and $W$ variables, obtained from the micro-engine in absence of the bubble. (c) and (d) show the MSD of $W_d$ and $W$ in presence of the bubble’s flow field. The MSD increases linearly with time as expected for a Wiener variable.
FIG. 5. Change in the amplitude of the input OU noise (a) and (b) show all the three Arcsine laws for $T_+$, $T_{last}$ and $T_{max}$ in red, green and blue respectively for the case when input amplitude is $A_1 = 1 \times (0.6 \times 10^{-6})^2 m^2/s$. In (c) the distribution of $x$ and $\lambda$ is plotted. We repeat the same for (d),(e),(f) for the case, $A_2 = 0.2 \times (0.6 \times 10^{-6})^2 m^2/s$.

FIG. 6. Change in the distance of the probe from the microbubble: (a) and (b) the three Arcsine laws for $T_+$, $T_{last}$ and $T_{max}$ in red, green and blue where distance from the microbubble is 5$\mu$m and 15$\mu$m, respectively. (c) The variation in the dissipated work for the two distances 5$\mu$m and 15$\mu$m are in red and blue respectively.