Experimental verification of a modified fluctuation-dissipation theorem for a Brownian particle in a non-equilibrium steady state

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Abstract. We verify experimentally a modified fluctuation-dissipation relation for displacement fluctuations of a micron-sized silica particle immersed in water in a non-equilibrium steady state (NESS) with non-vanishing probability current. A NESS is implemented by means of a toroidal optical trap created by a rotating laser beam with intensity modulation which exerts a constant non-conservative force on the particle against a sinusoidal potential on a circle. We measure the autocorrelation function of an observable related to the angular position of the particle, the corresponding integrated response function due to a small perturbation of the amplitude of the periodic potential, and a corrective term given by the constant probability current. We find that the correlation minus the corrective term times the inverse temperature of the surrounding water is equal to the integrated response. The results can be interpreted as an equilibrium-like fluctuation-dissipation relation in the Lagrangian frame moving at the mean local velocity of the particle determined by the probability current.

1 Introduction

The validity of the fluctuation-dissipation theorem (FDT) in systems out of thermal equilibrium has been the subject of intensive study during the last years. We recall that for systems in equilibrium with a thermal bath at temperature T, the FDT establishes a simple relation between the 2-time correlation function $C(t-s)$ of a given observable and the linear response function $R(t-s)$ of this observable to a weak external perturbation

$$\partial_s C(t-s) = k_B T R(t-s). \tag{1}$$

However, Eq. (1) is not necessarily fulfilled out of equilibrium and violations are observed in a variety of systems such as glassy systems [1,2], granular matter [3] and biophysical systems [4].

The lack of a general framework describing FD relations in non-equilibrium situations has motivated some theoretical works devoted to a comprehensive study of this issue in simple stochastic systems [5,6,7,8,9,10]. In this spirit, a modified fluctuation-dissipation theorem (MFDT) has been recently found for non-equilibrium steady systems with few degrees of freedom evolving according to a Langevin equation possibly including non-conservative forces [10]. In particular, this MFDT holds for the overdamped motion of a particle on a circle ($0 \leq \theta < 2\pi$) in the presence of a periodic potential $U(\theta) = U(\theta + 2\pi)$ and a non-conservative force $F(\theta) = F(\theta + 2\pi)$

$$\dot{\theta} = -\partial_\theta U(\theta) + F(\theta) + \zeta, \tag{2}$$

where $\int_0^{2\pi} F(\theta) d\theta \neq 0$, ζ is a white noise term of mean $\langle \zeta(t) \rangle = 0$ and covariance $\langle \zeta(t)\zeta(s) \rangle = 2D\delta(t-s)$, with D the diffusivity. The non-equilibrium steady state (NESS) associated to Eq. (2) is described by a constant non-vanishing probability current j along the circle and by an invariant probability density function $\rho_0(\theta)$ that allow to define a local mean velocity $v_0(\theta) = j/\rho_0(\theta)$. For a stochastic system in NESS evolving according to Eq. (2), the MFDT reads

$$\partial_s C(t-s) - b(t-s) = k_B T R(t-s), \tag{3}$$

where the 2-time correlation of a given observable $O(\theta)$ is defined by

$$C(t-s) = \langle O(\theta(t))O(\theta(s))\rangle_0, \tag{4}$$

and the linear response function to a time dependent perturbation $h(t)$ is given by the functional derivative

$$R(t-s) = \left.\frac{\delta}{\delta h(s)}\right|_{h=0} \langle O(\theta(t))\rangle_h. \tag{5}$$

In Eq. (4) $\langle ...\rangle_0$ stands for an ensemble average over NESS distributed according to ρ_0 whereas in Eq. (5), $\langle ...\rangle_h$ denotes the average over the time-dependent perturbed states. In the MFDT (3), the correlation $b(t-s)$ is given by

$$b(t-s) = \Theta(t-s)\langle O(\theta(t))v_0(\theta(s))\partial_\theta O(\theta(s))\rangle_0, \tag{6}$$

where Θ is the Heaviside step function. This new term plays the role of a corrective term of $C(t-s)$ in the usual fluctuation-dissipation relation (1) taking into account the effect of the probability current.

In the present work, we firstly show the experimental results describing the NESS attained by a micron-sized particle moving in a toroidal optical trap whose dynamics is claimed to be modeled by the Langevin dynamics of Eq. (2), namely the constant probability current, the invariant density and the local mean velocity. Further, by computing the correlation function $C(t)$, the integrated corrective term $B(t) \equiv \int_0^t b(t-s)ds$ and by directly measuring the integrated response function $\chi(t) = \int_0^t R(t-s)ds$ for a proper choice of the observable $O(\theta)$, we verify that Eq. (3), in its integral form,

$$C(0) - C(t) - B(t) = k_B T\chi(t), \tag{7}$$

is satisfied by this system within experimental accuracy. Accordingly, we follow the Lagrangian-frame interpretation discussed in [10] which implies that an equilibrium-like FD relation for the particle motion (similar in form to Eq. (1)) can be restored in the reference frame moving with the local mean velocity v_0.

2 Experimental description

In order to create experimentally a system with the Langevin dynamics of Eq. (2) in NESS, we prepare a small sample cell containing a extremely small volume fraction of spherical silica particles of radius $r = 1\ \mu$m diluted in ultrapure water. The experiment is performed at a room temperature $T = 20.0 \pm 0.5°$ C at which the dynamic viscosity of water is $\eta = (1.002 \mp 0.010) \times 10^{-3}$ Pa s. The sample cell is placed in an optical tweezers system with the purpose of trapping a single particle and isolate it from the rest during the experiment. Once the particle under study is far enough from all sources of perturbations, it is trapped by a toroidal optical trap. This kind of trap consists on a Nd :YAG diode pumped solid state laser beam (Laser Quantum, $\lambda = 1064$ nm) focused by a microscope objective (63×, NA = 1.4) whose waist represents the smallest circle of the torus scanning a larger circle of radius $a = 4.12\ \mu$m in the horizontal plane at a rotation frequency of 200 Hz. The rotation of the focused beam is accomplished by means of two coupled acousto-optic deflectors working with a $\pi/2$ phase shift. The toroidal trap is created 10 μm above the inner bottom surface of the sample cell where hydrodynamic boundary-coupling effects on the particle motion are negligible. At a rotation frequency of 200 Hz, the laser beam is not able to hold the particle but drags it regularly a small distance when passing through it [11]. The diffusive motion of the particle along the radial direction during the absence of the beam is small enough so that it remains confined in the circle of radius a. Therefore the angular position of the particle θ (measured modulo 2π) is the only relevant degree of freedom of the dynamics. In addition, the laser power is sinusoidally modulated around 30 mW with an amplitude of 7% of the mean power, synchronously with the deflection of the beam at 200 Hz in such a way that a static sinusoidal intensity profile is created along the circle. This trapping situation acts a constant non-conservative force f associated to the mean kick which pushes the particle against a periodic sinusoidal potential $U(\theta) = A\sin\theta$ due to the periodic intensity profile. The

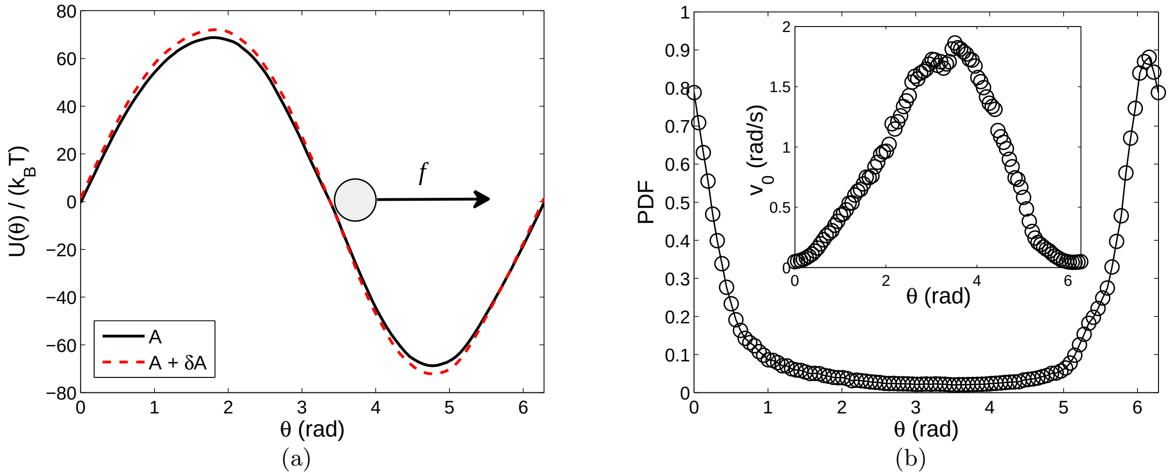

Fig. 1. (a) Profile of the periodic potential associated to the laser intensity modulation of the toroidal trap. The arrow indicates the direction of the non-conservative force f. (b) Invariant probability density function of the angulat position of the particle in NESS. Inset : Local mean velocity of the particle.

calibration and the determination of the values of the parameters f and A is accomplished by means of the method described in [12]. We obtain $f = 6.60 \times 10^{-14}$ N and $A = 68.8 k_B T = 2.78 \times 10^{-19}$ J. The experimental potential profile is shown in Fig. 1(a) (black solid line). In this way, the time evolution of θ is claimed to follow the Langevin dynamics of Eq. (2) [12] with $F = f/(6\pi\eta ra) = 0.85$ rad s^{-1}, $B = A/(6\pi\eta ra^2) = 0.87$ rad s^{-1}, and $D = k_B T/(6\pi\eta ra^2) = 1.26 \times 10^{-2}$ rad^2 s^{-1}. Images of the intensity contrast of the particle on the focal plane $x - y$ determined by the circle of radius a are recorded with a resolution of 160×130 pixels at a sampling rate of 150 frames per second for detection.

Tracking of the particle barycenter $(x(t), y(t))$ is achieved with an accuracy of a few nanometers from which the angular position of the particle $\theta(t)$ with respect to the trap center is found. We obtained 200 time series $\{\theta(t)\}$ of duration 66.67 s with different initial conditions $\{\theta_0 = \theta(0)\}$ sampled every 5 minutes for the determination of the stationary quantities C and B of Eq. (3). Additionally, 500 times series of duration 100 s were specially devoted for the determination of χ. In this case during each interval of 100 s we apply a Heaviside step-like perturbation to the amplitude of the potential $A(t) = A + \delta A[\Theta(t - t_0) - \Theta(t - (t_0 + T))]$ with $T = 33.33$ s the duration of the perturbation, $0 < t_0 < 66.67$ s the instant at which it is switched on, and δA the intensity of the perturbation. This is accomplish by suddenly switching the laser power modulation from 7% to 7.35% of the mean power (30 mW). By keeping constant the mean power during the switch we assure that the value of f remains also constant. The experimental shape of the perturbed potential of amplitude $A + \delta A$ is shown in Fig. 1(a) (red dashed line). In this way, we extracted 500 *perturbed* trajectories $\{\theta(t)\}_{\delta A}$ of duration $T = 33.33$ s. This duration is long enough to assure that after switching off the perturbation the system actually has attained a NESS before the beginning of the next step-like perturbation. We checked that the value of δA obtained by means of this procedure ($\delta A = 0.05A$) is small enough to remain within the linear response regime for time lags $0 \leq t \leq 3.5$ s. For 3.5 s $< t$ nonlinearities become important. Consequently, the response function is only measured for the first 3.5 s of the perturbed trajectories where linear response regime holds.

3 Results

We check that after a short transient, the particle motion attains a NESS described by a non-vanishing constant probability current j along the torus in the orientation of the laser beam rotation and by an

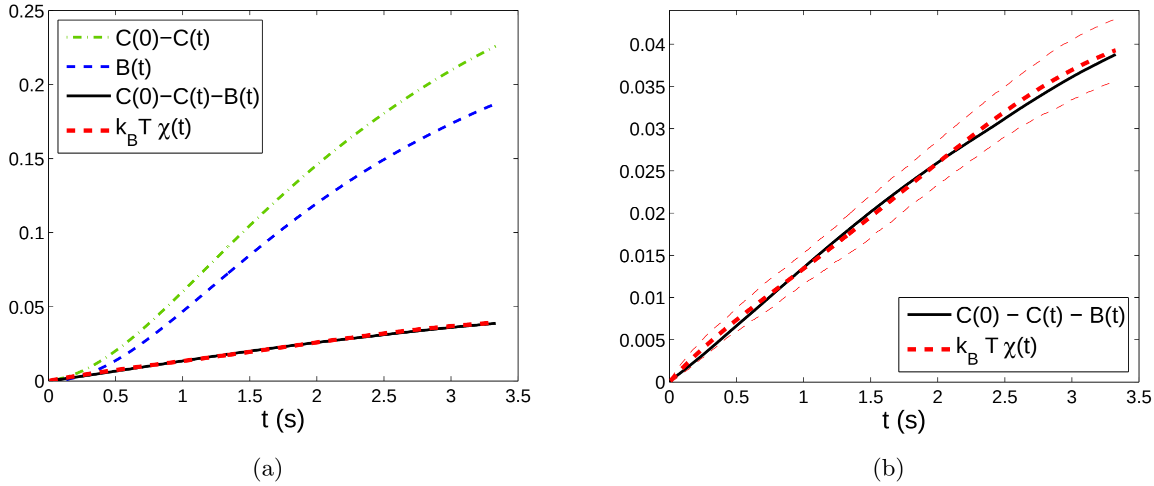

Fig. 2. a) Comparison between the different terms needed to verify Eq. (7), as functions on the time lag t. b) Expanded view of the comparison between $C(0) - C(t) - B(t)$ and $k_B T\chi(t)$ in Fig 2(a). The thin red dashed lines represent the error bars of the measurement of the integrated response.

invariant probability density function $\rho_0(\theta)$. The probability current is related to the global mean velocity of the particle by the expression $j = \langle\dot{\theta}\rangle_0/(2\pi)$. Hence, we determine the value of $\langle\dot{\theta}\rangle_0$ from the slope of the linear fit of the 200 trajectories (not taken modulo 2π) leading to $j = 3.76 \times 10^{-2}$ s^{-1}. The invariant density shown in Fig. 1(b) is computed from the histogram of each time series $\{\theta(t)\}$ averaged over the 200 different initial conditions. Note that in the corresponding equilibrium situation ($f = 0$) the probability maximum would be located at the minimum of $U(\theta)$ ($\theta = 3\pi/2$). However, in NESS the presence of the non-conservative force $f > 0$ shifts the maximun of $\rho_0(\theta)$ in its own direction. The position of the maximum depends on the value of f. In order to enhance the stochastic nature of the dyanmics, by chosing $F < B$ we purposely created a situation in which the particle stays long time around the maximum $\theta \approx 6$ whereas the rest of the circle is rarely visited during the mean rotation. In Fig. 1(b) we also show the local mean velocity $v_0(\theta)$ of the particle associated to the probability current through the torus. The local mean velocity is given by $v_0(\theta) = j/\rho_0(\theta)$ and its value at a given position θ represents the average of the instantaneous velocity $\dot{\theta}$ restricted to the ensemble of trajectories passing through θ.

With the purpose of determining correctly the different terms involved in Eq. (3), the observable $O(\theta)$ must be chosen consistently in both sides of such relation. The change of the potential $U(\theta) \to U(\theta) + \delta A \sin\theta$ due the application of the Heaviside step perturbation of its amplitude implies that $O(\theta) = \sin\theta$ is the observable that must be studied with $-\delta A$ as its conjugate variable. Hence, we compute the correlation function $C(t)$, the corrective term $B(t)$ (with $\partial_\theta O(\theta) = \cos\theta$ and the experimental curve $v_0(\theta)$ shown in Fig. 1(b)) and the integrated response $\chi(t)$ for this observable as a function of the time lag t.

The determination of $C(t)$ and $B(t)$ is straightforward according to Eqs. (4) and Eqs. (6). The stationarity of the system allows to perform an average over the time origin in addition to the ensemble average $\langle ...\rangle_0$ over the 200 different time series devoted to this purpose, which increases enormously the statistics. The dependence of the correlation term $C(0) - C(t)$ and the term $B(t)$ on the time lag t is shown in Fig. 2(a) in dotted-dashed green and blue dashed lines, respectively.

On the other hand, some subtleties must be taken into account for the determination of χ in the current non-equilibrium situation. From the linear response function defined in Eq. (5), the integrated response χ is given by

$$\chi(t) = \frac{\langle O(\theta(t))\rangle_{\delta A} - \langle O(\theta(t))\rangle_0}{-\delta A}. \tag{8}$$

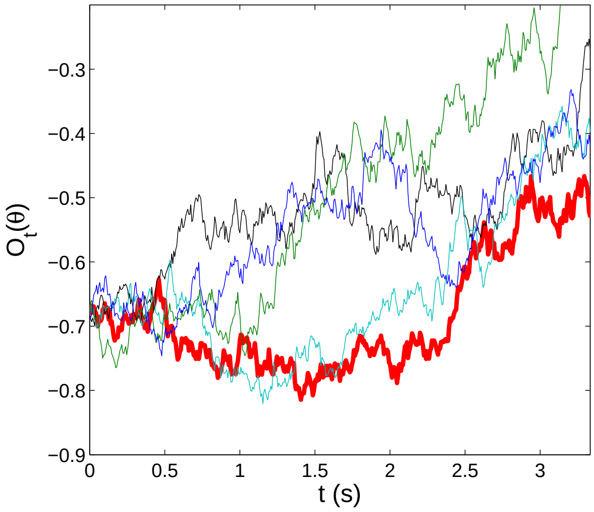

Fig. 3. Example of trajectories used to compute the intergrated response. For a given perturbed trajectory (thick red line) a set of unperturbed trajectories whose initial value is equal to that of the perturbed one at $t = 0$ (thin lines) is found.

In Eq. (8) the value $t = 0$ corresponds to instant when the perturbation of the potential amplitude δA is switched on. The numerator represents the mean deviation of the perturbed trajectories with respect to the unperturbed ones and its expression is exact only for a continuous sample of trajectories. However, due to the finite number of trajectories available in practice, some care is needed in order to compute it correctly. For a given perturbed trajectory $\theta(t)_{\delta A}$ we look for an unperturbed one $\theta(t)$ among the 200 time series $\{\theta(t)\}$ starting at a time t^* such that $O(\theta(t^*)) = O(\theta(0)_{\delta A})$. In order to improve the statistics, for each $\theta(t)_{\delta A}$ among the 500 perturbed time series $\{\theta(t)\}_{\delta A}$ we seek as many unperturbed trajectories as possible satisfying the initial condition for t^*, as shown in Fig. 3. The unperturbed trajectories found in this way allow us to define a subensemble over which the average $\langle O(\theta(t))\rangle_0$ in Eq. (8) is computed at a given t. The average $\langle O(\theta(t))\rangle_{\delta A}$ is simply computed over the 500 perturbed time series. In Fig. 2(a) we show in thick dashed red line the dependence of the integrated response on t.

The comparison between the different terms needed to verify Eq. (7) is shown in Fig. 2(a), for the time lag interval $0 < t < 3.5$ s where the linear response regime is valid. As expected, the usual FD relation (1) is strongly violated in this NESS with the correlation term $C(0) - C(t)$ being one order of magnitude larger than the response term $k_B T\chi(t)$. However, when we take into account the term $B(t)$ associated to the probability current as a correction of $C(0) - C(t)$, the term $C(0) - C(t) - B(t)$ shown in solid black line in Fig. 2(a), becomes equal to $k_B T\chi(t)$. In Fig. 2(b) we show an expanded view of the of the curves $C(0) - C(t) - B(t)$ and $k_B T\chi(t)$. We observe that within experimental accuracy, the agreement between both terms is quite good, verifying the integral form of the MFD relation given by Eq. (7). The error bars of the integrated response curve at each time lag t are obtained from the standard deviation of the subensemble of unperturbed trajectories found for each perturbed trajectory, like the ones shown in thin solid lines in Fig. 3. As shown in [10], a direct interpretation of the verification of this MFDT for the fluctuations of the angular position of the silica particle in NESS can be rendered in the Lagrangian frame moving with the local mean velocity shown in Fig. 1(b) along the toroidal trap. In this frame, the time-independent observable $O(\theta) = \sin\theta$ is replaced by an explicitly time-dependent one $O(t, \theta)$ evolving according to an advection equation with velocity field $v_0(\theta)$ for which the MFD relation reads

$$\partial_s C(t, s) = k_B T R(t, s). \tag{9}$$

Thus, an FD relation similar in form to the equilibrium one (Eq. (1)), can be restored in the corresponding Lagrangian frame of this experiemental system.

4 Conclusion

We have verified experimentally a modified fluctuation-dissipation relation describing the dyanmics of a system with one degree of freedom in NESS, namely a Brownian particle moving in a toroidal optical optical trap. We point out that the experimental results reported here represent an alternative approach of a fluctuation-dissipation relation extended to non-equilibrium stationary situations from the one described in [9] for velocity fluctuations relative to the local mean velocity for a similar experimental system. The approach followed in our work relies in an observable related to the particle position and besides quantifying the extent of the violation of the usual FDT by means of the term B, a transparent interpretation of the violation is possible.

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