Work Fluctuation Theorems for Harmonic Oscillators

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The work fluctuations of an oscillator in contact with a thermostat and driven out of equilibrium by an external force are studied experimentally and theoretically within the context of fluctuation theorems. The oscillator dynamics is modeled by a second order Langevin equation. Both the transient and stationary state fluctuation theorems hold and the finite time corrections are very different from those of a first order Langevin equation. The periodic forcing of the oscillator is also studied; it presents new and unexpected short time convergences. Analytical expressions are given in all cases.

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In this Letter, we investigate, within the context of the *fluctuation theorems* (FTs), the work fluctuations of a harmonic oscillator in contact with a thermostat and driven out of equilibrium by an external force. First found in dynamical systems [1,2] and later extended to stochastic systems [3–6], these conventional FTs give a relation between the probabilities to observe a positive value of the (time averaged) "entropy production rate" and a negative one. This relation is of the form $P(\sigma)/P(-\sigma) = \exp[\sigma\tau]$, where σ and $-\sigma$ are equal but opposite values for the entropy production rate, $P(\sigma)$ and $P(-\sigma)$ give their probabilities, and τ is the length of the interval over which σ is measured. In these systems, the above mentioned FT is derived for a mathematical quantity σ, which has a form similar to that of the entropy production rate in irreversible thermodynamics [7].

The proof of FTs is based on a certain number of hypotheses; experimenting on a real device is useful not only to check those hypotheses, but also to observe whether the predicted effects are observable or remain only a theoretical tool. There are not many experimental tests of FTs. Some of them are performed in dynamical systems [8] in which the interpretation of the results is very difficult. Other experiments are performed on stochastic systems, one on a Brownian particle in a moving optical trap [9] and another on electrical circuits driven out of equilibrium by injecting in it a small current [10]. The last two systems are described by first order Langevin equations and the results agree with the predictions of Refs. [5,6]. Systems described by second order Langevin equations have been studied [3], but to our knowledge no predictions on the finite time corrections, as the ones from [5,6] for the power injected into the system are available. The test using a harmonic oscillator is particularly important because the harmonic oscillator is the basis of many physical processes. Indeed the general predictions of FTs are valid only for $\tau \to \infty$ and the corrections for finite τ have been computed only for a first order Langevin dynamics.

In the present Letter, we address several important questions. We investigate first the *transient fluctuation theorem* (TFT) of the total external work done on the system in the transient state, i.e., considering a time interval of duration τ which starts immediately after the external force has been applied to the oscillator. We then analyze the *stationary state fluctuation theorem* (SSFT), which concerns fluctuations in the stationary state, i.e., in intervals of duration τ starting at a time long after the external force has been applied. We also study a new case of stationary behavior obtained when the system is driven periodically in time [11]. In this case, which is actually a very important one, no theoretical prediction is available. We show that the finite time corrections for SSFT are already very complex in both these rather simple situations.

To test the FT we measure the out-of-equilibrium fluctuations of a harmonic oscillator whose damping is mainly produced by the viscosity of the surrounding fluid, which acts as a thermal bath of temperature T. We recall here only the main features of the experimental setup, more details can be found in Refs. [12,13]. The oscillator is a torsion pendulum composed of a brass wire and a glass mirror glued in the middle of this wire. It is enclosed in a cell filled by a water-glycerol solution at 60% concentration. The motion of this pendulum can be described by a second order Langevin equation:

$$I_{\text{eff}} \frac{d^2\theta}{dt^2} + \nu \frac{d\theta}{dt} + C\theta = M + \sqrt{2\nu k_B T}\,\eta, \tag{1}$$

where θ is the angular displacement of the pendulum, I_{eff} is the total moment of inertia of the displaced masses, ν is the oscillator viscous damping, C is the elastic torsional stiffness of the wire, M is the external torque, k_B the Boltzmann constant, and η the noise, delta correlated in time. In our system the measured parameters are the stiffness $C = 4.5 \times 10^{-4}$ N m rad^{-1}, the resonant frequency $f_o = \sqrt{C/I_{\text{eff}}}/(2\pi) = 217$ Hz, and the relaxation time $\tau_\alpha = 2I_{\text{eff}}/\nu = 9.5$ ms. The external torque M is applied by means of a tiny electric current J flowing in a coil glued behind the mirror. The coil is inside a static magnetic field, hence $M \propto J$. The measurement of θ is performed by a differential interferometer, which uses two laser beams impinging on the pendulum mirror [12,13]. The measurement noise is 2 orders of magnitude smaller than the thermal fluctuations of the pendulum. $\theta(t)$ is acquired with a resolution of 24 bits at a sampling rate of 8192 Hz, which is about 40 times f_o. The calibration accuracy of the apparatus, tested at $M = 0$ using the fluctuation dissipation theorem, is better than 3% (see [13]).

To study SSFT and TFT we apply to the oscillator a time dependent torque $M(t)$ as depicted in Fig. 1(a), and we consider the work W_τ done by $M(t)$ over a time τ:

$$W_\tau = \frac{1}{k_B T}\int_{t_i}^{t_i+\tau}[M(t) - M(t_i)]\frac{d\theta}{dt}dt. \qquad (2)$$

The TFT implies that $t_i = 0$ whereas $t_i \geq 3\tau_\alpha$ for SSFT. As a second choice for $M(t)$, the linear ramp with a rising time τ_r is replaced by a sinusoidal forcing; this leads to a new form of stationary state which has never been considered in the context of FT. We examine first the linear forcing $M(t) = M_o t/\tau_r$ [Fig. 1(a)], with $M_o =$ 10.4 pN.m and $\tau_r = 0.1$ $s = 10.7$ τ_α. The response of the oscillator to this excitation is comparable to the thermal noise amplitude, as can be seen in Fig. 1(b) where $\theta(t)$ is plotted during the same time interval of Fig. 1(a). Because of thermal noise the power W_τ injected into the system [Eq. (2)] is itself a strongly fluctuating quantity.

We consider first the TFT. The probability density functions (PDFs) $P(W_\tau)$ of W_τ are plotted in Fig. 2(a) for different values of τ. We see that the PDFs are Gaussian for all τ and the mean value of W_τ is a few $k_B T$. We also notice that the probability of having negative values of W is rather high for the small τ. The function

$$S(W_\tau) \equiv \ln\left[\frac{P(W_\tau)}{P(-W_\tau)}\right] \qquad (3)$$

is plotted in Fig. 2(b). It is a linear function of W_τ for any τ, that is $S(W_\tau) = \Sigma(\tau) W_\tau$. Within experimental error, we measure the slope $\Sigma(\tau) = 1$. Thus,for our harmonic oscillator,the TFT is verified for any time τ. This was expected [5,7], and we give a derivation of this generic result for a second order Langevin dynamics at the end of the Letter.

We now consider the SSFT with $t_i \geq 3\tau_\alpha$ in Eq. (2). We find that the PDFs of W_τ, plotted in Fig. 3(a), are Gaussian with many negative values of W_τ for short τ. The function $S(W_\tau)$, plotted in Fig. 3(b), is still a linear function of W_τ, but, in contrast to TFT, the slope $\Sigma(\tau)$ depends on τ. In Fig. 3(c) the measured values of $\Sigma(\tau)$ are plotted as a

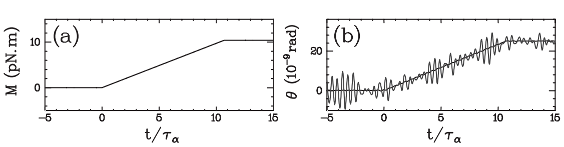

FIG. 1. (a) Typical driving torque applied to the oscillator. (b) Response of the oscillator to the external torque (gray line). The dark line represents the mean response $\bar{\theta}(t)$ to the applied torque $M(t)$.

function of τ. The function $\Sigma(\tau) \to 1$ for $\tau \gg \tau_\alpha$. Thus SSFT is verified only for large τ. The finite time corrections of SSFT, which present oscillations whose frequency is close to f_0, agree quite well with the theoretical prediction computed for a second order Langevin dynamics that we will discuss at the end of the Letter, see Eq. (10). We stress that the finite time correction is in this case very different from that computed in Refs. [5,6] for the first order Langevin equation.

The results of Figs. 2 and 3, have been checked for several M_o/τ_r without noticing any difference. The errors bars in the figure are within the size of the symbols, and they come only from the calibration errors of the harmonic oscillator parameters, and statistics of realizations (typically 5×10^5 cycles have been used).

Finally we want to briefly describe the results of the periodic forcing. In this case $M(t) = M_o \sin\omega_d t$ and the work expression [Eq. (2)] is replaced by

$$W_n = W_{\tau=\tau_n} = \frac{1}{k_B T}\int_{t_i}^{t_i+\tau_n} M(t)\frac{d\theta}{dt}dt, \qquad (4)$$

with $\tau_n = n2\pi/\omega_d$ with n integer. This is a stationary state that has never been studied before in the context of FT. We find that for any driving frequency ω_d the PDFs of W_n are Gaussian. The function $S(W_n)$, measured at $\omega_d/2\pi =$ 64 Hz and plotted in Fig. 4(a), is linear in W_n and the corresponding slope $\Sigma(n)$ is a function of n. The measured values of $\Sigma(n)$ are shown as function of n in Fig. 4(b), where the results obtained at $\omega_d/2\pi = 256$ Hz are plotted too. We see that the convergence rate is quite different in the two cases, which agree with our theoretical predictions for a second order Langevin equation [see Eq. (11)]. Also in the case of the sinusoidal forcing the agreement between the computed and measured finite time corrections is very good. These results prove not only that FTs asymptotically hold for any kind of forcing, but also that finite time corrections strongly depend on the specific dynamics. In the case of the sinusoidal forcing, the convergence is very slow: in Fig. 4(e), we see that it takes several dozens of excitation periods (500 ms for $\omega_d/2\pi = 64$ Hz) to get $\Sigma(n) = 1$ within 1%. On the contrary, for a ramp forcing this was achieved after a few τ_α (20 ms); see Fig. 3(c).

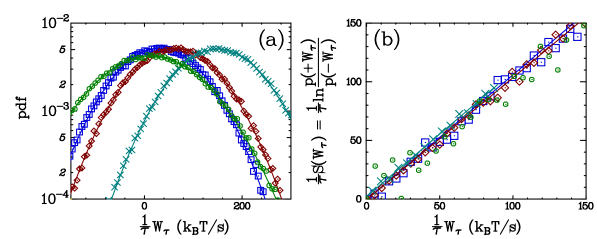

FIG. 2 (color online). TFT. (a) $P(W_\tau)$ for TFT for various τ/τ_α: 0.31 (○), 1.015 (□), 2.09 (◇), and 4.97 (×). Continuous lines are Gaussian fits. (b) TFT; §(W_τ) computed with the PDF of (a). The straight continuous lines are fits with slope 1, i.e., $\Sigma(\tau) = 1$, $\forall \tau$.

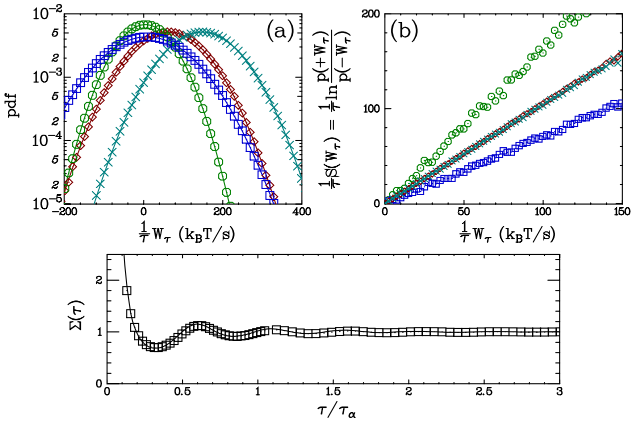

FIG. 3 (color online). SSFT with a ramp forcing. (a) PDF of W_τ for various τ/τ_α: 0.019 (○), 0.31 (□), 2.09 (◇), and 4.97 (×). (b) Corresponding functions $S(W_\tau)$. (c) The slope $\Sigma(\tau)$ of $S(W_\tau)$ is plotted versus τ [□: experimental values; continuous line: theoretical prediction Eq. (10) with no adjustable parameter].

Let us now compute the finite time corrections to TFT and SSFT [plotted in Figs. 3(c) and 4(e)] for the harmonic oscillator, applying a method very similar to the one already used in the context of the Jarzynski equality (see Ref. [13]). We write $\theta(t) = \bar{\theta}(t) + \delta\theta(t)$, where $\bar{\theta}(t)$ is the mean response of the system to the external torque and $\delta\theta(t)$ are the thermal fluctuations. The mean response $\bar{\theta}(t)$ [dark line in Figure 1(b) and 4(b)] is computed by performing an ensemble average of $\theta(t)$ over 10^3 responses to the $M(t)$ of Fig. 1(a), respectively, Fig. 4(a). It turns out that the measured $\bar{\theta}(t)$ is equal to the solution of Eq. (1) with $\eta = 0$ and with M equal to the applied time dependent torque. Once the mean behavior is known, it is useful to compare the statistical properties of $\delta\theta(t)$ measured at $M(t) \neq 0$ with the equilibrium ones. In Fig. 5(a) we plot the Gaussian fit of the PDF of $\delta\theta(t)$ measured at equilibrium $M(t) = 0$ (continuous line) and at $M(t) \neq 0$ (○). The two curves are equal within experimental errors. Thus we conclude that the external driving does not perturb the equilibrium PDF, which is a Gaussian of variance k_BT/C. In Fig. 5(b) we plot the power spectra of $\delta\theta$ in equilibrium (continuous line) and out equilibrium (dotted line). The two spectra are equal and they coincide with the theoretical spectrum of an equilibrium second order Langevin dynamics [Eq. (1) with $M = 0$] computed using the oscillator parameters. Thus we clearly see that, within statistical accuracy, the fluctuations of $\delta\theta$ measured at $M(t) \neq 0$ are those of equilibrium. This important observation is the key point to estimate the finite time corrections of FTs.

In order to compute $\Sigma(\tau)$, we first decompose the total work of the external torque into the sum of a mean part and a fluctuating one, i.e., $W_\tau = \bar{W}_\tau + \delta W_\tau$, where

$$\bar{W}_\tau = \frac{1}{k_BT}\int_{t_i}^{t_i+\tau}[M(t) - aM(t_i)]\frac{d\bar{\theta}}{dt}dt, \quad (5)$$

$$\delta W_\tau = \frac{1}{k_BT}\int_{t_i}^{t_i+\tau}[M(t) - aM(t_i)]\frac{d\delta\theta}{dt}dt, \quad (6)$$

with $a = 1$ for the ramp forcing [Eq. (2)] or $a = 0$ for sinusoidal driving [Eq. (4)]. From those equations and the aforementioned experimental observation on the fluctuations $\delta\theta$, we see that the fluctuations δW_τ have a Gaussian distribution, so the PDF of the work W_τ done by the external torque is fully characterized by its mean $\bar{W}_\tau$ and variance $\sigma^2 = \langle(\delta W_\tau)^2\rangle$, where $\langle . \rangle$ stands for ensemble

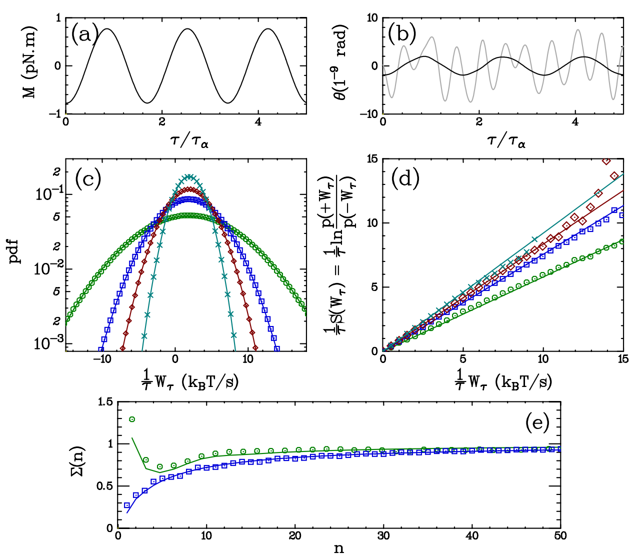

FIG. 4 (color online). Sinusoidal forcing. (a) Sinusoidal driving torque applied to the oscillator. (b) Response of the oscillator to this periodic forcing (gray line); the dark line represents the mean response $\bar{\theta}(t)$. (c) PDF of the work W_n integrated over n periods of forcing, with $n = 7$ (○), $n = 15$ (□), $n = 25$ (◇), and $n = 50$ (×). (d) The function $S(W_n)$ measured at $\omega_d/2\pi = 64$ Hz is plotted as a function of W_n for several n: (○) $n = 7$; (□) $n = 15$ (◇), $n = 25$; (×) $n = 50$. Continuous lines are functions $S(W_n)$ computed from Gaussian fits of PDF (in Fig. b). (e) The slopes $\Sigma(n)$, plotted as a function of n for two different driving frequencies $\omega_d = 64$ Hz (□) and 256 Hz (○); continuous lines are theoretical predictions from Eq. (11) with no adjustable parameter.

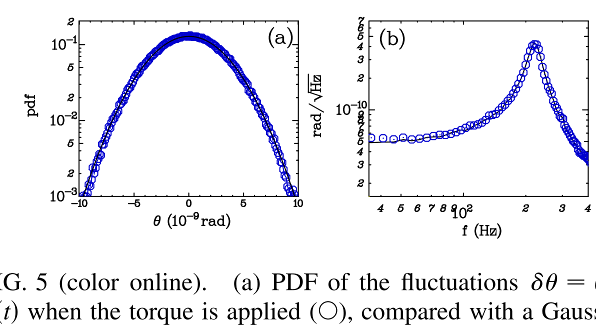

FIG. 5 (color online). (a) PDF of the fluctuations $\delta\theta = \theta - \bar{\theta}(t)$ when the torque is applied (○), compared with a Gaussian fit of the PDF at equilibrium (continuous line). (b) The measured spectrum of $\delta\theta$ (○) is compared with the prediction of fluctuation dissipation theorem in equilibrium (continuous line).

average. In such a case the FTs take a simple form:

$$S(W_\tau) = \frac{2\bar{W}}{\sigma^2} W_\tau = \Sigma(\tau) W_\tau, \tag{7}$$

where $\Sigma(\tau) \equiv [1 - \epsilon(\tau)]^{-1}$ following the notation of Ref. [6]. From Eq. (7) it is clear that to estimate the finite time correction $\Sigma(\tau)$ we need only to compute $\bar{W}_\tau$ and σ^2.

$\bar{W}_\tau$ is simply obtained by inserting in Eq. (5) the expression of $M(t)$ and the solution $\bar{\theta}$ of Eq. (1) with $\eta = 0$. The variance σ^2 is computed using Eq. (6). For the linear ramp of Fig. 1, we obtain

$$\sigma^2 = \frac{1}{(k_B T)^2} \frac{M_0^2}{\tau_r^2} \left[\tau^2 \langle \delta\theta^2(\tau) \rangle + \left\langle \left(\int_0^\tau \delta\theta(t) dt \right)^2 \right\rangle - 2\tau \int_0^\tau \langle \delta\theta(\tau)\delta\theta(t) \rangle dt \right], \tag{8}$$

which can be computed using the correlation function $R(\tau) = \langle \delta\theta(\tau)\delta\theta(0) \rangle$. As already explained, the experimental data indicate that the statistical properties of $\delta\theta(t)$ on the ramp are the same as the properties of the equilibrium fluctuations, which are well described by a second order Langevin dynamics. Thus we can use for $R(\tau)$ the known equilibrium correlation function of the thermal fluctuations, which for a second order Langevin dynamics is [13]

$$R(\tau) = \frac{k_B T}{C} \frac{\sin(\psi|\tau| + \varphi)}{\sin\varphi} \exp(-\alpha|\tau|), \tag{9}$$

where $\alpha = 1/\tau_\alpha$, $\alpha^2 + \psi^2 = \omega_0^2 = C/I_{\mathrm{eff}}$ and $\sin\varphi = \psi/\omega_0$. We checked our method on a first order Langevin equation, for which the exact results of Refs. [5,6] are available. We find that $\epsilon(\tau)$ for the work computed with our technique in a first order Langevin dynamics is the same as in Refs. [5,6] both for TFT and SSFT. Thus we can now safely apply our technique to a second order Langevin equation. We find that in the case of the TFT $\epsilon = 0 \forall \tau$, whereas in the case of the SSFT

$$\epsilon(\tau) = \frac{2}{\psi\tau} \left\{ \frac{\sin 3\varphi}{\omega_0 \tau} - \exp(-\alpha\tau) \times \left(\sin(2\varphi + \psi\tau) + \frac{\sin(3\varphi + \psi\tau)}{\omega_0 \tau} \right) \right\}. \tag{10}$$

The same calculations can be performed for any kind of $M(t)$. For example, with a sinusoidal forcing $M(t) = M_o \sin(\omega_d t)$, SSFT for the work W_n defined in Eq. (4) gives

$$\epsilon(\tau_n) = -\frac{\cos 2\gamma}{2\alpha\tau_n} \frac{\omega_0^2 + \omega_d^2}{\omega_d^2} + \frac{1}{\tau_n} \mathcal{O}(e^{-\alpha\tau_n}), \tag{11}$$

where γ is the phase shift between $\bar{\theta}(t)$ and $M(t)$, i.e, $\tan(\gamma) = -2\alpha\omega_d/(\omega_0^2 - \omega_d^2)$ and $\tau_n = 2n\pi/\omega_d$ with n integer. In Eq. (11), $\mathcal{O}(e^{-\alpha\tau_n})$ is a term that vanishes exponentially in $\alpha\tau_n$, the expression of which is complicated and will be reported in a longer article, together with many other interesting features.

These analytical results agree remarkably well with the experimental results for TFT and SSFT for the work fluctuations in a harmonic oscillator [see Figs. 3(c) and 4(e)].

In conclusion, we have applied the FTs to the work fluctuations of an oscillator driven out of equilibrium by an external force. The TFT holds for any time whereas the SSFT presents a complex convergence to the asymptotic behavior which strongly depends on the form of the driving. The exact formula of this convergence can be computed using several experimental evidences of the statistics of the fluctuation. These results are useful for many applications going from biological systems to nanotechnology, where the harmonic oscillator is the simplest building block.

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