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Abstract. – Heat fluctuations of a harmonic oscillator in contact with a thermostat and driven out of equilibrium by an external deterministic force are studied experimentally and theoretically within the context of Fluctuation Theorems. We consider the case of a periodic forcing of the oscillator, and we calculate the analytic probability density function of heat fluctuations. The limit of large time is discussed and we show that heat fluctuations satisfy the conventional fluctuation theorem, even if a different fluctuation relation exists for this quantity. Experimental results are also given for a transient state.

Out-of-equilibrium fluctuations have recently received a lot of attention, especially in the context of nanosystems and biomolecules where fluctuations are large. In the last decade, Fluctuation Theorems (FTs) appeared in nonequilibrium physics. They quantify the asymmetry of fluctuations of entropy production for a large class of systems, possibly far from equilibrium. These theorems were first demonstrated in deterministic dynamics of many degrees of freedom [1,2] and later extended to stochastic systems [3–6]. The FT states that the probability $P(\sigma = a)$ of observing an entropy production rate σ, measured over a time τ, with a value a, satisfies

$$\frac{P(\sigma = a)}{P(\sigma = -a)} \rightarrow \exp(a\tau) \quad \text{for large } \tau \text{ and any value } a \tag{1}$$

There are two classes of FTs. The *Stationary State Fluctuation Theorem* (SSFT) considers a nonequilibrium stationary state. SSFT holds in the limit of large time τ. The *Transient Fluctuation Theorem* (TFT) describes transient nonequilibrium states where τ measures the time interval since the system left its equilibrium state. Contrary to SSFT, the TFT holds for all times, *i.e.*, equation (1) is an equality even for arbitrarily small values of τ.

In this letter, we consider the heat Q_τ dissipated on a time interval τ in a thermostated system at temperature T rather than the entropy production. We study the fluctuations of the heat dissipated by a harmonic oscillator in contact with the thermostat and driven out of equilibrium by an external force. Experimental tests of FTs are rare. Some of them are performed in dynamical systems [8], in which the interpretation of the results is very difficult. In stochastic systems, a laboratory experiment was carried out by Wang *et al* using a Brownian particle in a moving optical trap [9]. Work fluctuations were shown to obey the

predictions of ref. [5]. Work and heat fluctuations were also studied in an electrical circuit by Garnier and Ciliberto [10] ; the theoretical predictions for both heat and work fluctuations were satisfied [5, 6]. These two systems are described by a first order Langevin equation. Systems described by a second order Langevin equation have been studied [3], and tested experimentally for work fluctuations W_τ [14]; as far as we know, no analytical results for the probability density functions (PDFs) of heat fluctuations Q_τ are available in this case. In ref. [5], van Zon and Cohen have calculated the Fourier transform of the PDF of Q_τ for a first order Langevin dynamics but no exact expression of the PDF itself is known.

In the following, we consider first a stationary state obtained by driving the system periodically in time. We calculate exactly the probability density function of Q_τ. We then compare our finding with new experimental results, and show that SSFT holds, and we discuss the large time limit. Finally experimental results of a TFT for the heat are reported.

To study FT, we measure the out-of-equilibrium fluctuations of a harmonic oscillator whose damping is due to the viscosity of a surrounding fluid, acting as a thermal bath. The oscillator is a torsion pendulum composed of a brass wire and a glass mirror glued in the middle of this wire. The elastic torsional stiffness of the wire is $C = 4.7 \times 10^{-4}$ N m rad^{-1}. It is enclosed in a cell filled by a water-glycerol solution at 60% concentration. The system is a harmonic oscillator with resonant frequency $f_o = \sqrt{C/I_{\text{eff}}}/(2\pi) = 217$ Hz and a relaxation time $\tau_\alpha = 2I_{\text{eff}}/\nu = 9.5$ ms. I_{eff} is the total moment of inertia of the displaced masses and ν is the oscillator viscous damping. The angular displacement of the pendulum θ is measured by a differential interferometer [12, 13]. The measurement noise is two orders of magnitude smaller than 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 using the Fluctuation Dissipation Theorem, is better than 3% (see [13]). We drive the system out-of-equilibrium by forcing the system with an external torque M by means of a small electric current J flowing in a coil glued behind the mirror. The coil is inside a static magnetic field, hence $M \propto J$. More details on the set-up can be found in ref. [12, 13]. The system is very well described by a second order Langevin equation:

$$I_{\text{eff}}\,\frac{\mathrm{d}^2\theta}{\mathrm{d}t^2} + \nu\,\frac{\mathrm{d}\theta}{\mathrm{d}t} + C\,\theta = M + \sqrt{2k_B T\nu}\,\eta, \qquad (2)$$

with η the noise, delta-correlated in time, $\beta = (k_B T)^{-1}$ with k_B Boltzmann's constant and T the temperature of the system. At equilibrium ($M = 0$ pN.m), the PDF of the thermal fluctuations $\delta\theta$ is Gaussian with variance $k_B T/C$. We apply two kinds of forcing. First, we study a periodic forcing : $M(t) = M_o \sin(\omega_d t)$ with $M_o = 0.78$ pN.m and $\omega_d/2\pi = 64$ Hz. We then analyze a linear forcing $M(t) = M_o t/\tau_r$ with $M_o = 6.2$ pN.m and $\tau_r = 0.01$ s $= 1.07\,\tau_\alpha$.

The change in internal energy ΔU_τ of the oscillator over a time τ, starting at a time t_i, is written as:

$$\Delta U_\tau = U(t_i + \tau) - U(t_i) = Q_\tau + W_\tau \qquad (3)$$

which is the first law of thermodynamics. W_τ is the work done on the system over a time τ:

$$W_\tau = \frac{1}{k_B\,T}\int_{t_i}^{t_i+\tau} M(t')\frac{\mathrm{d}\theta}{\mathrm{d}t}(t')dt' \qquad (4)$$

and Q_τ is the heat given to the system. Equivalently, $(-Q_\tau)$ is the heat dissipated by the system.

For a harmonic oscillator described by a second order Langevin equation, the internal

energy has two contributions : the kinetic and potential energies:

$$U(t) = \frac{1}{k_B \; T} \left[\frac{1}{2} I_{\text{eff}} \left[\frac{\mathrm{d}\theta(\mathrm{t})}{\mathrm{dt}} \right]^2 + \frac{1}{2} C\theta(t)^2 \right] \tag{5}$$

Multiplying eq. (2) by $\frac{\mathrm{d}\theta}{\mathrm{dt}}$ and integrating between t_i and $t_i + \tau$, we obtain exactly the first law of thermodynamics eq. (3) and have the following expression for the heat:

$$Q_\tau = \Delta U_\tau - W_\tau = -\frac{1}{k_B \; T} \int_{t_i}^{t_i+\tau} \nu \left[\frac{\mathrm{d}\theta}{\mathrm{dt}}(t') \right]^2 dt' + \frac{1}{k_B \; T} \int_{t_i}^{t_i+\tau} \eta(t') \frac{\mathrm{d}\theta}{\mathrm{dt}}(t')dt' . \tag{6}$$

The first term corresponds to the viscous dissipation and is always positive, whereas the second term can be interpreted as the work of the thermal noise which have a fluctuating sign.

We rescale the work W_τ (the heat Q_τ) by the average work $\langle W_\tau \rangle$ (the average heat $\langle Q_\tau \rangle$) and define: $w_\tau = \frac{W_\tau}{\langle W_\tau \rangle}$ ($q_\tau = \frac{Q_\tau}{\langle Q_\tau \rangle}$). Averages are obtained experimentally as time-averages, and they are proportional to τ on the stationary state under consideration.

We consider first the periodic forcing. In this case, we choose τ as a multiple of the period of the driving ($\tau = 2n\pi/\omega_d$ with n integer). The starting phase $t_i\omega_d$ is averaged over all possible t_i to increase statistics. The PDFs of w_τ, ΔU_τ and q_τ are plotted in Fig. 1 for different values of n. The average of ΔU_τ is clearly zero because the time τ is a multiple of the period of the forcing. The PDFs of the work (fig. 1a) are Gaussian for any n whereas the PDFs of heat fluctuations q_τ have exponential tails (fig. 1c). These exponential PDFs can be understood noticing that, from eq. (6), $-Q_\tau = W_\tau - \Delta U_\tau$ and that ΔU_τ has an exponential PDF independent of n (fig. 1b). Therefore, on a first approximation, the PDF of q_τ is the convolution between an exponential and a Gaussian.

To quantify the symmetry of the PDF around the origin, we define the function S as:

$$S(e_\tau) \equiv \frac{1}{\langle E_\tau \rangle} \ln \left[\frac{P(e_\tau)}{P(-e_\tau)} \right] \tag{7}$$

where e_τ stands for either w_τ or q_τ and E_τ stands for either W_τ or Q_τ. The question we ask is whether:

$$\lim_{\tau \to \infty} S(e_\tau) = e_\tau \tag{8}$$

as required by SSFT. $S(q_\tau)$ is plotted in Fig. 1d for different values of n ; three regions appear:

(I) For large fluctuations q_τ, $S(q_\tau)$ equals 2. When τ tends to infinity, this region spans from $q_\tau = 3$ to infinity.

(II) For small fluctuations q_τ, $S(q_\tau)$ is a linear function of q_τ. We then define $\Sigma_q(n)$ as the slope of the function $S(q_\tau)$, *i.e.* $S(q_\tau) = \Sigma_q(n) \, q_\tau$. This slope is plotted in Fig. 1e where we see that it tends to 1 when τ is increased. So, SSFT holds in this region II which spans from $q_\tau = 0$ up to $q_\tau = 1$ for large τ.

(III) A smooth connection between the two behaviors.

The PDF of the work being Gaussian, the functions $S(w_\tau)$ are proportional to w_τ for any τ, *i.e.* $S(w_\tau) = \Sigma_w(n) \, w_\tau$ (ref. [14]). $\Sigma_w(n)$ is plotted in Fig. 1e and we observe that it matches experimentally $\Sigma_q(n)$, for all values of n. So the finite time corrections to the FT for the heat are the same than the ones of FT for work [14] : $\Sigma_w(n) = \Sigma_q(n) = 1 + K/n + 1/n\mathcal{O}\left(e^{-\tau_n/\tau_\alpha} \right)$, where K is a constant.

We now give an analytical expression of the PDF of heat Q_τ. In order to do this, we write $\theta = \bar{\theta} + \delta\theta$ where $\bar{\theta}$ is the mean response of the system to the external work and $\delta\theta$ the thermal fluctuations. We suppose that fluctuations at $M(t) \neq 0$ are those of equilibrium, *i.e.*
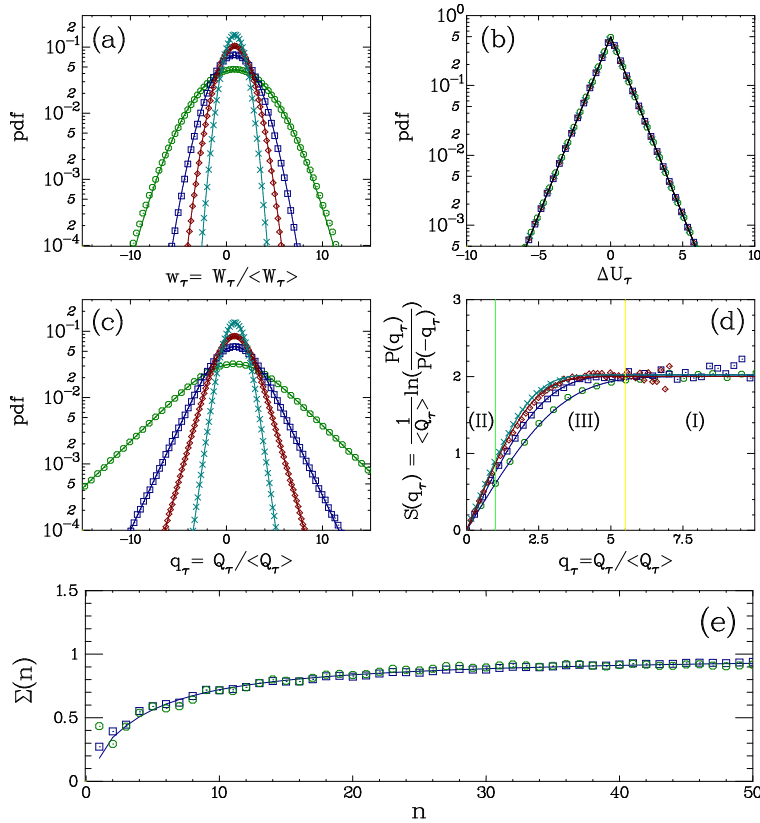
Fig. 1 – Sinusoidal forcing. a) PDFs of w_τ. b) PDFs of ΔU_τ. c) PDFs of q_τ. d) Functions $S(q_\tau)$. In all plots, the integration time τ is a multiple of the period of forcing, $\tau = 2n\pi/\omega_d$, with $n = 7$ ($\circ$), $n = 15$ ($\square$), $n = 25$ ($\diamond$) and $n = 50$ ($\times$). Continuous lines are theoretical predictions with no adjustable parameter. e) The slope $\Sigma_q(n)$ of $S(q_\tau)$ for $q_\tau < 1$, plotted as a function of n ($\circ$). The slope $\Sigma_w(n)$ of $S(w_\tau)$ plotted as a function of $n(\square)$. Continuous line is theoretical prediction.

that the external driving does not perturb the equilibrium PDF. This hypothesis is supported experimentally as shown in ref. [13] and detailed in [14]. Times τ under consideration are taken larger than $\tau_\alpha = 9, 5$ ms, so that exponential corrections to the autocorrelation functions, which are scaling like $\exp(-\tau/\tau_\alpha)$, can be neglected. Experimentally, $\tau/\tau_\alpha = 1.64\,n$, so this is correct as soon as n is larger than 3 or 4. Within this assumption, $\theta(t_i + \tau)$ and $\theta(t_i)$ are independent, and so are $\frac{d\theta}{dt}(t_i + \tau)$ and $\frac{d\theta}{dt}(t_i)$. Eq. (2) is second order in time, so $\theta(t)$ and $\frac{d\theta}{dt}(t)$ are independent at any given time t. Just like in our experiment, we choose the integration time τ to be a multiple of the period of the forcing, so $\langle \Delta U_\tau \rangle = 0$ and therefore $\langle W_\tau \rangle = -\langle Q_\tau \rangle$. Within this framework, we find that the PDF of ΔU_τ is exponential:

$$P(\Delta U_\tau) = \frac{1}{2} \exp(-|\Delta U_\tau|). \tag{9}$$

It is independent of τ because ΔU_τ depends only on θ and $\frac{d\theta}{dt}$ at times t_i and $t_i + \tau$ which are uncorrelated. This expression is in perfect agreement with the experimental PDFs for all times (see Fig. 1b). Experimentally, work fluctuations have a Gaussian distribution so it is fully characterized by its mean $\langle W_\tau \rangle$ and its variance σ_W^2. In a Gaussian case, the FTs

take a simple form $S(w_\tau) = \frac{2\langle W_\tau \rangle}{\sigma_W^2} w_\tau = \Sigma_w(n) w_\tau$. We have already computed the analytic expression of $\Sigma_w(n)$ [14] and this gives a relation between the variance and the mean value of W_τ: $\sigma_W^2 = 2\langle W_\tau \rangle + \mathcal{O}(\frac{1}{\tau})$ and so $\sigma_W^2 = 2|\langle Q_\tau \rangle| + \mathcal{O}(\frac{1}{\tau})$.

To obtain the PDF $P(Q_\tau)$ of the heat, we define its Fourier transform (characteristic function) as

$$\hat{P}_\tau(s) \equiv \int_{-\infty}^{\infty} dq_\tau e^{isq_\tau} P(q_\tau) \tag{10}$$

which can be computed exactly [5]. We then write $P(q_\tau)$ using eq. (6) as:

$$P(q_\tau) = \int \int d\theta d\dot{\theta} \tilde{P}\left(\Delta U_\tau - Q_\tau, \theta(t_i + \tau), \theta(t_i), \dot{\theta}(t_i + \tau), \dot{\theta}(t_i)\right) \tag{11}$$

where $\tilde{P}$ is the joint distribution of the work W_τ, θ and $\frac{d\theta}{dt}$ at the beginning and at the end of the time interval τ. This distribution is expected to be Gaussian because W_τ is linear in $\dot{\theta}$ and additionally θ, $\dot{\theta}$ and W_τ are Gaussian. Some algebra then yields:

$$\hat{P}_\tau(s) = \frac{1}{1+s^2} \exp\left(i\langle Q_\tau \rangle s - \frac{\sigma_W^2}{2} s^2\right) \tag{12}$$

The characteristic function of heat fluctuations is therefore the product of the characteristic function of an exponential distribution ($\frac{1}{1+s^2}$) with the one of a Gaussian distribution ($\exp\left(i\langle Q_\tau \rangle s - \frac{\sigma_W^2}{2} s^2\right)$). Thus the PDF of heat fluctuations is nothing but the convolution of a Gaussian and an exponential PDF, just as if W_τ and ΔU_τ were independent. The inverse Fourier transform can be computed exactly:

$$P(Q_\tau) = \frac{1}{4} \exp\left(\frac{\sigma_W^2}{2}\right)\left[e^{Q_\tau - \langle Q_\tau \rangle} \mathrm{erfc}\left(\frac{Q_\tau - \langle Q_\tau \rangle + \sigma_W^2}{\sqrt{2\sigma_W^2}}\right) + \right.$$
$$\left. e^{-(Q_\tau - \langle Q_\tau \rangle)} \mathrm{erfc}\left(\frac{-Q_\tau + \langle Q_\tau \rangle + \sigma_W^2}{\sqrt{2\sigma_W^2}}\right)\right], \tag{13}$$

where erfc stands for the complementary Erf function. In Fig. 1c, we have plotted the analytical PDF from eq. (13) together with the experimental ones, using values of σ_W^2 and $\langle Q_\tau \rangle$ from the experiment. The agreement is perfect for all values of n and with no adjustable parameters. Using eq. (13), we isolate three different regions for $S(q_\tau)$:

(I) if $Q_\tau > \sigma_W^2 + |\langle Q_\tau \rangle| = 3|\langle Q_\tau \rangle| + \mathcal{O}(1)$, then $S(q_\tau) = 2 + \mathcal{O}(\frac{1}{\tau})$. This case corresponds to large fluctuations and the PDF can be pictured as exponential with a non-vanishing mean.

(II) if $Q_\tau < \sigma_W^2 - |\langle Q_\tau \rangle| = |\langle Q_\tau \rangle| + \mathcal{O}(1)$, then $S(q_\tau) = \Sigma(n) q_\tau + \mathcal{O}(\frac{1}{\tau})$ with $\Sigma_q(n) = \frac{2|\langle Q_\tau \rangle|}{\sigma_W^2} = \Sigma_w(n)$. In this case, heat fluctuations are small and behave like work fluctuations. The slope $\Sigma(\tau)$ is the same as the one found in the case of work fluctuations. The exact correction to the asymptotic value is plotted in Fig. 1e and again it matches perfectly the experimental behavior.

(III) for $\sigma_W^2 - |\langle Q_\tau \rangle| < Q_\tau < \sigma_W^2 + |\langle Q_\tau \rangle|$, there is an intermediate region connecting cases (I) and (II) by a second order polynomial $S(q_\tau) = 2. - \frac{\Sigma(\tau)}{4}(q_\tau - (1 + \frac{2}{\Sigma(\tau)}))^2 + \mathcal{O}(\frac{1}{\tau})$.

Those three regions offer a perfect description of the three domains observed experimentally (Fig. 1d).

Now, we examine the limit of infinite τ in which SSFT is supposed to hold but which depends on the variable we use : either the heat Q_τ or the normalized heat q_τ. First we discuss

Q_τ. The asymptotic behavior of the PDF of Q_τ (eq. (13)) for large τ is Gaussian with variance σ_W^2. Thus, the PDF of Q_τ coincides with the PDF of W_τ for τ strictly infinite. As we have already shown, work fluctuations satisfy the conventional SSFT, therefore heat fluctuations also satisfy the conventional SSFT (eq. (1)). We have found three different regions defined by two characteristic values, but in the limit of infinite time τ, only region (II) is relevant: region (II) is bounded from above by $|\langle Q_\tau \rangle| + \mathcal{O}(1)$ with the average $\langle Q_\tau \rangle$ being linear in τ. We see that all the behavior of the fluctuations of Q_τ is captured by region (II) where $S(q_\tau)$ is linear for all Q_τ and SSFT holds. Second, we turn to the normalized heat q_τ. As the average value of Q_τ is linear with τ, rescaling by $\langle Q_\tau \rangle$ is equivalent to a division by τ ; the mean of q_τ is then 1. This normalization changes into constants the two characteristic values which delimit the three regions : the boundary between (II) and (III) is now $1 + \mathcal{O}(1/\tau)$ and the boundary between (III) and (I) is $3 + \mathcal{O}(1/\tau)$. The function $S(q_\tau)$ is not linear for large values of q_τ but only in region (II) ($q_\tau < 1$), for small fluctuations. So SSFT is satisfied only for small fluctuations but not for all values of q_τ, and instead of a FT, we rather have a fluctuation relation. These two descriptions in terms of Q_τ or q_τ are in fact a problem of two non-commutative limits. The first description implies that one takes the limit τ infinite before taking the limit of large Q_τ. The second description does the opposite. However, the probability to have large fluctuations decreases with τ and experimentally, for large τ, only the region (II) where SSFT holds ($q_\tau < 1$) can be seen.

Finally, we briefly report results for the Transient Fluctuation Theorem. For this, we choose a torque $M(t) = M_o t/\tau_r$ linear in time, and the system is at equilibrium at $t_i = 0$ ($M(t_i = 0) = 0$ pN.m). Unlike with the periodic driving, the average of the variation of internal energy $\langle \Delta U_\tau \rangle$ is not vanishing. The work done by $M(t)$ is used by the system to increase his internal energy but a small amount of energy is lost by viscous dissipation and exchange with the thermostat. The PDF of W_τ/τ, $\Delta U_\tau/\tau$ and Q_τ/τ are plotted in Fig. 2 for different values of τ/τ_α. Averages $\langle W_\tau/\tau \rangle$ and $\langle \Delta U_\tau/\tau \rangle$ are linear in τ. However, their difference (eq. (6)) $\langle Q_\tau/\tau \rangle$ is constant and of the order of a few $k_B T/s$ (Fig. 2c). The shapes of the PDF are different from the ones obtained with the periodic forcing. Work fluctuations have a Gaussian PDF for any values of τ, moreover TFT holds for W_τ [14]. In contrast ΔU_τ have a different probability distribution. Fig. 2b shows that the PDF are not symmetric around the mean value. Extreme events have again an exponential distribution. For exactly the same reasons as in the case of a periodic forcing, the PDFs of Q_τ/τ are not Gaussian and have exponential tails for extreme fluctuations : $P(Q_\tau/\tau) = A \exp(-\alpha |Q_\tau/\tau|)$. As we can see in fig. 2c, the PDFs are not symmetric around the mean value, thus there are two pairs (α, A), one for positive value of large fluctuations (α_+, A_+) and one for negative (α_-, A_-). Thus, there is a simple expression of $S(q_\tau)$ for large fluctuations:

$$S(q_\tau) = (\alpha_+ - \alpha_-)q_\tau + \frac{1}{\langle Q_\tau \rangle} \ln \left(\frac{A_+}{A_-} \right) \tag{14}$$

It can be seen experimentally in fig. 2d. $S(q_\tau)$ is not proportional to q_τ, therefore TFT is not satisfied for finite time. However, for large value of τ, the PDF of Q_τ becomes symmetric around the mean value and only the Gaussian behavior remains. Thus, TFT appears to be satisfied experimentally in the limit of infinite τ. This breaks the expected property of TFT to be valid at any time.

In conclusion we have proposed an SSFT for heat fluctuations of a harmonic oscillator driven in a stationary out of equilibrium state by a periodic external force. An exact expression of the PDFs of the heat Q_τ averaged over a time τ is given. This PDF is in perfect agreement with experimental data. For finite times, we have isolated different behaviors: one
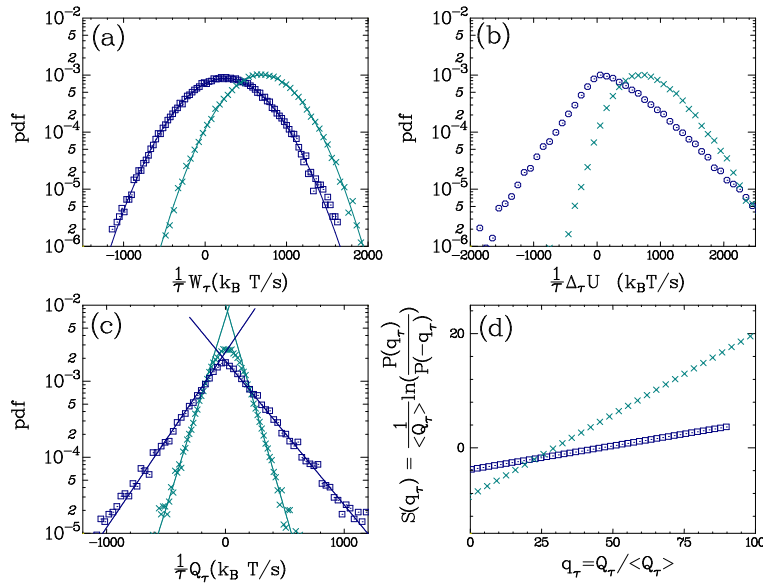
Fig. 2 – TFT. All plots are given for two values of τ/τ_α: 0.3 ($\square$) and 1.04 ($\times$). a) PDFs of $\frac{1}{\tau}W_\tau$. Continuous lines are Gaussian fits. b) PDFs of $\frac{1}{\tau}\Delta U_\tau$. c) PDFs of $\frac{1}{\tau}Q_\tau$. Continuous lines are exponential fits for large Q_τ. d) Functions $S(q_\tau)$ for large heat fluctuations q_τ computed using exponential fits.

for small fluctuations (Gaussian behavior) and the other for extreme fluctuations (exponential behavior). SSFT holds for infinite time. We have also studied a TFT for Q_τ using linear forcing and found that FT is satisfied only in the limit of large times.

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