

Work fluctuations in a nematic liquid crystal

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Abstract. The orientation fluctuations of the director of a liquid crystal are measured, by a sensitive polarization interferometer, close to the Fréedericksz transition, which is a second order transition driven by an electric field. Using mean field theory, we define the work injected into the system by a change of the electric field and we calibrate it using Fluctuation-Dissipation Theorem. We show that the work fluctuations satisfy the Transient Fluctuation Theorem. An analytical justification of this result is given. The open problems for the out of equilibrium case are finally discussed.

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1. Introduction

The physics of small systems used in nanotechnology and biology has recently received an increased interest. In such systems, fluctuations of the work injected become of the order of the mean value, leading to unexpected and undesired effects. For example, the instantaneous energy transfer can flow from a cold source to a hot one. The probabilities of getting positive and negative injected work are quantitatively related in non equilibrium system using Fluctuation Theorem (FT) [1]-[5]. This theorem has been both theoretically and experimentally studied in Brownian systems described by a Langevin equation [6]-[14]. The *Transient Fluctuation Theorem* (TFT) of the injected work W_τ considers the 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 system. For these systems, FT holds for all integration time and all fluctuation magnitudes :

$$S(a) = \ln \left[\frac{p(W_\tau = +a)}{p(W_\tau = -a)} \right] = \frac{a}{k_B T} \quad \forall a \quad \forall \tau \quad (1)$$

where k_B is the Boltzmann constant, T the temperature of the heat bath and $p(W_\tau)$ is the probability density function (PDF) of the injected work W_τ . S is called symmetry function. In a recent paper [15], some surprising results have been obtained for the TFT in a spatially extended system where the fluctuations of the work injected by an electric

field into a nematic liquid crystal (LC) have been studied. More precisely, the authors find an agreement between experimental results and TFT only for particular values of the observation time τ . We report in this article experimental results on the same system using a different measurement technic. We show that in our experiment, TFT holds for all integration time and all fluctuation magnitude. The paper is organized as follow. In the section 2, we describe the experimental setup. In the section 3, the work and the free energy of the LC system are defined. The experimental results are presented in section 4. Finally, we conclude in section 5.

2. Experimental setup

In our experimental apparatus, we measure the spatially averaged alignment of the LC molecules, whose local direction of alignment is defined by the unit vectors \vec{n} . The LC is confined between two parallel glass plates at a distance L . The inner surfaces of the confining plates have transparent Indium-Tin-Oxyde (ITO) electrodes, used to apply an electric field \vec{E} . The plates surfaces are coated by a thin layer of polymer which is mechanically rubbed into one direction. This treatment of the glass plates align the molecules of the LC in a unique direction parallel to the surface (planar alignment), *i.e.* all the molecules near the surfaces have the same director parallel to x -axis and $\vec{n} = (1, 0, 0)$ (see figure 1a). This kind of coating excludes weak anchoring effect described in the literature [17]. The experiments are performed on nematic liquid crystal (5CB) produced by Merck in a cell of thickness $L = 9 \mu\text{m}$.

The LC is submitted to an electric field perpendicular to the plates by applying a voltage V between the ITO electrodes. In order to avoid electrohydrodynamic effects of the motion of the ions invariably present in the liquid crystal, we apply an AC voltage at a frequency of $f_V = 1 \text{ kHz}$ ($V = \sqrt{2}V_0 \cos(2\pi f_V t)$) [18, 19]. When the voltage V_0 exceeds a threshold value V_c , the planar state becomes unstable and the LC molecules try to align parallel to the electric field (fig. 1a). This is known as the Fréedericksz transition which is second order. Above the critical value of the field, the equilibrium structure of the LC depends on the reduced control parameter defined as $\epsilon = \frac{V_0^2}{V_c^2} - 1$. The sample is thermostated at room temperature, *i.e.* $T = 295 \text{ K}$. The stability of the temperature is better than 0.5 K over the duration of the experiment.

The measurement of the alignment of the LC molecules uses the anisotropic optical properties of the LC, *i.e.* the cell is a birefringent plate whose local optical axis is parallel to the director \vec{n} . This optical anisotropy can be precisely estimated by measuring the phase shift Φ between two linearly polarized beams which cross the cell, one polarized along x -axis (ordinary ray) and the other along the y -axis (extraordinary ray). The experimental set-up employed is schematically shown in fig. 1c. The beam produced by a stabilized He-Ne laser ($\lambda = 632.8 \text{ nm}$) passes through the LC cell and it is finally back-reflected into the LC cell by a mirror. The lens between the mirror and the LC cell is used to compensate the divergency of the beam inside the interferometer. Inside the cell, the laser beam is parallel and has a diameter of about 2 mm. The beam

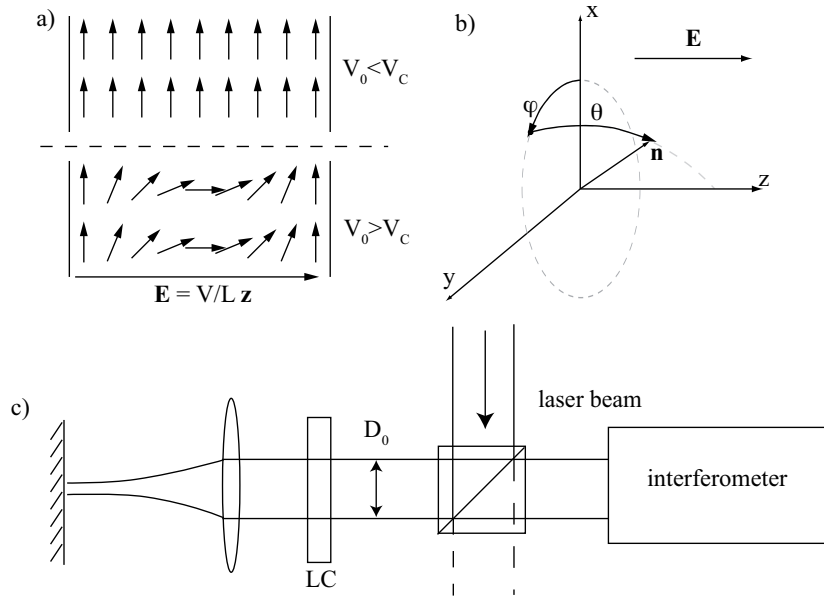


Figure 1. a) The geometry of Fréedericksz transition: director configuration for $V_0 < V_c$ and director configuration for $V_0 > V_c$. b) Definition of angular displacements (θ, φ) of the director of one nematic \vec{n} . c) Experimental setup. A laser beam passes through the LC cell and a polarization interferometer measures the phase shift Φ between the ordinary and extraordinary rays [21].

is at normal incidence to the cell and linearly polarized at 45° from the x -axis, *i.e.*, can be decomposed in an extraordinary beam and in an ordinary one. The optical path difference, between the ordinary and extraordinary beams, is measured by a very sensitive polarization interferometer [21].

The phase shift Φ takes into account the angular displacement in xz -plane, θ , and in xy -plane, φ (fig. 1b). For V_0 larger than the threshold value V_c , Φ depend only on θ and can be written as :

$$\Phi = \frac{1}{\mathcal{A}} \iint_{\mathcal{A}} dx dy \left[\frac{2\pi}{\lambda} \int_0^L \left(\frac{n_o n_e}{\sqrt{n_o^2 \cos^2(\theta)^2 + n_e^2 \sin^2(\theta)^2}} - n_o \right) dz \right] \quad (2)$$

with (n_o, n_e) the two anisotropic refractive indices [18, 19] and $\mathcal{A} = \pi D_0^2/4$ is the area of the measuring region of diameter D_0 in the (x, y) plane. The global variable of our interest is ζ defined as the spatially averaged alignment of the LC molecules, and more precisely :

$$\zeta = \frac{2}{L} \frac{1}{\mathcal{A}} \iint_{\mathcal{A}} dx dy \int_0^L (1 - n_x^2) dz \quad (3)$$

Close to the critical value V_c , using boundary conditions, the space dependence of θ along z has the following form : $\theta = \theta_0(x, y, t) \sin(\pi z/L)$, thus $n_x = \cos \theta$ [18, 19, 20]. If θ_0 remains small, ζ takes a simple form in terms of θ_0 :

$$\zeta = \frac{1}{\mathcal{A}} \iint_{\mathcal{A}} dx dy \theta_0^2 \quad (4)$$

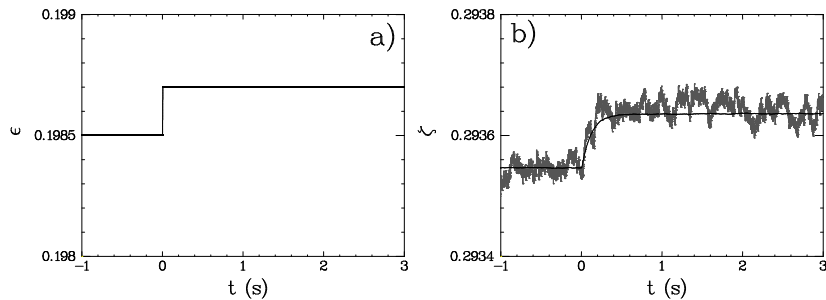


Figure 2. Typical measurement cycle. a) Dependence of ϵ on time. The perturbation $\delta\epsilon$ is switched on at $t = 0$. b) One realization of the response of ζ when the amplitude of the control parameter is changed from $\epsilon_0 = 0.1985$ and $\delta\epsilon = 2 \cdot 10^{-4}$ as shown in a). The thin line represents the average response $\langle \zeta \rangle$ done over 2500 experiments.

After some algebra, we can show that the phase shift is a linear function of ζ :

$$\Phi = \Phi_0 \left(1 - \frac{n_e(n_e + n_o)}{4n_o^2} \zeta \right) \quad (5)$$

$$\Phi_0 \equiv \frac{2\pi}{\lambda} (n_e - n_o) L \quad (6)$$

The phase Φ , measured by the interferometer, is acquired with a resolution of 24 bits at a sampling rate of 16384 Hz and then filtered at 500 Hz in order to suppress the AC voltage at f_V . The need of such a high acquisition frequency will be explained in the next section. The instrumental noise of the apparatus [21] is three orders of magnitude smaller than the amplitude $\delta\Phi$ of the fluctuations of Φ induced by the thermal fluctuations of ζ . In a recent paper, we have shown that ζ is characterized by a mean value $\langle \zeta \rangle \propto \epsilon$ and fluctuations which have a lorentzian spectrum [22, 16].

We are interested here in the fluctuations of the work injected into the LC when it is driven away from an equilibrium state to another using a small change of the voltage (*i.e.* the control parameter), specifically:

$$\epsilon = \begin{cases} \epsilon_0 & t < 0 \\ \epsilon_0 + \delta\epsilon & t \geq 0 \end{cases} \quad (7)$$

We take care that the system is in equilibrium at $t = 0$ by applying the value ϵ_0 for a time much larger than the relaxation time (τ_r) of the system. A typical measurement cycle is depicted in Fig. 2 where we plot $\epsilon(t)$ (Fig. 2a) and $\zeta(t)$ (Fig. 2b) as functions of time. We repeat the same cycle 2500 times to compute, over this ensemble of experiments, the probability density function of the work W_τ injected during a time τ . We choose the amplitude of $\delta\epsilon$ sufficiently small such that the mean response of the system to this excitation is comparable to the thermal noise amplitude, as can be seen in figure 2b) for $\epsilon_0 = 0.1985$ and $\delta\epsilon = 2 \cdot 10^{-4}$.

3. Landau equation and definition of the work

We define now the work injected into this system using the kind of forcing plotted in fig. 2a. The free energy is the sum of the electric energy and the potential energy :

$$F = \iiint_{\mathcal{V}} dx dy dz (f_{elastic} + f_{electric}) \quad (8)$$

Elastic free energy is a function of the director \vec{n} and its spatial derivative [18, 19] and electric free energy is $f_{elect} = -1/2\epsilon_0\epsilon_a(\vec{E}\cdot\vec{n})^2$ where ϵ_a is the dielectric anisotropy of the LC (ϵ_a is positive for 5CB). As the area of the measuring beam is much larger than the correlation length, we can neglect spatial derivative over x and y . Using the sinusoidal approximation for θ , *i.e.* $\theta(x, y, z, t) = \theta_0(x, y, t) \sin(\pi z/L)$ [18], free energy has the following form :

$$F = \iint_{\mathcal{A}} dx dy \left(\frac{\pi^2 K_1}{4L} (\theta_0^2 + \frac{\kappa}{4} \theta_0^4) - \frac{\epsilon_0 \epsilon_a}{4L} V^2 (\theta_0^2 - \frac{1}{4} \theta_0^4) \right) \quad (9)$$

$$F = \iint_{\mathcal{A}} dx dy \left(\frac{\pi^2 K_1}{4L} \left[-\epsilon \theta_0^2 + \frac{1}{4} (\kappa + \epsilon + 1) \theta_0^4 \right] \right) \quad (10)$$

where κ is equal to $(K_3 - K_1)/K_1$, $\epsilon = V_0^2/V_c^2 - 1$ and K_1 , K_2 and K_3 are the three elastic constants of the LC. To simplify the analysis the dependance of θ_0 in x and y can be neglected and the free energy takes the simple form:

$$F = B \left[-\epsilon \theta_0^2 + \frac{1}{4} (\kappa + \epsilon + 1) \theta_0^4 \right] \quad (11)$$

where $B = \mathcal{A} \pi^2 K_1 / 4L$. From eq.11 we can obtain the equation of momentum [20]:

$$\frac{\gamma \mathcal{A} L}{2} \frac{d\theta_0}{dt} = B \left[2 \epsilon \theta_0 - (\kappa + \epsilon + 1) \theta_0^3 \right] + \eta \quad (12)$$

where γ is a viscosity coefficient and η a thermal noise delta-correlated in time, such that :

$$\langle \eta(t) \rangle = 0 \quad \text{and} \quad \langle \eta(t_1) \eta(t_2) \rangle = k_B T \gamma \mathcal{A} L \delta(t_2 - t_1) \quad (13)$$

where $\langle \cdot \rangle$ stands for ensemble average. First, we have to calibrate the system, *i.e.* determine the value of the constant B that is a function of the area \mathcal{A} which is very difficult to precisely determine on the experiment. For this calibration, we use Fluctuation-Dissipation Theorem (FDT). We apply a small change of the voltage V_0 and, so of the control parameter, *i.e.* $\epsilon = \epsilon_0 + \delta\epsilon$. We separate θ_0 into the average part at ϵ_0 and a deviation due to $\delta\epsilon$: $\theta_0(t) = \psi_0 + \Delta(t)$, where $\psi_0 = 2\epsilon_0/(\kappa + \epsilon_0 + 1)$ is the stationary solution of eq.12 at $\epsilon = \epsilon_0$. If the response is linear, the average value of Δ is of the order of $\delta\epsilon$. In this limit, the equation of momentum can be rewritten :

$$\frac{\gamma \mathcal{A} L}{2} \frac{d\Delta}{dt} = B \left[2 \epsilon_0 - (\kappa + \epsilon_0 + 1) 3\psi_0^2 \right] \Delta + 2 B \delta\epsilon \psi_0 \left(1 - \frac{\psi_0^2}{2} \right) + \eta + O(\Delta^2) \quad (14)$$

The external torque, and so the conjugate variable to θ_0 , is equal to

$$\Gamma_{ext} = 2B \delta\epsilon \psi_0 \left(1 - \frac{\psi_0^2}{2} \right). \quad (15)$$

We define the integrated linear response function using an heaviside for $\delta\epsilon$:

$$R(\tau) = \frac{\langle \Delta(\tau) \rangle}{\Gamma_{ext}} = \frac{1}{2B \psi_0 \left(1 - \frac{\psi_0^2}{2}\right)} R_{\delta\epsilon} \quad (16)$$

where $R_{\delta\epsilon}$ is the integrated response function of θ_0 to $\delta\epsilon$. R is related by FDT to the autocorrelation function, $C_{\theta_0}(\tau) = \langle \delta\theta_0(t + \tau) \delta\theta_0(t) \rangle$, of the fluctuations $\delta\theta_0$ of θ_0 , that is:

$$R(\tau) = \frac{1}{k_B T} (C_{\theta_0}(0) - C_{\theta_0}(\tau)) \quad (17)$$

The global variable measured by the interferometer and defined in eq. 3 is $\zeta = 1/\mathcal{A} \iint \theta_0^2 dx dy \simeq \psi_0^2 + 2\psi_0 \delta\theta_0$. Thus the mean value of ζ is $\zeta_0 = \psi_0^2$ and the fluctuations $\delta\zeta$ of ζ can be related to the fluctuations of θ_0 : $\delta\zeta = 2\psi_0 \delta\theta_0$. We measure the autocorrelation function of $\delta\zeta$, $C_\zeta = 4\psi_0^2 C_{\theta_0}$. The response function of ζ to $\delta\epsilon$ is also related to $R_{\delta\epsilon}$: $R_{\zeta, \delta\epsilon} = 2\psi_0 R_{\delta\epsilon}$. Using fluctuation-dissipation theorem, we can measure the constant B :

$$\frac{1}{B'} R_{\zeta, \delta\epsilon} = \frac{1}{k_B T} (C_\zeta(0) - C_\zeta(\tau)) \quad (18)$$

where $B' = B (1 - \psi_0^2/2)$.

We define now the work, W_τ , injected into the system when the control parameter ϵ is suddenly changed from ϵ_0 to $\epsilon = \epsilon_0 + \delta\epsilon$ at $t = 0$ (eq.7). Using equation 12, W_τ can be written as :

$$W_\tau = B \int_0^\tau \delta\epsilon(t') \frac{d\theta_0}{dt} (2\theta_0 - \theta_0^3) dt' \quad (19)$$

we consider that the external torque is $B\delta\epsilon(t') (2\theta_0 - \theta_0^3)$. This expression can be integrated into :

$$W_\tau = B\delta\epsilon \left(\theta_0^2(\tau) - \theta_0^2(0) - \frac{\theta_0^4(\tau) - \theta_0^4(0)}{4} \right), \quad (20)$$

Using the assumption of the limit of small θ_0 and neglecting its dependance in the x and y directions, the work has a simple form in terms of $\zeta = \theta_0^2$:

$$W_\tau = B\delta\epsilon \left(\zeta(\tau) - \zeta(0) - \frac{\zeta^2(\tau) - \zeta^2(0)}{4} \right) \quad (21)$$

Notice that if, ϵ_0 is not too large, the quadratic term in this expression can be neglected, but it is not the case in general. The work W_τ is of course a fluctuating quantity because of the fluctuation of ζ .

4. Fluctuations of the work injected into the system

4.1. Experimental results

The first step consists in calibrating the system. Indeed as we have already mentioned the parameter B' appearing in eq.18 depends on \mathcal{A} , i.e. the effective area of the laser beam inside the cell that is very difficult to estimate with a good accuracy. In order to

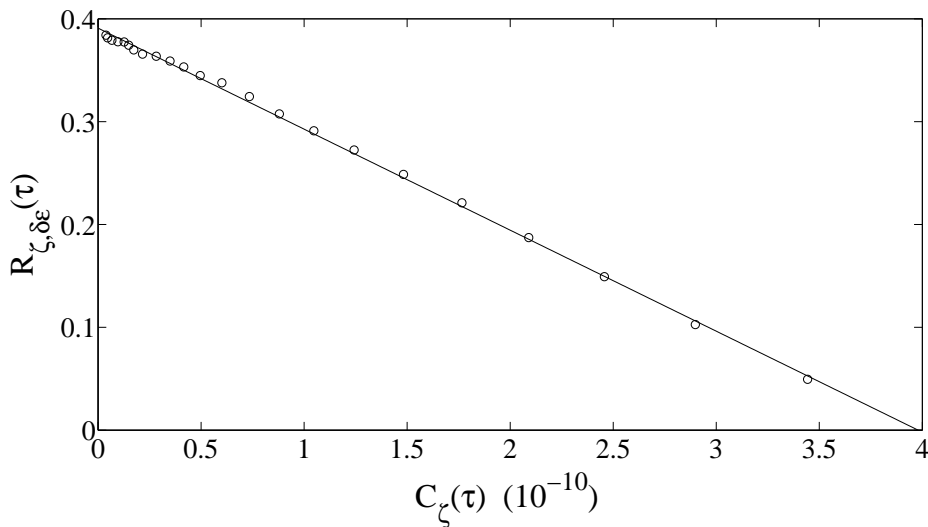


Figure 3. Fluctuation-dissipation theorem. $R_{\zeta, \delta\epsilon}(\tau)$ is represented as a function of $C_{\delta\zeta}(\tau)$. Continuous line represents linear fits. The measurement has been done at $\epsilon = 0.1985$ with $\delta\epsilon = 2 \cdot 10^{-4}$

calibrate the system we use FDT. In this case we use a sufficiently small perturbation $\delta\epsilon$ to insure that the response is linear. Thus we obtain the integrated linear response of the system : $R_{\zeta, \delta\epsilon}(\tau)$. Then we measure the correlation C_{ζ} . The function $R_{\zeta, \delta\epsilon}(\tau)/B'$ is a linear function of C , as can be seen in figure 3 and the slope seems to be independent of $\langle\zeta\rangle$. Using FDT, the slope is equal to $B'/k_B T$; by this method, we can calibrate the constant B' . We measure $B' = 4.03 \cdot 10^{-2} \pm 2 \cdot 10^{-3}$. Using in the equation for $B' = (1 - \psi_0^2/2)(\pi^2 K_1 \mathcal{A})/(4L)$, the measured values of B' and ψ_0 , $K_1 = 6.4 \cdot 10^{-12}$ N (value for the LC 5CB) and $L = 9 \mu\text{m}$ we estimate $\mathcal{A} = 2.4 \cdot 10^{-6}$ m². This value is consistent with a $D_0 = 1.75$ mm which is very close to the estimated diameter of the laser beam inside the cell which is about 2 mm. This justify our approach.

The system is at equilibrium at $t = 0$ s and we are now interested in the work done by the change of $\delta\epsilon$ during a time τ , W_{τ} . We need a high acquisition frequency to reduce experimental errors in the determination of the first point of each cycle $t = 0$. The probability density function of W_{τ} are plotted in figure 4 for different values of τ . We find that the PDFs are Gaussian for all values of τ and the average value of W_{τ} is equal to a few $k_B T$. We also notice that the probability of having negative values decreases when τ is increased. The symmetry function is plotted in fig. 4b. It is a linear function of W_{τ} for any τ , that is $S(W_{\tau}) = \Sigma(\tau)W_{\tau}$. Within experimental errors, we measure the slope $\Sigma(\tau) = 1$. Thus, for our experimental system, the TFT is verified for any time τ .

To test the importance of the quartic terms in the expression of the work, we have done the same experiments using a higher value of the control parameter $\epsilon_0 = 1.039$. The perturbation remains small $\delta\epsilon = 4 \cdot 10^{-4}$ (see figure 5). The quadratic terms in the expression of the work cannot be neglected in this situation. We find that the PDFs of the work are also Gaussian for all the integration times. This is normal because the

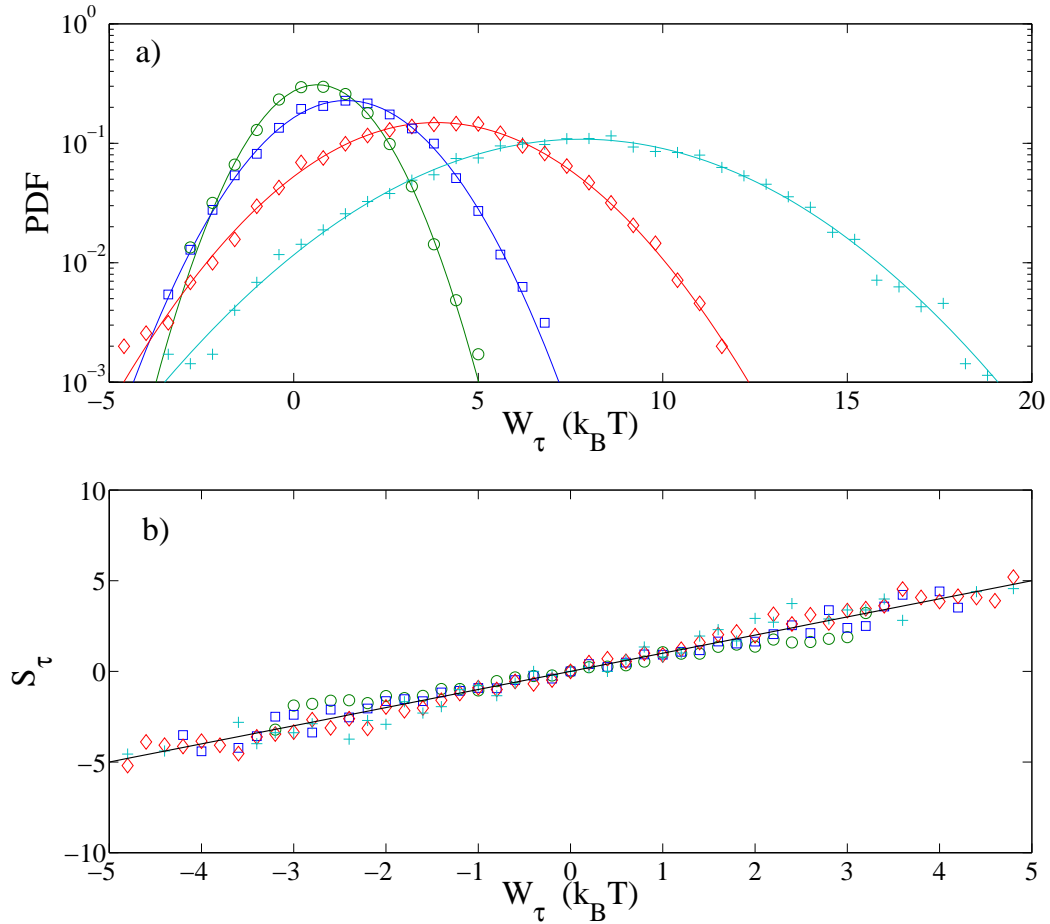


Figure 4. Work fluctuations a) PDFs of the work W_τ injected into the system at $\epsilon_0 = 0.1985$ by a perturbation $\delta\epsilon = 2 \cdot 10^{-4}$, for various τ : 10 ms (\circ), 20 ms (\square), 50 ms (\diamond) and 100 ms ($+$). Continuous lines are Gaussian fits. b) Symmetry functions $S(W_\tau)$ computed using the PDF of a) for the same values of τ . The straight continuous line is a line with slope 1 for all values of τ .

perturbation is such small that we are in the realm of the applicability of the FDT. The symmetry function is plotted in fig. 5b. It is a linear function of W_τ for any τ . Within experimental errors, we measure the slope equal to 1. Thus, for our experimental system, the TFT is verified for any time τ .

4.2. Comparison with theory

These results can be justified for small ϵ_0 and $\delta\epsilon$. Decomposing W_τ into the sum of a mean part $\langle W_\tau \rangle$ and a fluctuating one δW_τ , *i.e.* $W_\tau = \langle W_\tau \rangle + \delta W_\tau$. The distributions of the injected work are Gaussian for all values of τ , so the symmetry function takes a simple form :

$$S(W_\tau) = \frac{2\langle W_\tau \rangle}{\sigma_W^2} W_\tau \quad (22)$$

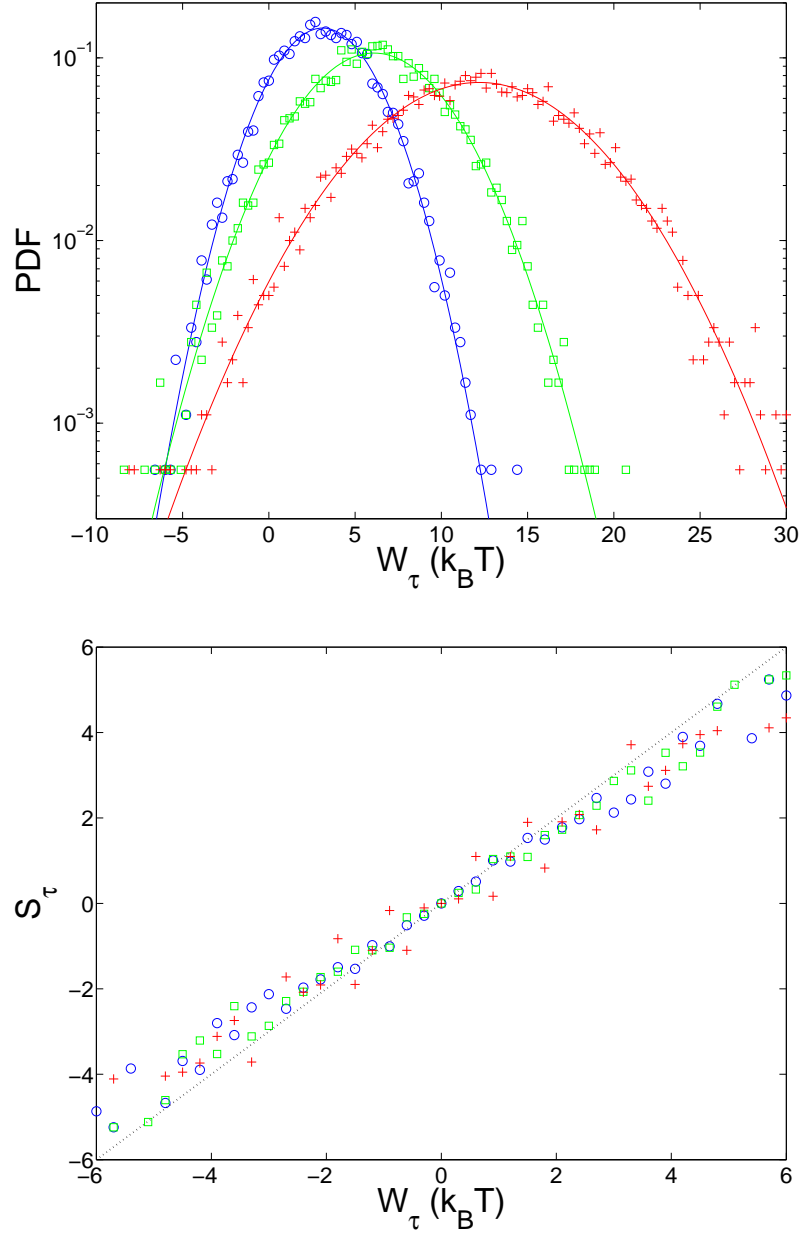


Figure 5. Work fluctuations a) PDFs of the work W_τ injected into the system at $\epsilon_0 = 1.039$ by a perturbation $\delta\epsilon = 4 \cdot 10^{-4}$, for various τ : 10 ms (o), 20 ms (\square), 50 ms (+). Continuous lines are Gaussian fits. b) Symmetry functions $S(W_\tau)$ computed using the PDF of a) for the same values of τ . The dashed line is a line with slope 1 for all values of τ .

where σ_W^2 is the variance of the fluctuation of the injected work. The mean value of the work is for small ϵ_0 and small $\delta\epsilon$:

$$\langle W_\tau \rangle = B\delta\epsilon(\langle \zeta(\tau) \rangle - \langle \zeta(0) \rangle) \quad (23)$$

$$= R_{\zeta, \delta\epsilon}(\tau) B^2 \delta\epsilon^2 \quad (24)$$

The variance of the fluctuations is given by :

$$\sigma_W^2 = B^2 \delta\epsilon^2 (\langle \delta\zeta(\tau)^2 \rangle + \langle \delta\zeta(0)^2 \rangle - 2\langle \delta\zeta(\tau)\delta\zeta(0) \rangle) \quad (25)$$

$$= 2B^2 \delta\epsilon^2 (C(0) - C(\tau)) \quad (26)$$

Using FDT, we obtain that $2k_B T \langle W_\tau \rangle = \sigma_W^2$, thus FT is satisfied for all integration times and all fluctuation magnitudes as can be seen in fig. 4b).

5. Conclusion

In conclusion, we have defined in a LC cell close to Fréedericksz transition the work injected into the system by the change of $\delta\epsilon$ during a time τ using the assumption of small angle θ_0 and neglecting its dependance in x and y directions. We have shown that the TFT holds for the work injected in a spatially extended system for all integration times and all fluctuation magnitudes. We have tested this result for another thickness of the LC cell and other values of ϵ . It is interesting to notice that this experimental test is the first for a spatially extended system in the presence of a non linear potential. This is in contrast to the result of ref [15]. The reason of this discrepancy is that they neglect in the definition of the work (eq.21) the square term in ζ^2 . This cannot be done when a large ϵ_0 is used as in the case of ref.[15].

Our results are a preliminary study for future investigation on unsolved problems such as the applicability of Transient Fluctuation Theorem in an out-of-equilibrium system such as an aging system or a rapid quench.

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