

# Fluctuation dissipation and response in out of equilibrium systems

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# Outline

- Modified Fluctuation Dissipation Theorems (MFDTs) for Non Equilibrium Steady State (NESS)
- Fluctuation Theorem for systems driven out of equilibrium by a random force.

# Modified Fluctuation Dissipation Theorems (MFDT)

Motivation: Test in an experiment the new FDT for  
an out of equilibrium system

Outline :

- MFDT Three formulations: 1) Lagrangian FDT  
2) Frenesy FDT  
3) Generalized FDT
- Langevin dynamics
- Experimental realisation
- Results

# Fluctuation Dissipation Theorem (FDT)

In equilibrium FDT takes the form :

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

Observable  $O_t(\theta)$  of the dynamical process  $\theta$   
and its conjugated variable  $h$

Correlation function

$$C(t - s) = \langle O_t O_s \rangle$$

Response function to a delta perturbation of  $h$

$$R(t - s) = \langle \frac{\delta O_s}{h} \rangle$$

## Langevin Dynamics

$$\nu \dot{x} = -\partial_x U(x) + G + \eta$$

$$\text{with } \langle \eta(t)\eta(t') \rangle = 2k_B T \nu \delta(t - t')$$

$G$  non conservative force

with  $G = 0$  equilibrium FDT holds

with  $G \neq 0$  the system is driven into  
a non equilibrium steady state (**NESS**)  
where equilibrium FDT does not hold

# Modified Fluctuation Dissipation Theorem (MFDT) for NESS

**R. Chetrite, G. Falkovich, and K. Gawedzki, J. Stat. Mech.P08005 (2008).**

$$R^L(t, s) = \frac{1}{k_B T} \partial_s C^L(t, s)$$

$R^L$  and  $C^L$  are measured in the Lagrangian frame  
moving at mean local velocity  $v_0(\theta)$

The new observable  $O(t, \theta)$  evolves according to :

$$\partial_t O(t, \theta) + v_0(\theta) \cdot \nabla O(t, \theta) = 0$$

## MFDT for NESS

The MFDT in the lagrangian frame,

$$R^L(t, s) = -\frac{1}{k_B T} \partial_s C^L(t, s),$$

can also be written in the laboratory frame, replacing  $\partial_s$  in FDT with the convective derivative  $\partial_s + \nabla \cdot v_0(\theta)$

MFDT becomes:

This is the equality that we want to test

$$R(t - s) k_B T = \partial_s C(t - s) - b(t - s)$$

where

$$b(t - s) = \langle O(t, \theta) v_0(\theta(s)) \partial_\theta O(s, \theta) \rangle$$

## MFDT for NESS

In experiments it is much safer to use the integral form :

$$\chi(t-s) k_B T = [C(0) - C(t-s)] - B(t-s)$$

$\chi(t-s)$  is the integrated response and

$$B(t-s) = \int_0^s b(t-t') dt'$$

**We test this equality on the Langevin dynamics**

$$\gamma \dot{x} = -\partial_x U(x) + G + \eta$$

with  $\langle \eta(t)\eta(t') \rangle = 2k_B T \nu \delta(t-t')$

$G = \text{constant} \neq 0$  non conservative force



## Experiment with optical trap

Let us consider first the case with  $U = 0$

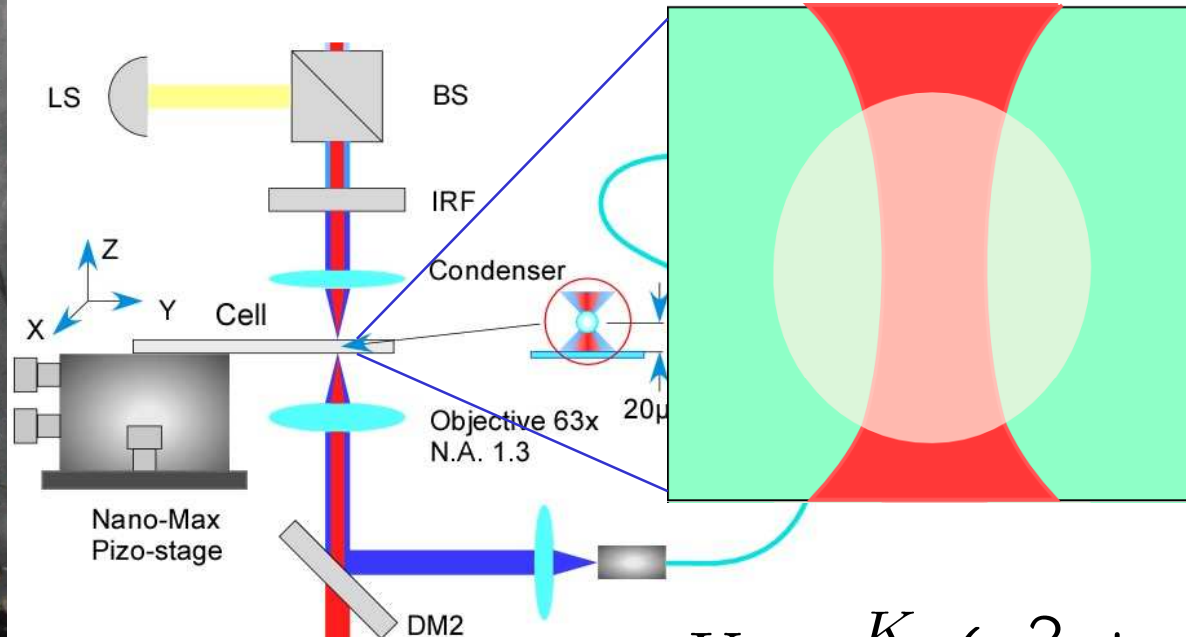
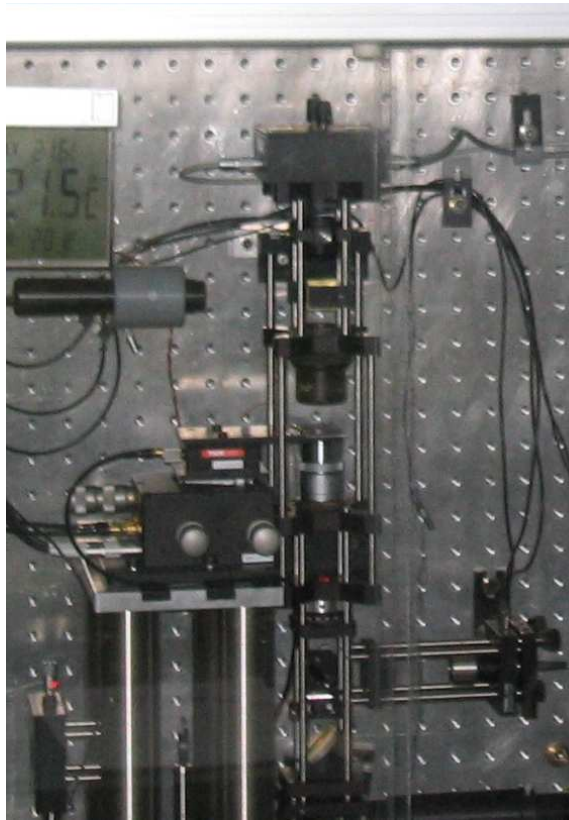
$$\nu \dot{x} = G + \eta$$

$G = \text{constant} \neq 0$  non conservative force

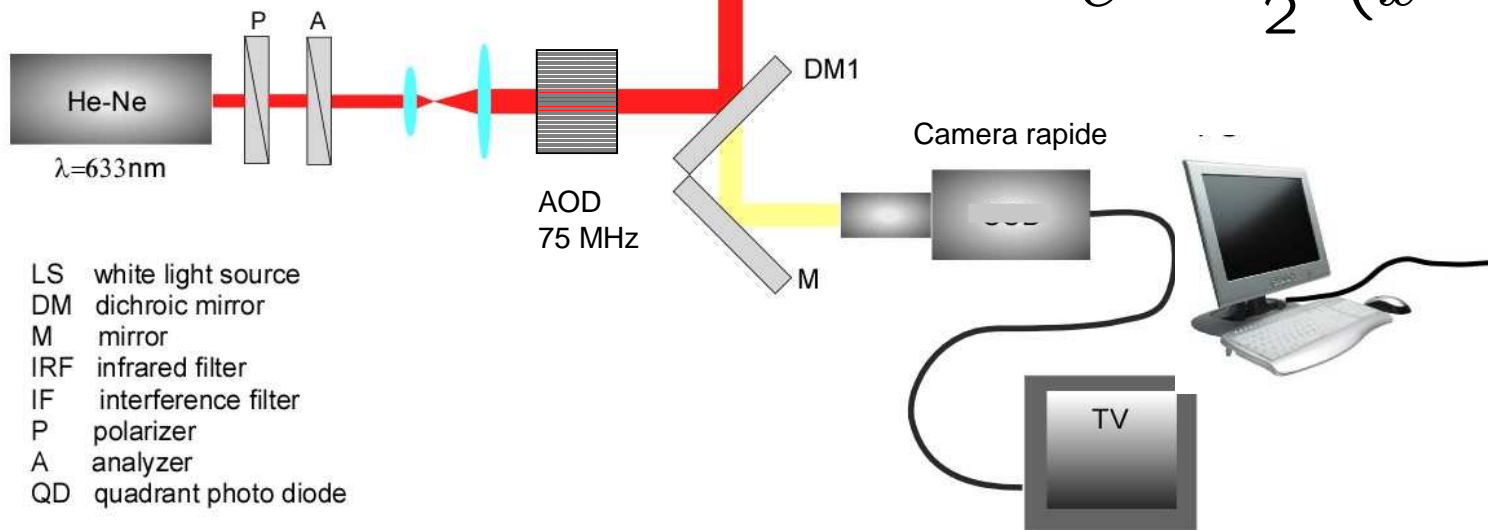
We use a Brownian particle confined in an optical trap

### The experimental set up

# Optical traps

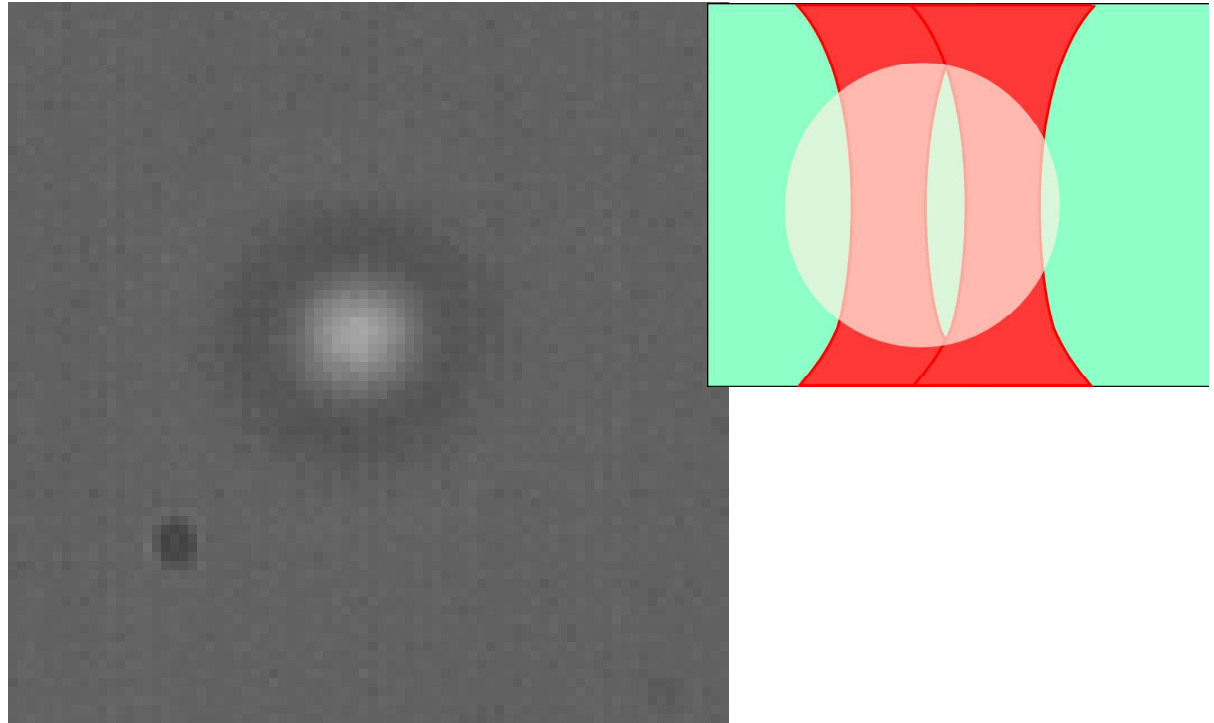
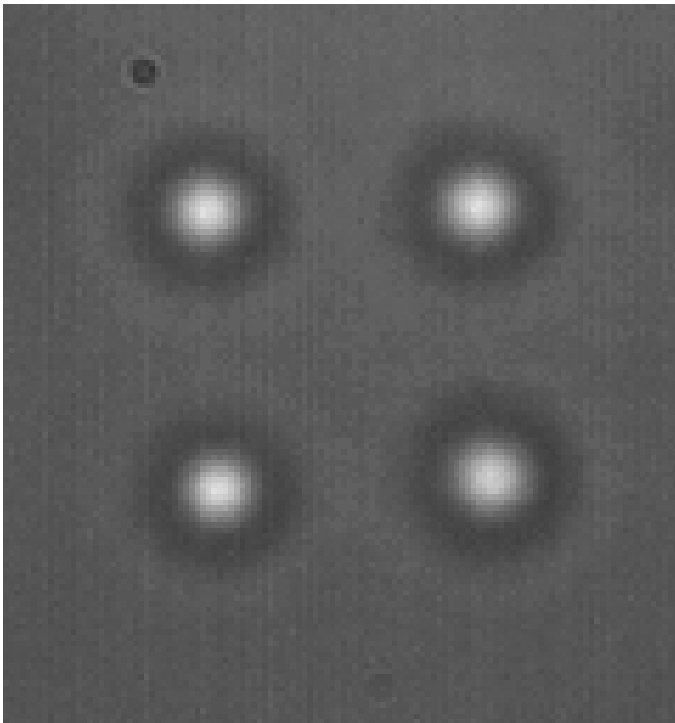


$$U = \frac{K}{2} (x^2 + y^2)$$



- LS white light source
- DM dichroic mirror
- M mirror
- IRF infrared filter
- IF interference filter
- P polarizer
- A analyzer
- QD quadrant photo diode

## Examples of traps



The Kramer rate is

$$r_k = \tau_0^{-1} \exp\left(-\frac{\delta U}{k_B T}\right)$$

## Experiment with optical trap

Let us consider first the case with  $U = 0$

$$\nu \dot{x} = G + \eta$$

$G = \text{constant} \neq 0$  non conservative force

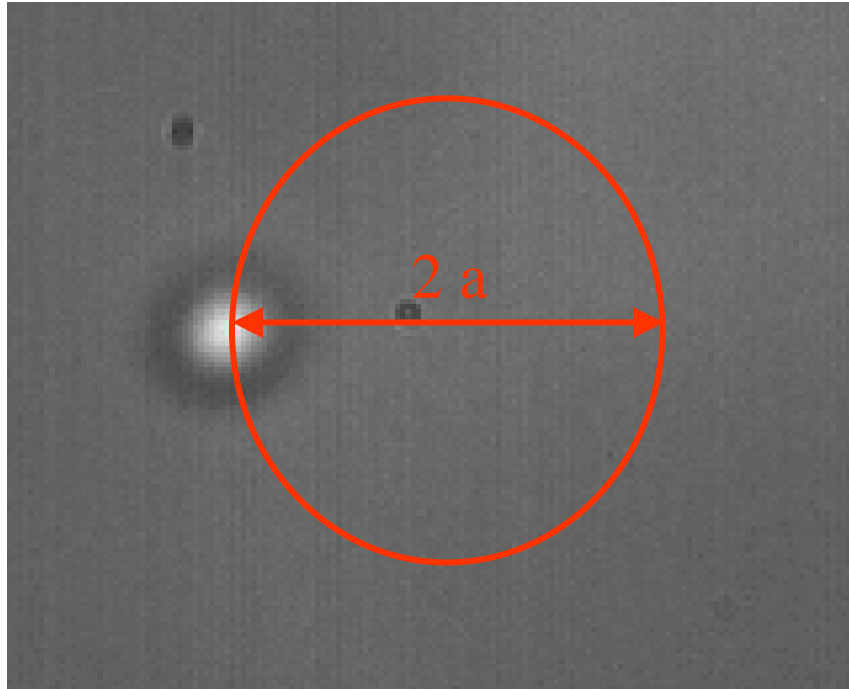
The motion of the particle is confined on a circle of radius  $a$

$$x = a \theta \quad \text{with} \quad 0 \leq \theta \leq 2\pi$$

This is achieved by a circular sweeping of the laser beam

How  $G$  is obtained ?

## Particle motion with $U=0$



$$\nu a \dot{\theta} = G + \eta$$

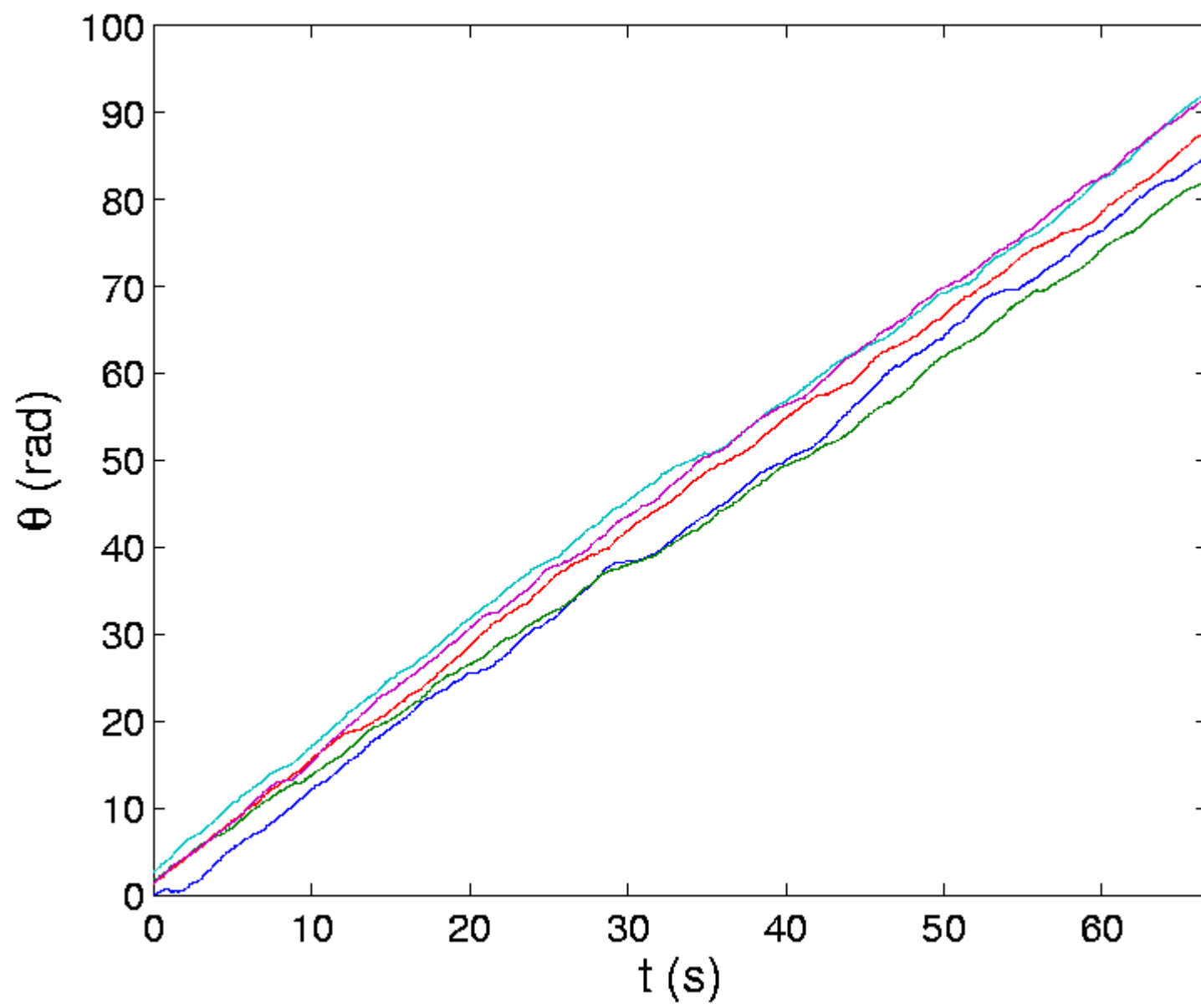
$$\nu = 6\pi r \eta$$

$$v_0 = \langle \dot{\theta} \rangle = G/\nu$$

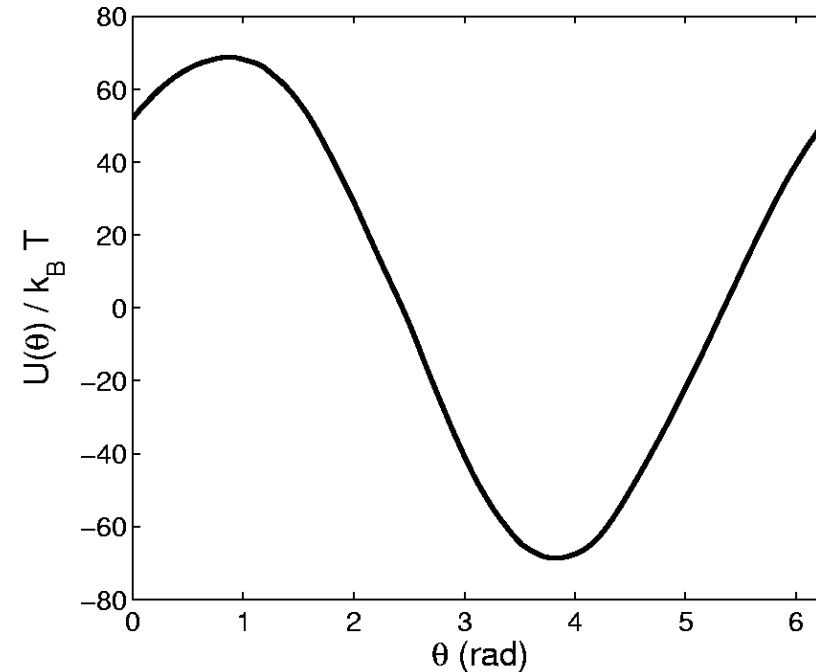
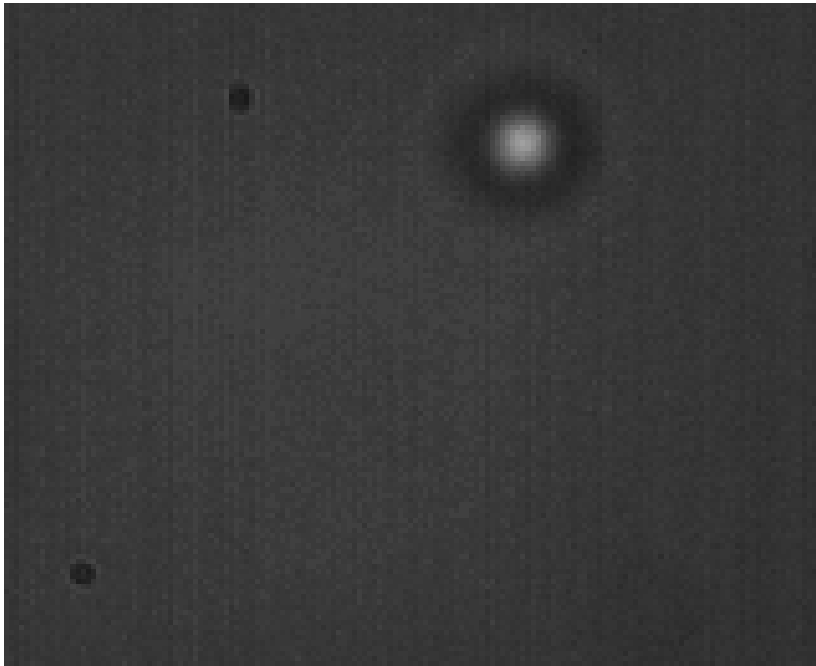
$a = 4.5\mu m$ ,  $r = 1\mu m$ ,  $\eta = 10^{-3} Pa$ ,

We obtain  $G = 6.60 \cdot 10^{-14} N$  from

the measure of  $v_0 = 0.85 rad/s$



# Particle motion with potential



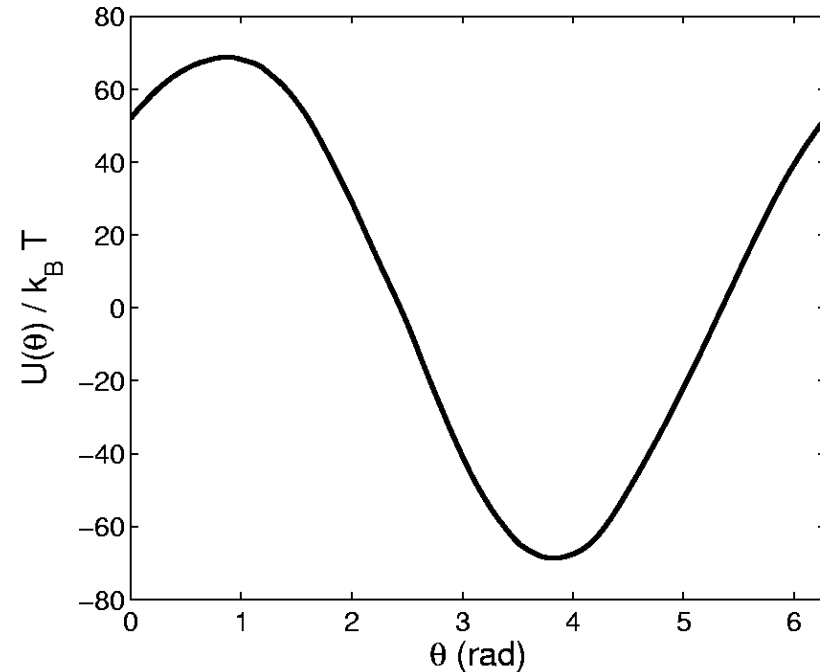
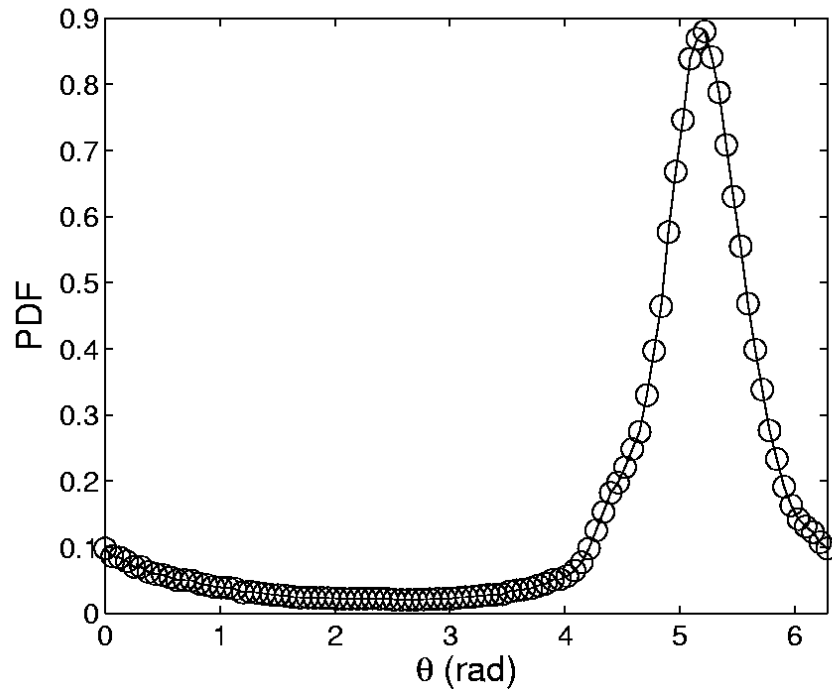
Periodic potential  $U = A \sin(\theta + \varphi)$

$$\nu a^2 \dot{\theta} = -\partial_{\theta} U(\theta) + F + \eta a$$

$F = G a = \text{constant non conservative torque}$

The potential  $U$  is produced by a modulation (5%) of the laser intensity

## PDF of the particle position

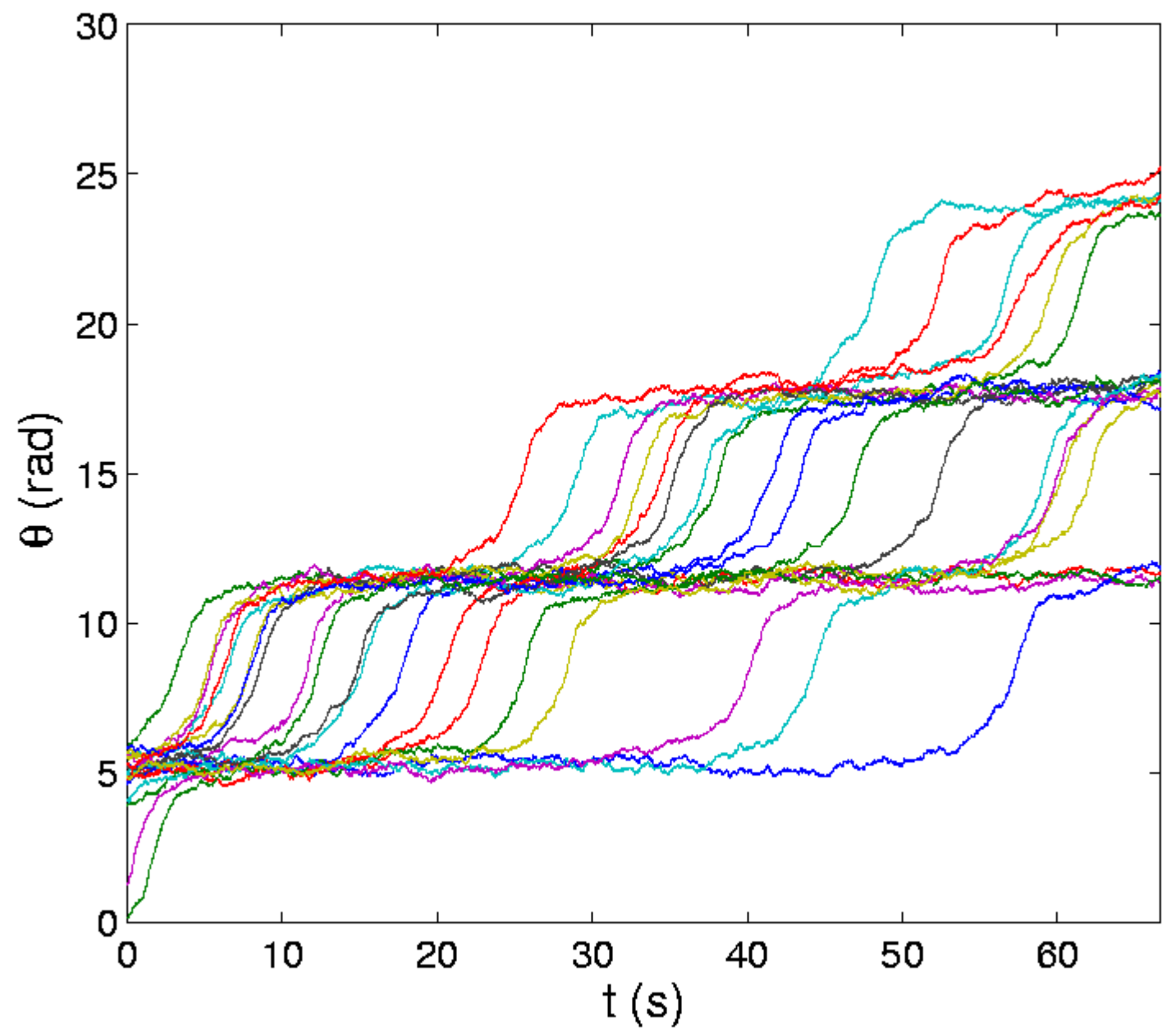


$j$  = probability current =  $\langle \dot{\theta} \rangle_0 / (2\pi)$

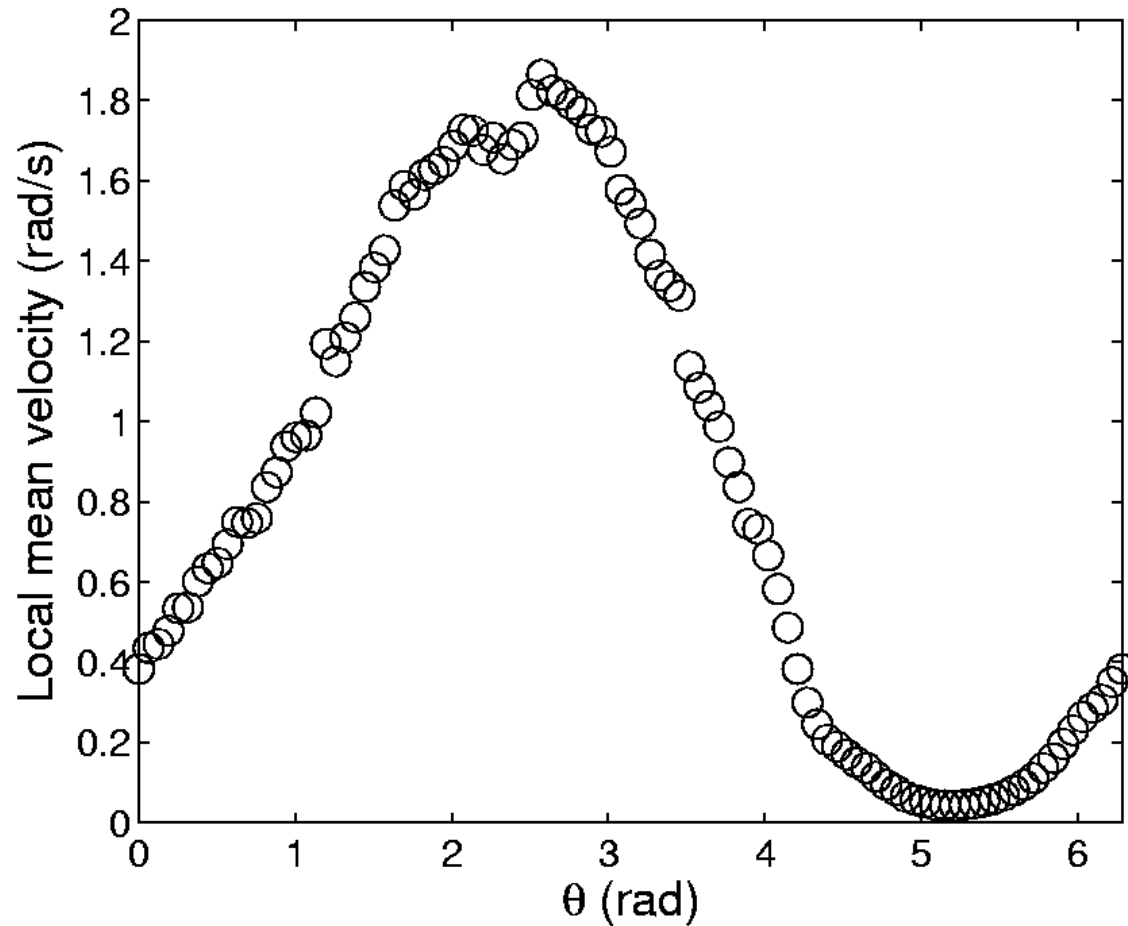
$\rho(\theta)$  = probability density

$$v_0(\theta) = \frac{j}{\rho(\theta)}$$





## Local mean velocity



$j$  = probability current =  $\langle \dot{\theta} \rangle_0 / (2\pi)$

$\rho(\theta)$  = probability density

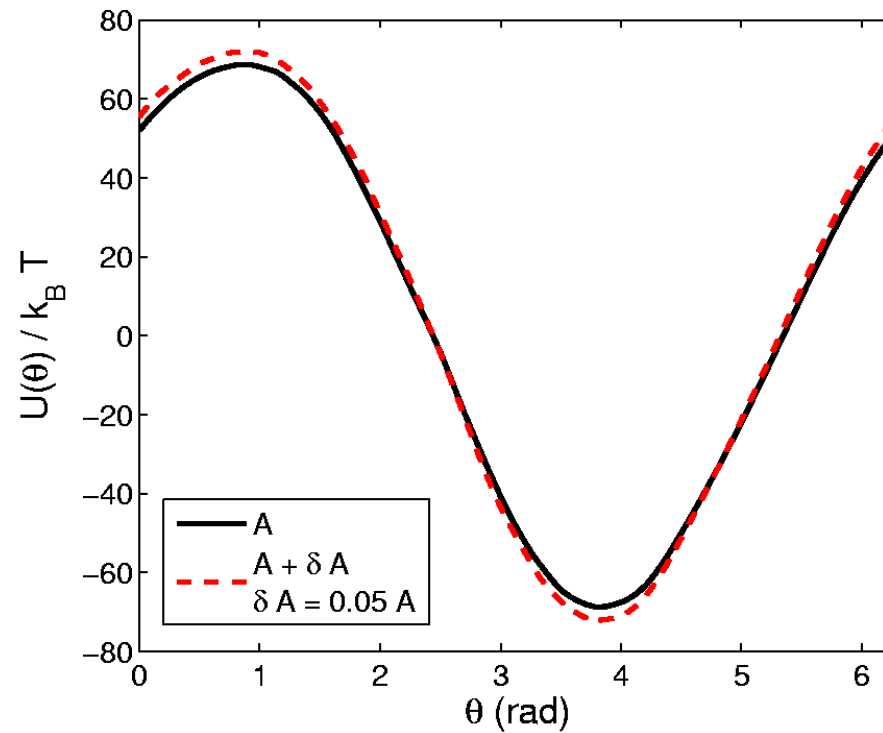
$$v_0(\theta) = \frac{j}{\rho(\theta)}$$

## The observable $O(\theta)$

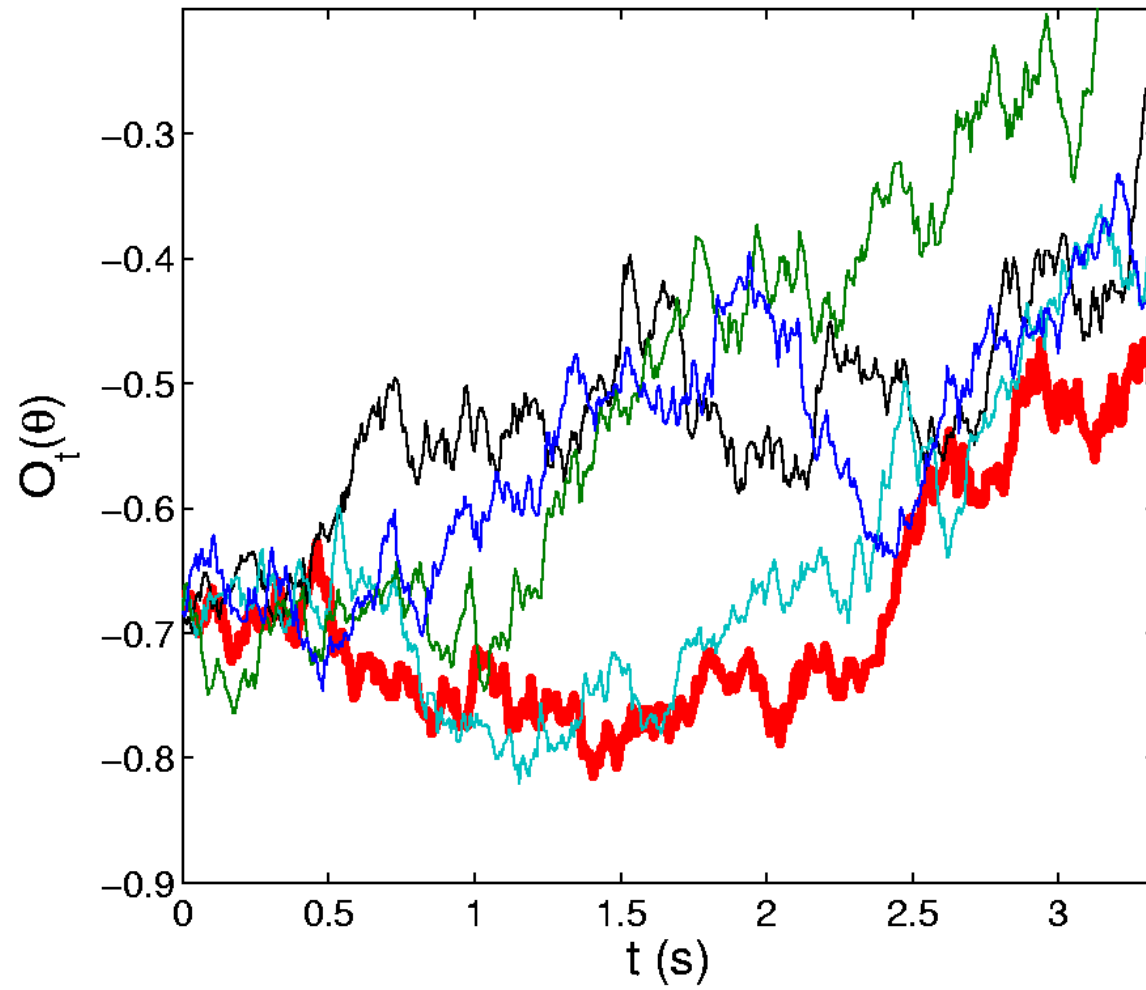
To measure the response, the perturbation is applied in the following way:

$$U(\theta) \longrightarrow U(\theta) + \delta A \sin(\theta + \varphi)$$

Thus the observable is  $O(\theta(t)) = \sin(\theta(t) + \varphi)$



# Time evolution of $O(t)$



## MFDT for NESS

The observable is :  $O(\theta(t)) = \sin(\theta(t) + \varphi)$

$$\chi(t) k_B T = [C(0) - C(t)] - B(t)$$

with

$$B(t) = \int_0^t \langle O(\theta(t')) v_0(\theta(t')) \partial_\theta O(\theta(0)) \rangle dt'$$

$$C(t) = \langle O(\theta(t)) O(\theta(0)) \rangle$$

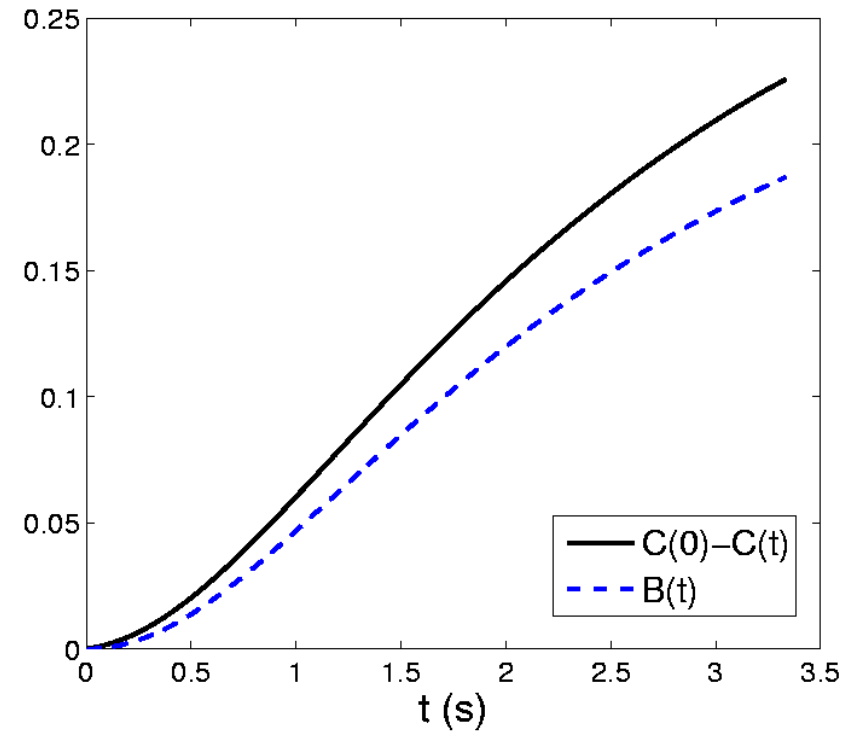
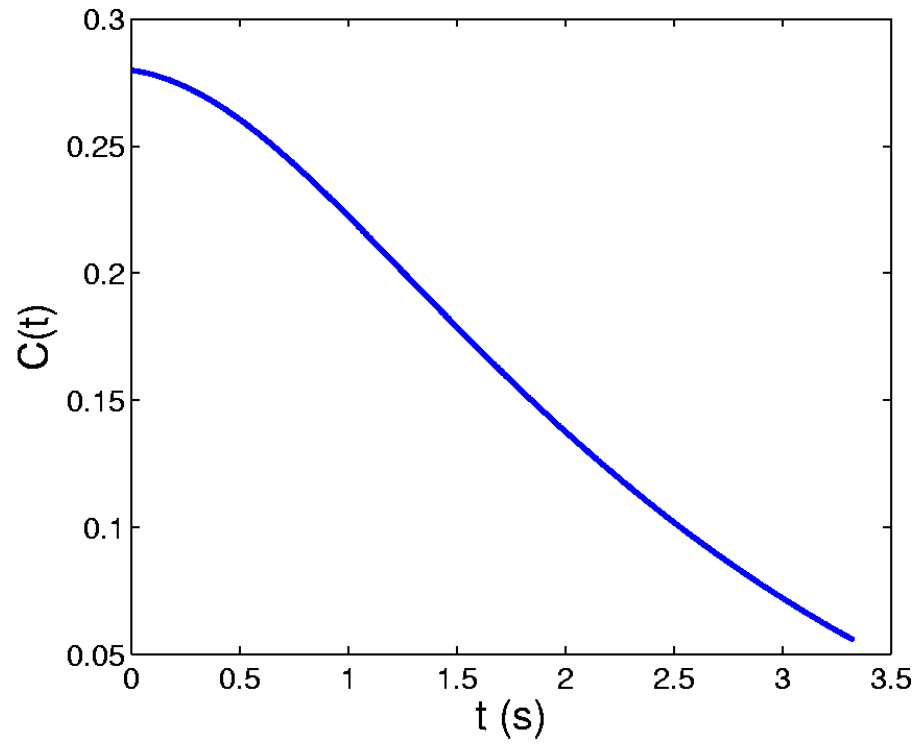
The integrated response  $\chi(t)$

to a Heaviside perturbation of  $A$ , switched on at  $t = 0$ , is:

$$\chi(t) = \frac{\langle O(\theta(t))_{\delta A} - O(\theta(t + t^*))_0 \rangle}{\delta A}$$

such that  $O(\theta(0))_{\delta A} = O(\theta(t^*))_0$

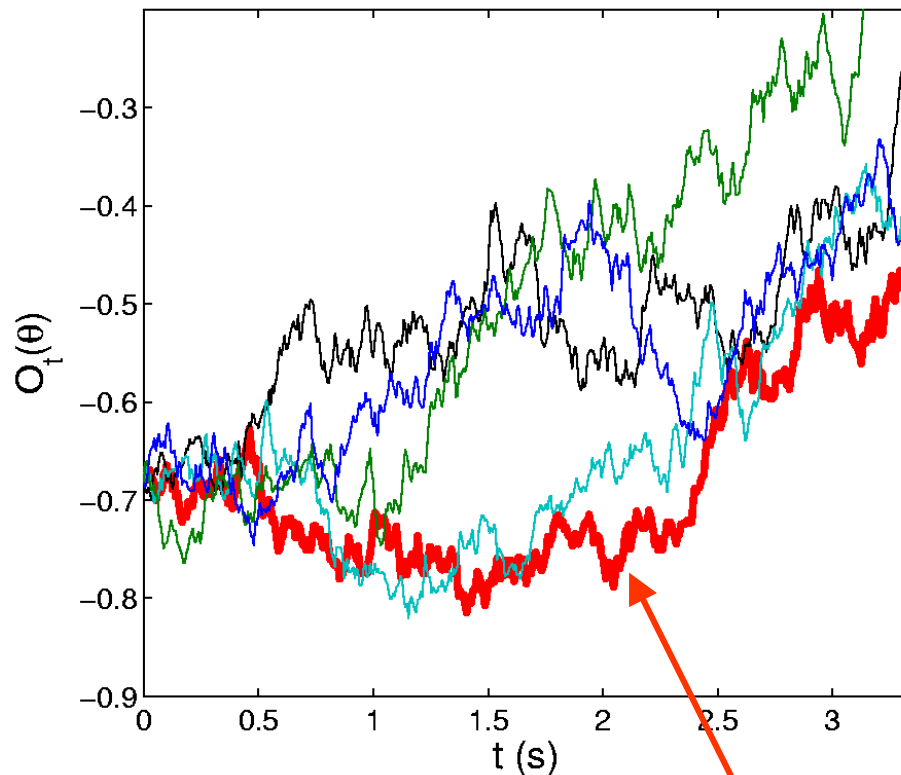
# Correlation function and B(t)



## The integrated response

to a Heaviside perturbation of  $A$ , switched on at  $t = 0$ , is:

$$\chi(t) = \frac{\langle O(\theta(t))_{\delta A} - O(\theta(t + t^*))_0 \rangle}{\delta A}, \text{ with } O(\theta(0))_{\delta A} = O(\theta(t^*))_0$$



- Select 200 unperturbed trajectories such that at time  $t^*$

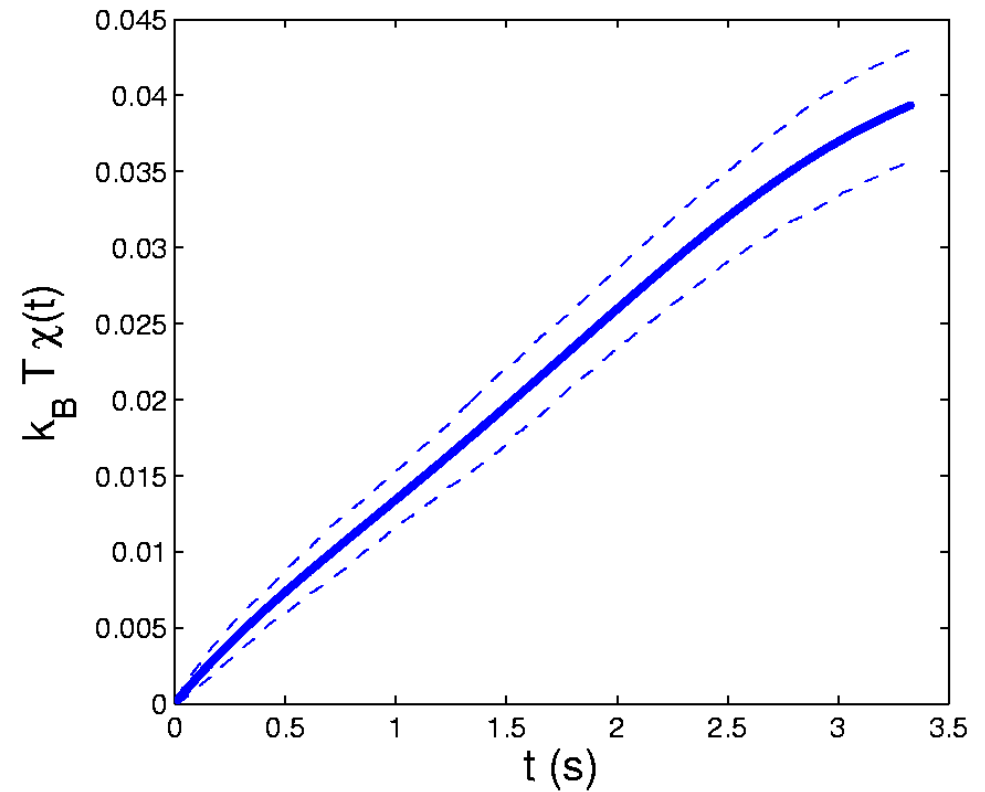
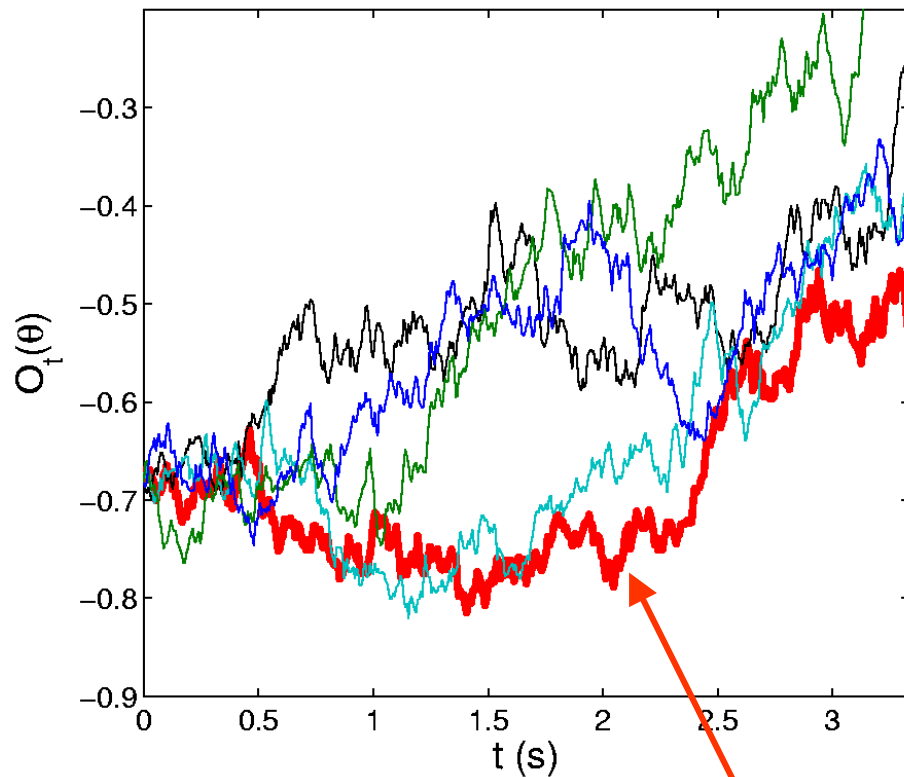
$$O(\theta(0))_{\delta A} = O(\theta(t^*))_0$$

- Compute the mean of  $O(\theta(t))_{\delta A} - O(\theta(t + t^*))_0$  on the 200 trajectories
- Repeat the procedure on several perturbations and make the average

## The integrated response

to a Heaviside perturbation of  $A$ , switched on at  $t = 0$ , is:

$$\chi(t) = \frac{\langle O(\theta(t))_{\delta A} - O(\theta(t + t^*))_0 \rangle}{\delta A}, \text{ with } O(\theta(0))_{\delta A} = O(\theta(t^*))_0$$

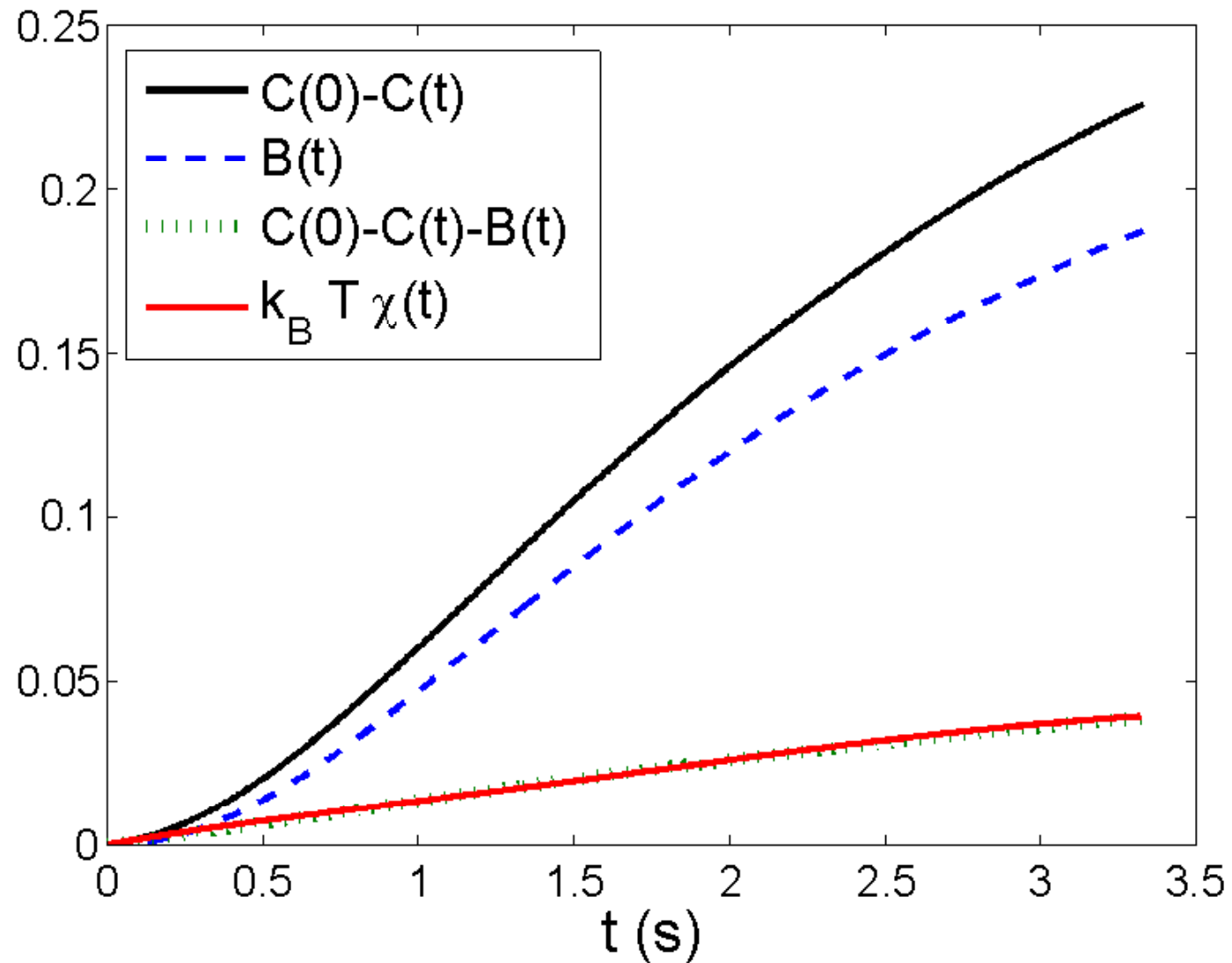


**Perturbed trajectory**



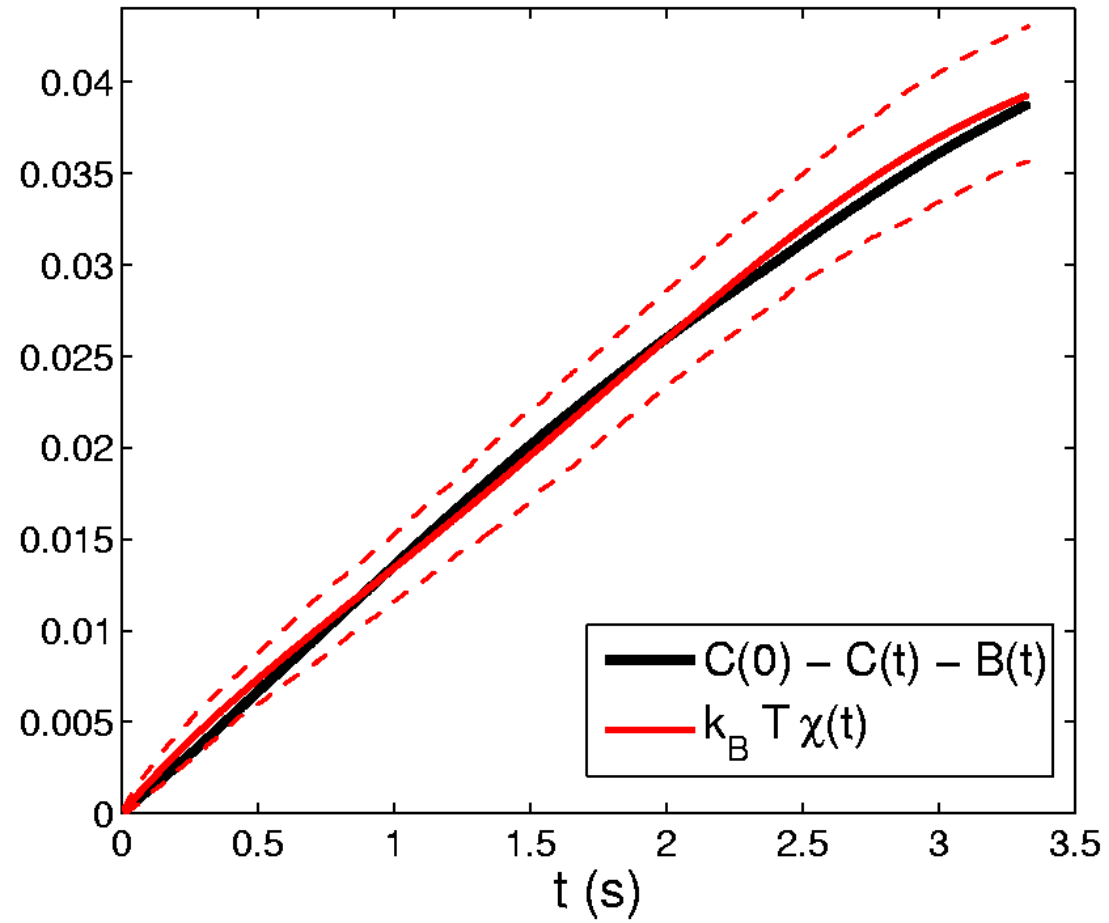
## MFDT

$$\chi(t) k_B T = [C(0) - C(t)] - B(t)$$



# MFDT

$$\chi(t) k_B T = [C(0) - C(t)] - B(t)$$



# Conclusions on MFDT

based on a Lagrangian formulation

- We have shown that MFDT holds for a bead moving in a toroidal optical trap and submitted to a non conservative force and to a periodic potential.
- The results can be interpreted as an equilibrium like **FDT in the Lagrangian frame moving at the velocity** determined by the probability current.
- **The main drawback of the method** is the knowledge of the probability current which is not always obvious to measure.

## MFDT based on frenesy

E. Lippiello, F. Corberi, and M. Zannetti, Phys. Rev.E, 71 (036104) 2005.

M. Baiesi, C. Maes and B. Wynants, Phys. Rev. Lett., 103 (010602) 2009.

$$U(q) \rightarrow U(q) - h_s V(q)$$

$$\chi_{OV}(t) = \int_0^t R_{OV}(t, s) ds = \frac{\langle O(q_t) \rangle_h - \langle O(q_t) \rangle_0}{h},$$

$$\chi_{QV}(t) = \frac{\beta}{2} [C_{OV}(0) - C_{OV}(t) + K(t)],$$

$$C_{OV}(t) = \langle V(q_0) O(q_t) \rangle_0,$$

$$K(t) = - \int_0^t \langle LV(q_s) O(q_t) \rangle_0 ds,$$

The frenesy  $\beta LV(q)$  can be regarded as a generalized escape rate of a trajectory from a given phase-space point  $q$  (*Baiesi et al*).

## MFDT based on frenesy

### In our experiment

$$\nu a^2 \dot{\theta} = -A \frac{\partial O(\theta)}{\partial \theta} + F + \eta a$$

$$U(\theta) = A O(\theta) \text{ with } O(\theta) \simeq \sin(\theta + \varphi)$$

and  $V(\theta) = U(\theta)$

$$U(\theta) \rightarrow U(\theta) - \frac{\delta A}{A} U(\theta)$$

$$\chi_{OV}(t) = \frac{A(\langle O(\theta(t)) \rangle_h - \langle O(\theta(t+t^*)) \rangle_0)}{\delta A} = A \chi(t)$$

$$\text{with } O(\theta(0))_{\delta A} = O(\theta(t^*))_0$$

## MFDT based on frenesy

$$\chi_{QV}(t) = \frac{\beta}{2}[C_{OV}(0) - C_{OV}(t) + K(t)],$$

$$K(t) = - \int_0^t \langle LV(q_s)O(q_t) \rangle_0 ds,$$

For the Langevin dynamics of  $\theta$  the analytical expression of the generator  $L$  is

$$L = \frac{1}{\nu a^2} [(F - AO'(\theta))\partial_\theta + k_B T \partial_\theta^2].$$

Hence in this case

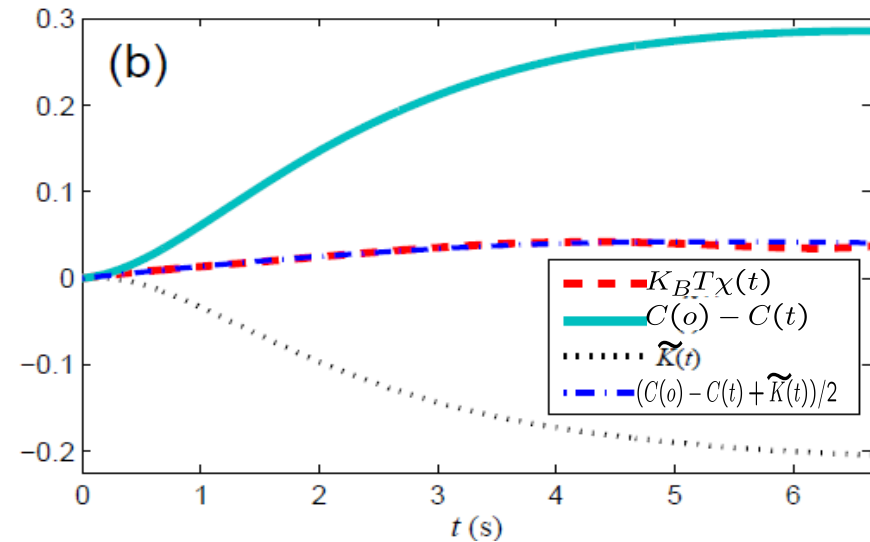
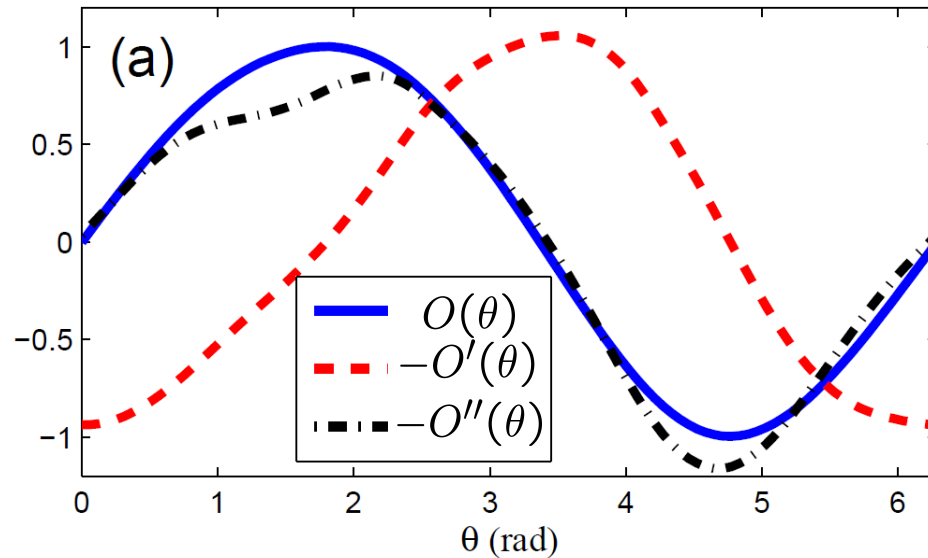
$$k_B T \chi(t) = \frac{C(0) - C(t) + \tilde{K}(t)}{2},$$

where the entropic and frenetic terms are

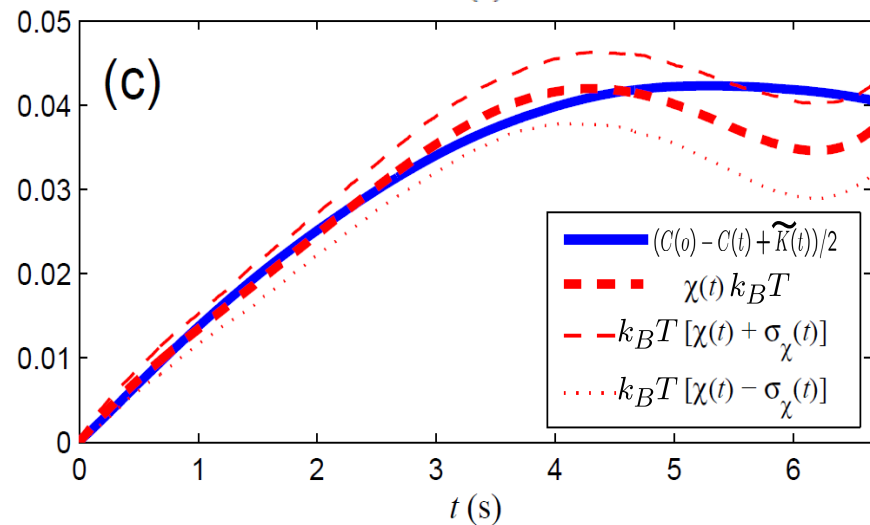
$$A C(t) = A \langle O(\theta_0)O(\theta_t) \rangle_0 = C_{OV}(t)$$

$$K(t) = A\tilde{K}(t) = -\frac{A}{\nu a^2} \int_0^t ds \langle [k_B T O''(\theta_s) + (F - A O'(\theta_s)) O'(\theta_s)] O(\theta_t) \rangle_0,$$

## Results on the FDT based on frenesy



- The method is sensitive to the values of the experimental derivative of the potential
- Two drawbacks:
  - a) the knowledge of the generator of the dynamics;
  - b) The Markofian nature of the system



# Generalized FDT

The starting point is the Hatano-Sasa relation for Markofian process

$$\rho_{ss}(c, \lambda^{ss}) = \exp[-\phi(c, \lambda^{ss})].$$

Probability density  
for a NESS

$$\phi(c; \lambda) = -\log[\rho_{ss}(c; \lambda)]$$

Pseudo-potential

$$\left\langle \exp \left\{ - \int_{t_i}^{t_f} dt \dot{\lambda}_\alpha(t) \frac{\partial \phi(c(t); \lambda(t))}{\partial \lambda_\alpha} \right\} \right\rangle = 1,$$

Hatano-Sasa

The average is taken over a large number of realizations of a given dynamical process defined by the variation of  $\lambda(t)$



## Generalized FDT

$$\left\langle \exp \left\{ - \int_{t_i}^{t_f} dt \dot{\lambda}_\alpha(t) \frac{\partial \phi(c(t); \lambda(t))}{\partial \lambda_\alpha} \right\} \right\rangle = 1,$$

Consider small variations of the control parameters around a steady-state value  $\lambda^{\text{ss}}$ .

$$\delta \lambda(t) = \lambda(t) - \lambda^{\text{ss}} \quad \text{with} \quad \delta \lambda(t_i) = 0$$

Expanding to second order in the integrand and taking into account the normalisation conditions one obtains:

## Generalized FDT

$$\left\langle \frac{\partial \phi(c(t); \lambda^{ss})}{\partial \lambda_\alpha} \right\rangle = \int_{t_i}^t R_{\alpha\gamma}(t - t') \delta \lambda_\gamma(t') dt',$$

$$\begin{aligned} R_{\alpha\gamma}(t - t') &= \frac{d}{dt} C_{\alpha\gamma}(t - t') \\ &= \frac{d}{dt} \left\langle \frac{\partial \phi(c(t); \lambda^{ss})}{\partial \lambda_\alpha} \frac{\partial \phi(c(t'); \lambda^{ss})}{\partial \lambda_\gamma} \right\rangle_{ss}. \end{aligned}$$

Where now  $\langle \cdot \rangle_{ss}$  is computed on the stationary state

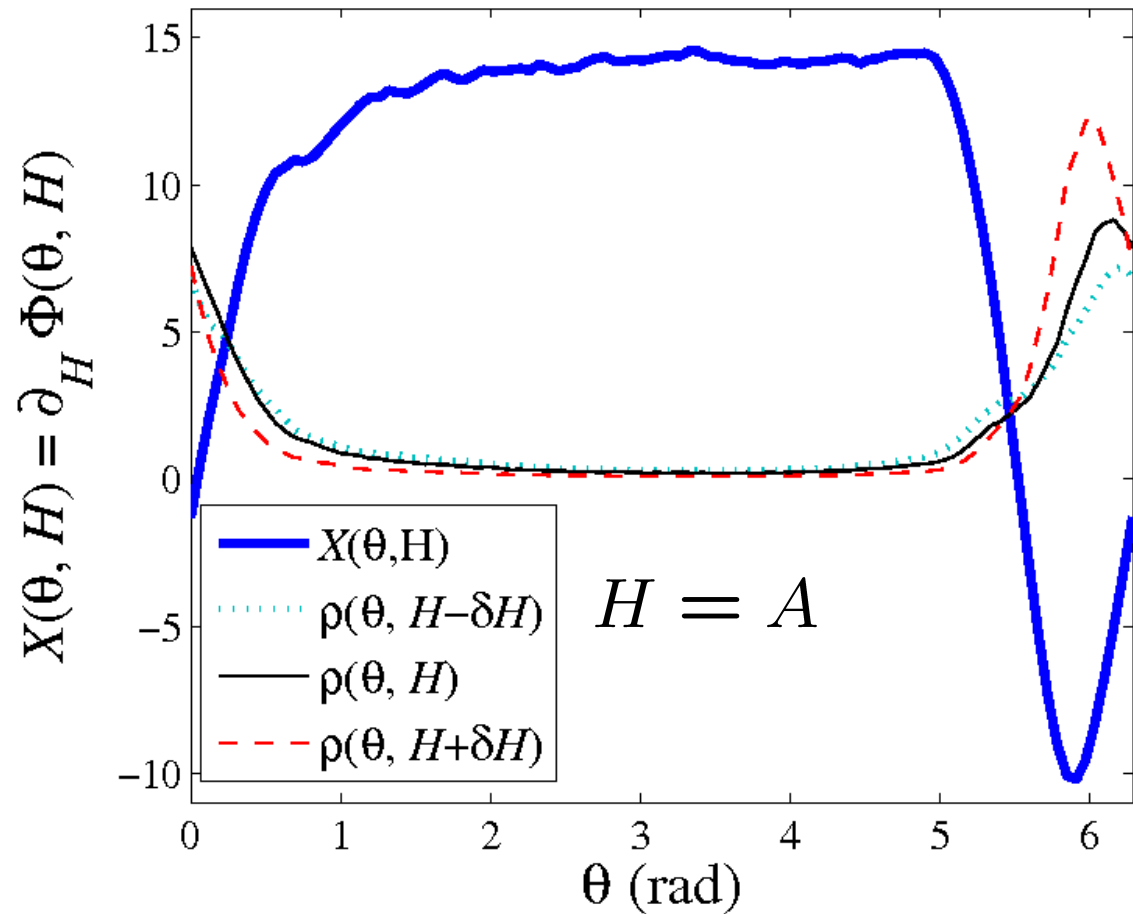
$$X_\alpha(t) = \frac{\partial \phi(t)}{\partial \lambda_\alpha} \text{ is the observable}$$

J. Prost, J.F. Joanny, J.M. Parrondo, PRL 103, 090601 (2009)

# Generalized FDT

In the experiment

$$X(t) = \frac{\partial \phi(\theta(t), A)}{\partial A}$$

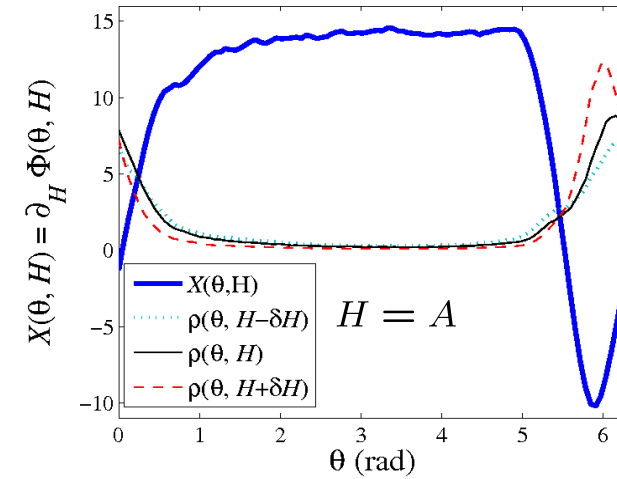
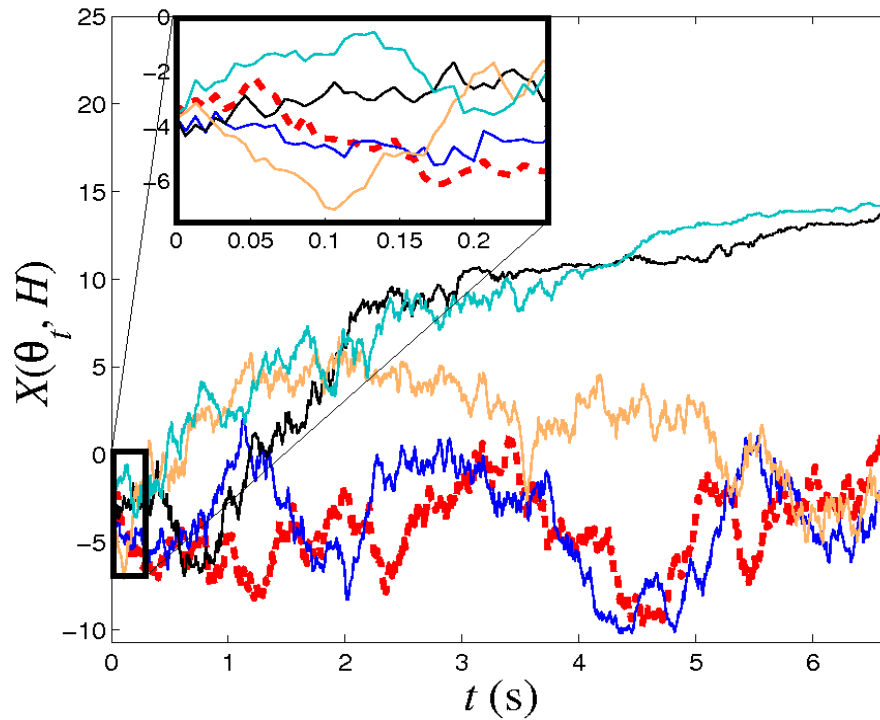


# Generalized FDT

In the experiment

$$X(t) = \frac{\partial \phi(\theta(t), A)}{\partial A}$$

Time evolution of X



# Generalized FDT

The response function in the experiment

$$\left\langle \frac{\partial \phi(\theta(t))}{\partial A} \right\rangle = \int_0^t R(t-t') \delta A(t') dt'$$

If  $\delta A$  is an Heaviside perturbation

$$\left\langle \frac{\partial \phi(\theta(t))}{\partial A} \right\rangle \neq (C(0) - C(t)) \delta A$$

where

$$C(t) = \left\langle \frac{\partial \phi(\theta(t))}{\partial A} \frac{\partial \phi(\theta(0))}{\partial A} \right\rangle_{ss}$$

Experimentally it does not work

## Generalized FDT

The response function in the experiment

$$\left\langle \frac{\partial \phi(\theta(t))}{\partial A} \right\rangle = \int_0^t R(t - t') \delta A(t') dt'$$

$$\left\langle \frac{\partial \phi(\theta(t))}{\partial A} \right\rangle - \left\langle \frac{\partial \phi(\theta(t))}{\partial A} \right\rangle_{ss} = \int_0^t R(t - t') \delta A(t') dt'$$

This is zero in the case of infinite sampling

In the case of finite sampling two effects has to be taken into account

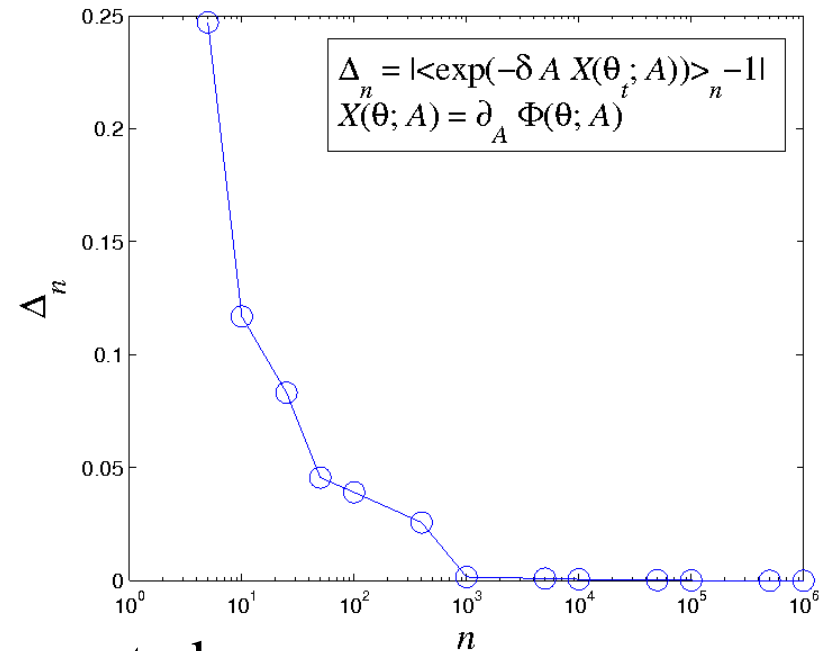
# Generalized FDT

In the case of finite sampling two effects has to be taken into account

The Hatano-Sasa relation  
is not exactly 1

and

$$\left\langle \frac{\partial \phi(\theta(t))}{\partial A} \right\rangle_{ss} \neq 0$$



The integrated response must be computed :

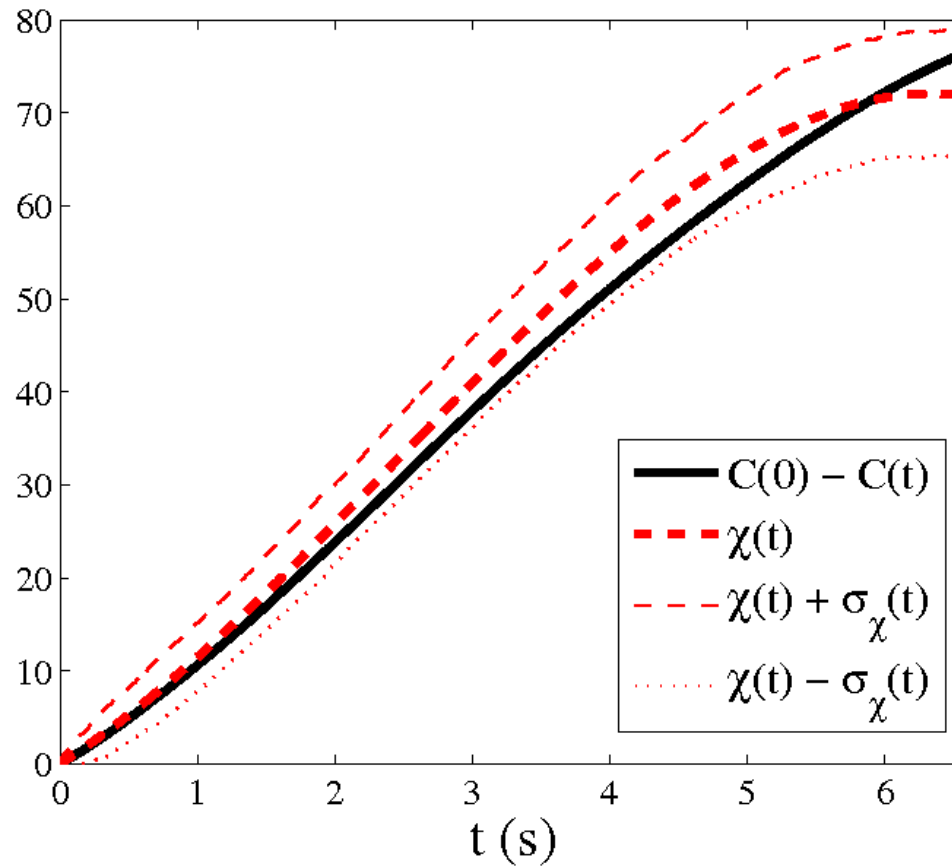
$$\frac{\left\langle \frac{\partial \phi(\theta(t))}{\partial A} \right\rangle - \left\langle \frac{\partial \phi(\theta(t+t^*))}{\partial A} \right\rangle_{ss}}{\delta A} = \chi(t)$$

under the contion  $\left\langle \frac{\partial \phi(\theta(0))}{\partial A} \right\rangle = \left\langle \frac{\partial \phi(\theta(t^*))}{\partial A} \right\rangle_{ss}$

# Generalized FDT

$$C(t) = \left\langle \frac{\partial \phi(\theta(t))}{\partial A} \frac{\partial \phi(\theta(0))}{\partial A} \right\rangle_{ss}$$

$$\chi(t) = (C(0) - C(t))$$





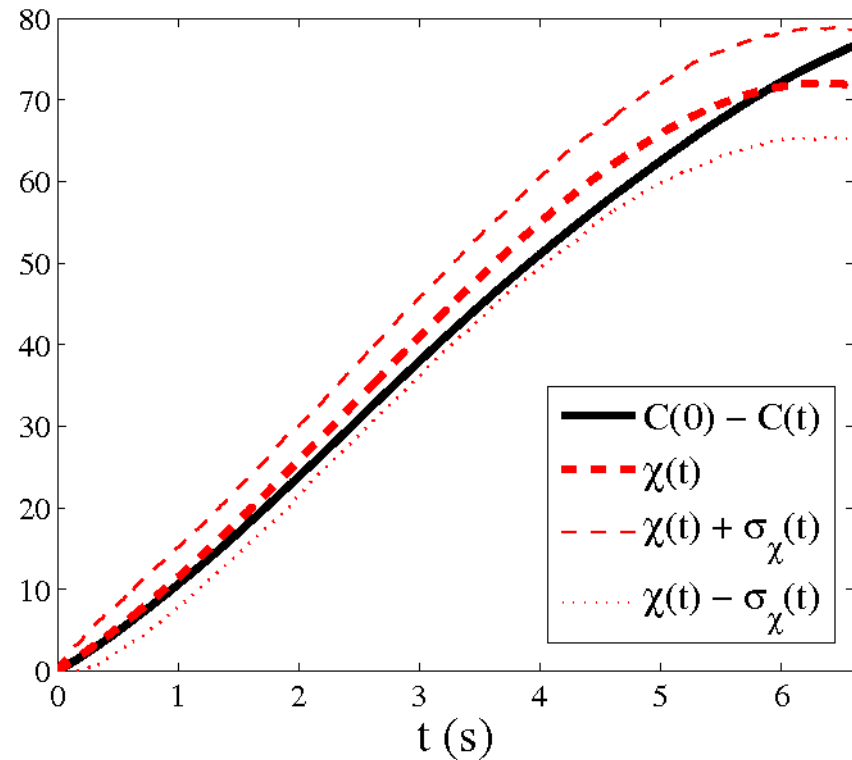
# Generalized FDT

$$C(t) = \left\langle \frac{\partial \phi(\theta(t))}{\partial A} \frac{\partial \phi(\theta(0))}{\partial A} \right\rangle_{ss}$$

$$\chi(t) = (C(0) - C(t))$$

## Conclusions

- GFDT as been checked on the experimental data taking into account the finite sampling.
- It is certainly the more general formulation for a Markofian dynamics



## Conclusions on FDT on NESS

- We checked three formulations of FDT for NESS
- The Lagrangian formulation can be applied to any process but the knowledge of the current is needed
- The frequency formulation needs the knowledge of the generator of the dynamics, but it can in principle be applied to non-stationary cases. It is limited to Markovian systems
- The formulation based on Hatano-Sasa relation is certainly the most general of the three if the dynamics is Markovian

## FR for NESS driven by a random forcing

Motivation :

We consider a Langevin dynamics driven out of equilibrium

$$m\ddot{x} + \gamma\dot{x} = -kx + \zeta_T + f_0.$$

and

$$w_\tau = \frac{1}{k_B T} \int_t^{t+\tau} \dot{x}(t') f_0(t') dt'.$$

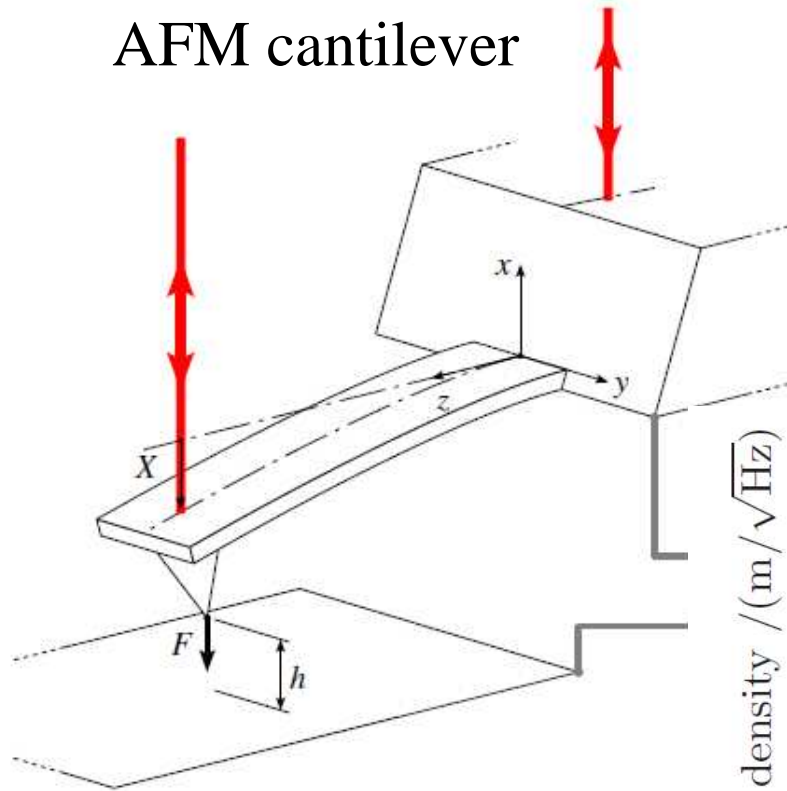
if  $f_0$  is deterministic the Fluctuation Theorem holds.

$$\ln \frac{P(W_\tau = W)}{P(W_\tau = -W)} \rightarrow \frac{W}{k_B T}, \quad \tau \rightarrow \infty$$

What happens if the external force is random ?

# FR for NESS driven by a random forcing

AFM cantilever



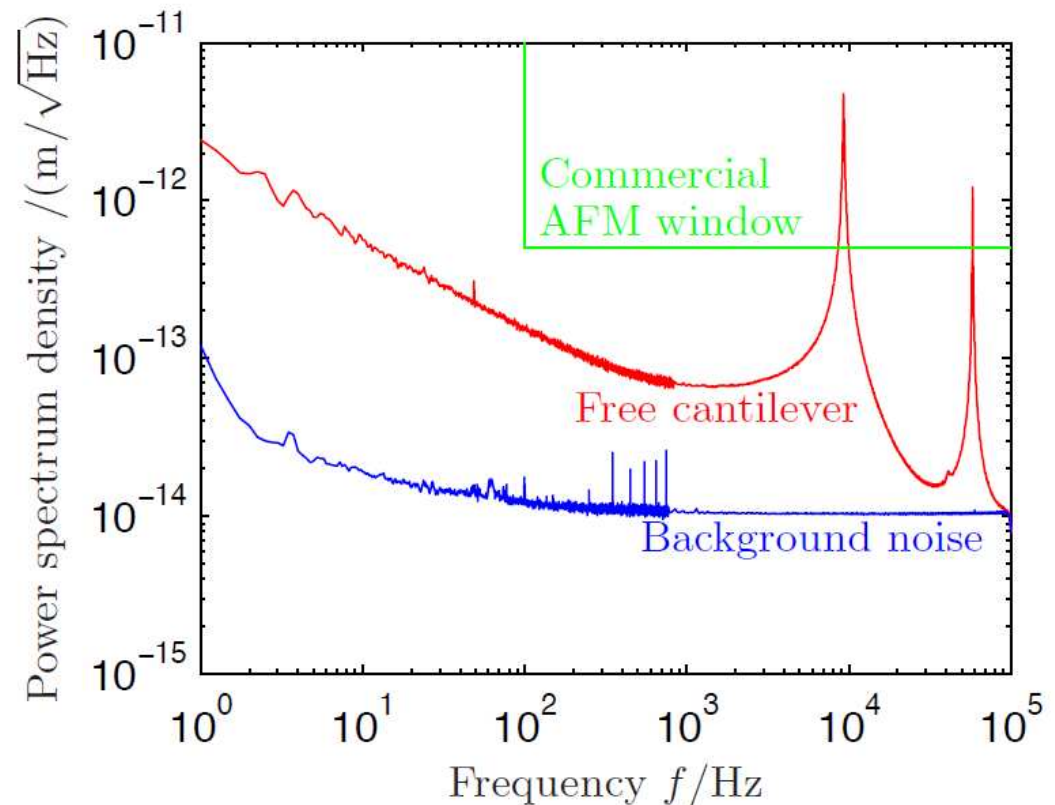
$$E_c = \frac{1}{2} C(X) V^2$$

$$F = -\partial_X E_c = -aV^2$$

$$a = 15 \text{ pN/V}^2$$

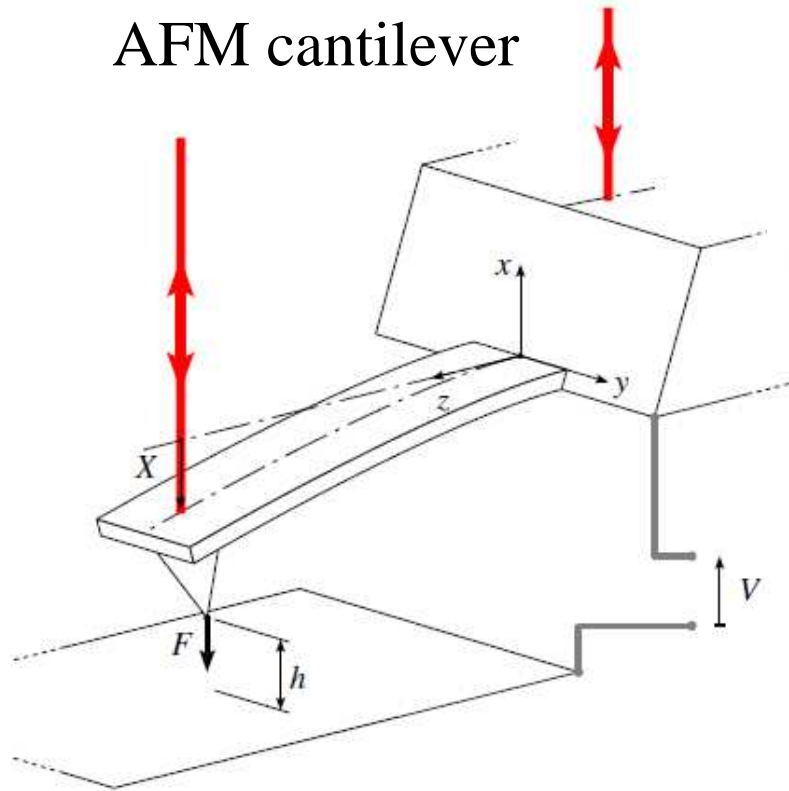
The experiment

$$m\ddot{X} + \gamma\dot{X} = -kX + \zeta_T$$



## FR for NESS driven by a random forcing

AFM cantilever



$$E_c = \frac{1}{2} C(X) V^2$$

$$F = -\partial_X E_c = -aV^2$$

$$a = 15 \text{ pN/V}^2$$

The experiment

$$m\ddot{X} + \gamma\dot{X} = -kX + \zeta_T + F$$

$$F = \bar{F} + f_0$$

$$x = X - \bar{X} \quad \text{with} \quad \bar{X} = \bar{F}/k$$

$$m\ddot{x} + \gamma\dot{x} = -kx + \zeta_T + f_0.$$

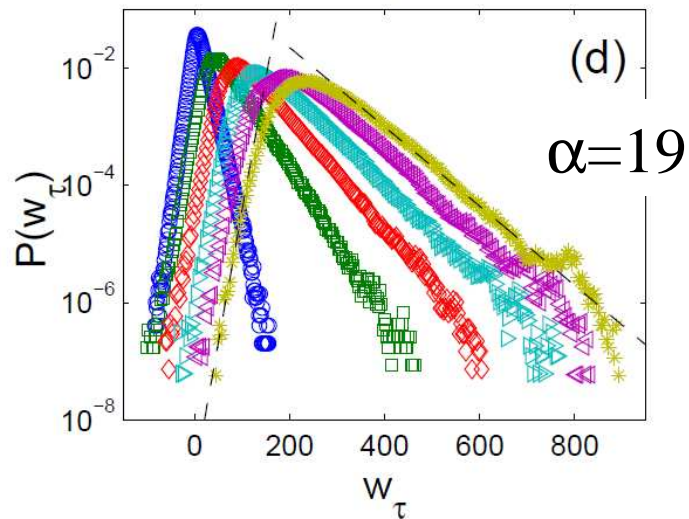
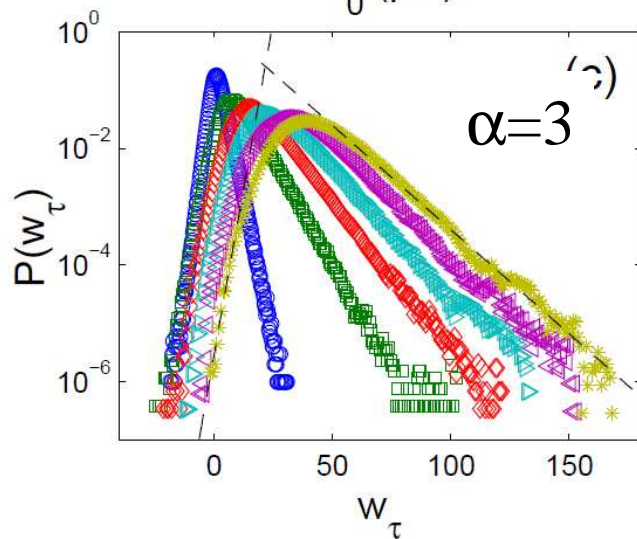
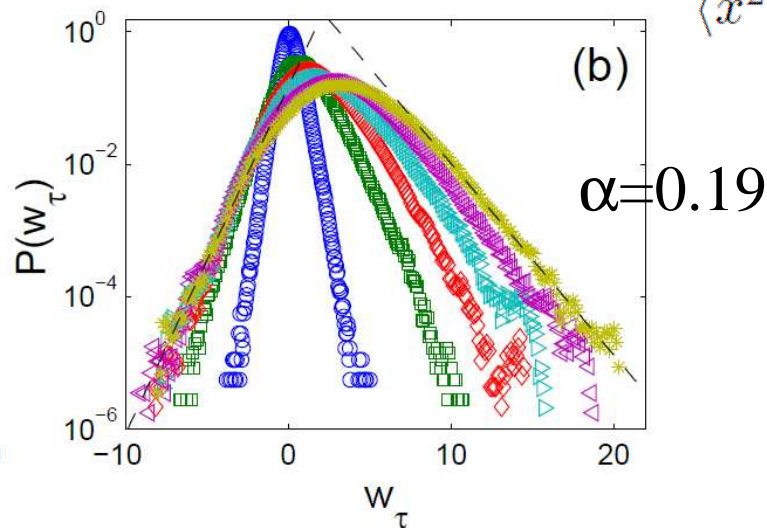
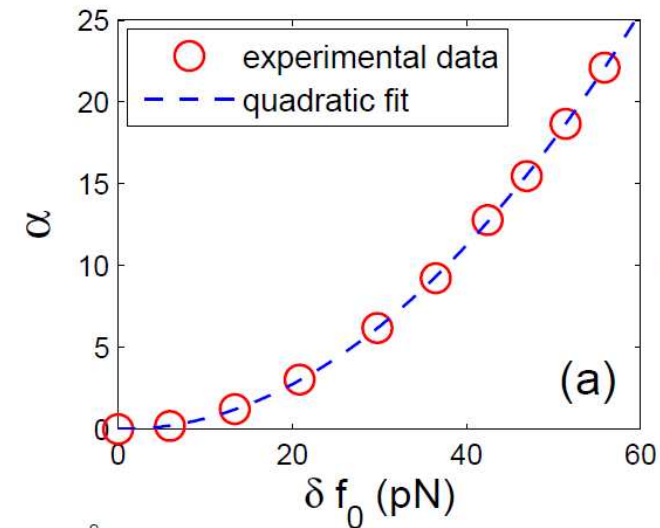
$f_0$  is random Gaussian force with white spectrum

# FR for NESS driven by a random forcing

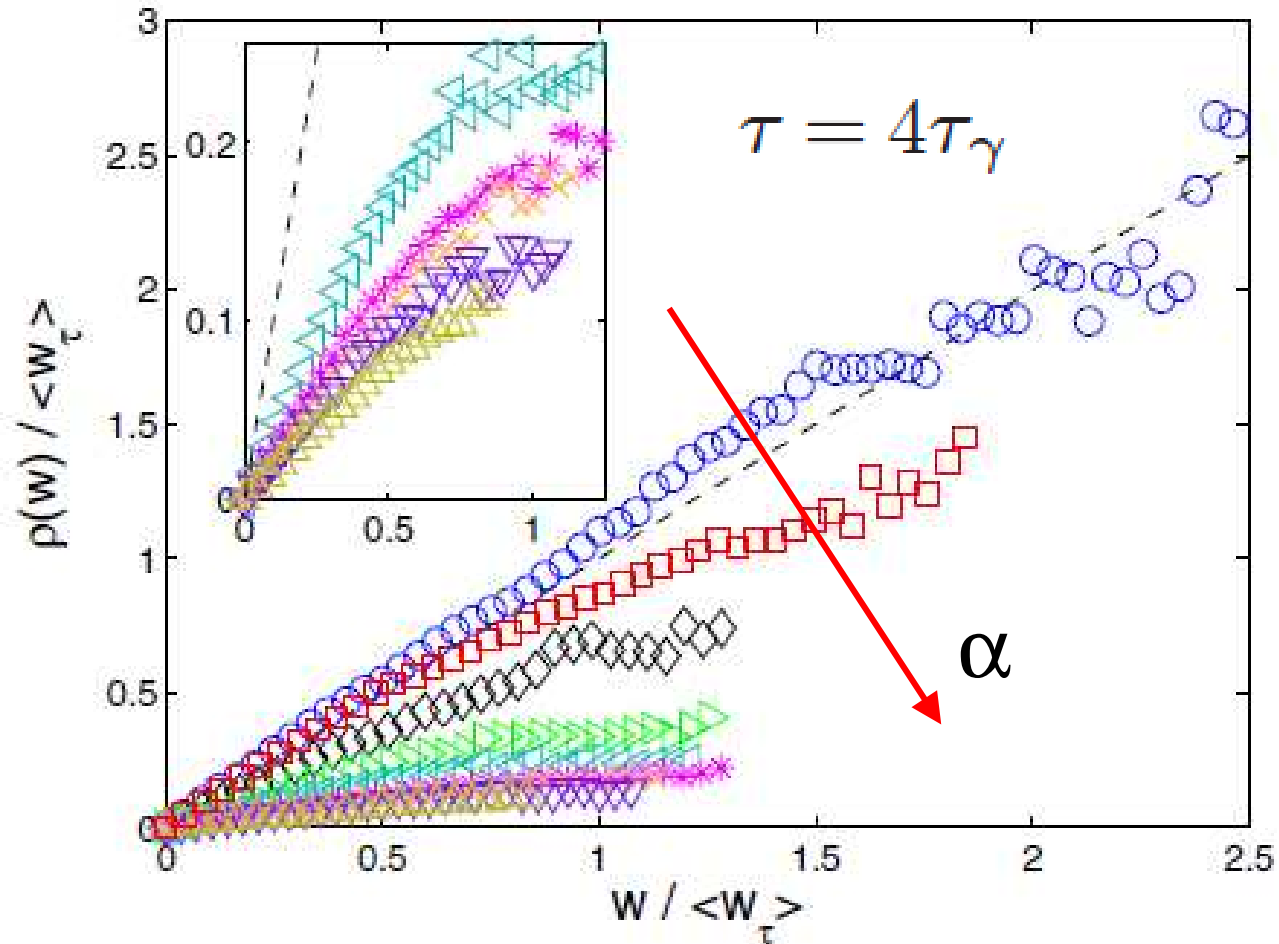
The relevant control parameter is

$$\alpha = \frac{\langle x^2 \rangle}{\langle x^2 \rangle_{eq}} - 1, \quad \text{where}$$

$$\langle x^2 \rangle_{eq} = k_B T / k$$



## FR for NESS driven by a random forcing



$$\rho(w) = \ln \frac{P(w_\tau = w)}{P(w_\tau = -w)}$$

## FR for NESS driven by a random forcing

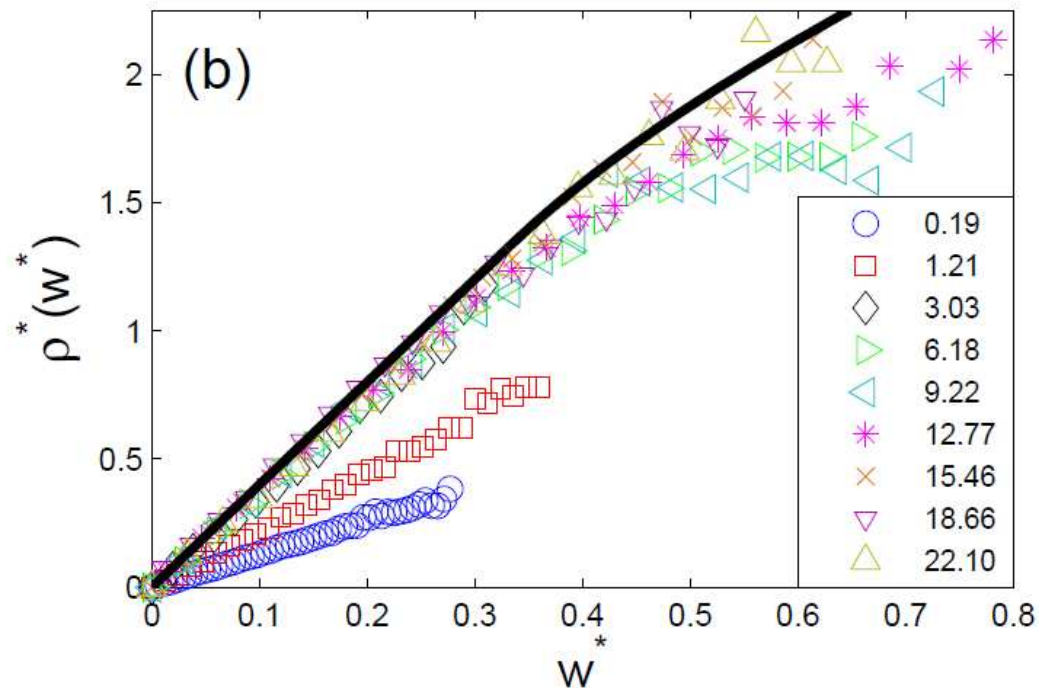
$$w_\tau^* = \frac{\tau_c}{\tau} \frac{w_\tau}{1 + \alpha}$$

$$k\langle x^2 \rangle / k_B = (1 + \alpha)T \approx \alpha T$$

$$\rho^*(w^*) = \lim_{\tau/\tau_c \rightarrow \infty} \frac{\tau_c}{\tau} \ln \frac{P(w_\tau^* = w^*)}{P(w_\tau^* = -w^*)}$$

$$\rho^*(w^*) = \begin{cases} 4w^* & w^* < 1/3 \\ \frac{7}{4}w^* + \frac{3}{2} - \frac{1}{4w^*} & w^* \geq 1/3 \end{cases}$$

Farago J., Physica A, 331 (2004) 69.





# FR for NESS driven by a random forcing

## Conclusions

- We have studied the FT for the work fluctuations in two experimental systems in contact with a thermal bath and driven out of equilibrium by a stochastic force.
- The main result of our study is that the validity of FT is controlled by the parameter  $\alpha$ . For  $\alpha < 1$  we have shown that the validity of the steady-state FT is a very robust result.
- In contrast for  $\alpha > 1$ , when the randomness of the system becomes dominated by the external stochastic forcing, we have shown that FT is violated.
- For  $\alpha \gg 1$  the data can be described by a master curve with a suitable effective temperature