

# **Fluctuations near a critical point**

# Fluctuations near a critical point

## Motivations

Why is interesting to study these fluctuations near a second order phase transition?

1) The probability density functions of global variables

S. T. Bramwell, P. Holdsworth, J.-F. Pinton, Nature vol. 396, 552 (1998)

*Universality of rare fluctuations in turbulence and critical phenomena*

E. Bertin, Phys. Rev. Lett, 95 170601 (2005).

*Global fluctuations in Gumbel Statistics*

2) Aging at critical point

L. Berthier, P. Holdsworth, Europhys. Lett. 58, 35 (2002)

*Surfing on a critical line: Rejuvenation without chaos, memory without a hierarchical phase space*

P. Calabrese and A. Gambassi, cond-mat/0410357V2

*Aging Properties of Critical Systems*

# Fluctuation Dissipation Ratio (FDR) during aging

**In equilibrium**  $\chi(t, t_w) = \frac{1}{k_B T} (C_\theta(t, t) - C_\theta(t, t_w))$

$$\chi(t, t_w) = \frac{\langle \Delta \rangle}{\Gamma_{ext}} = \int_{t_w}^t R(t, t') dt' \quad C_\theta(t, t_w) = \langle \delta\theta(t) \delta\theta(t_w) \rangle$$

**Out equilibrium** (Cugliandolo and Kurchan 1992) **FDR**

$$\chi(t, t_w) = \frac{X(t, t_w)}{k_B T} (C_\theta(t, t) - C_\theta(t, t_w)) \quad X(t, t_w) = \frac{T}{T_{eff}(t, t_w)}$$

**Experimentally this idea has been tested in**

- Spin glasses
- Colloids
- Polymers

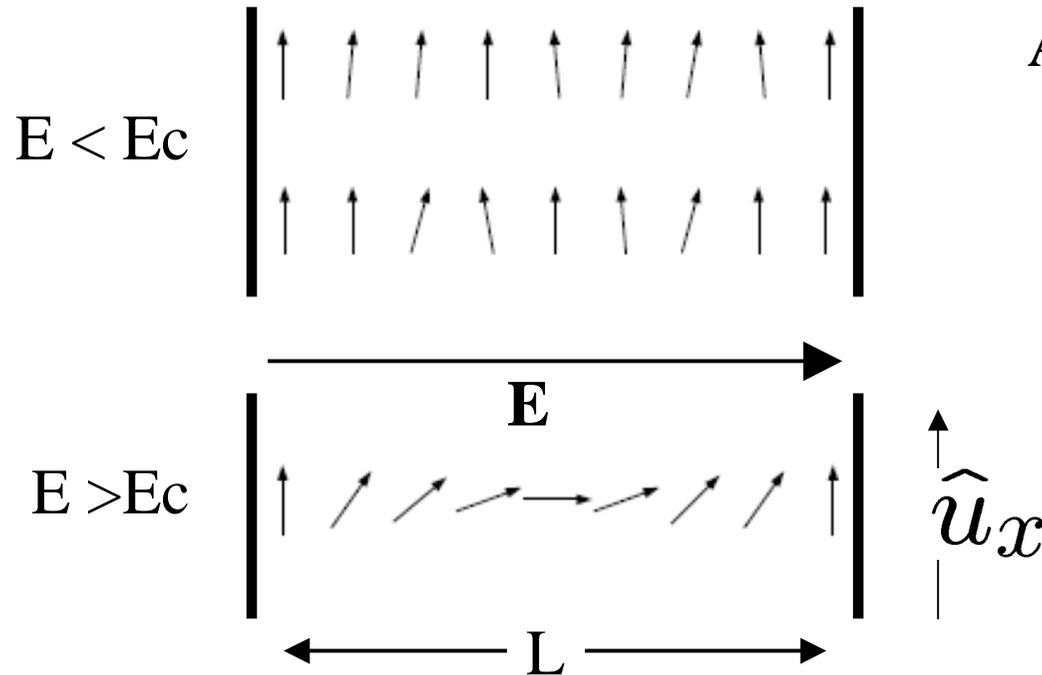
Controversial results

Need for experiments where the results  
can be directly compared with theoretical models

# Outline

- 1) The Fréedericksz transition in liquid crystals
- 2) Second order phase transition and the global variable of interest
- 3) Experimental system
- 4) Experimental results on PDF: the universal PDF for global variables
- 5) Aging at critical point
- 6) Experimental results on aging
- 7) Conclusions

## Liquid Crystals and Fréedericksz transition (I)



A liquid crystal consists of elongated molecules

$\hat{n}$  is the director

Surface treatment.  
Parallel anchored  
(planar allignement)

Competition between :

- Elastic energy  $\hat{n} // \hat{u}_x$
- Electrostatic energy  $\hat{n} // \vec{E}$

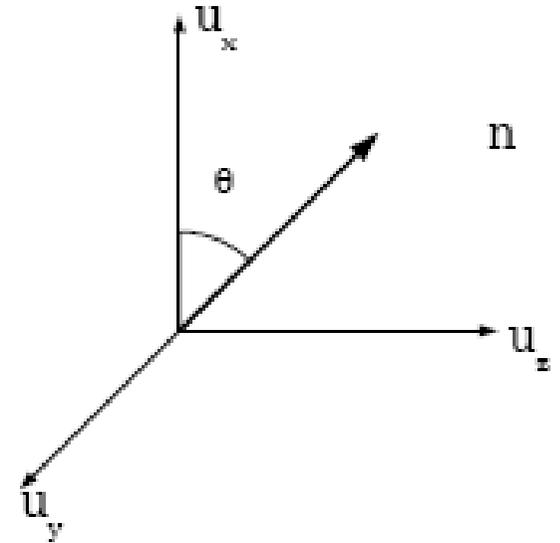
Control parameter :  
voltage difference  $U$

## Liquid Crystals and Fréedericksz transition (II)

$$\hat{n} = \cos(\theta) \hat{u}_x + \sin(\theta) \hat{u}_z$$

With boundary conditions

$$\theta(z=0) = \theta(z=L) = 0$$



Solution of the form:  $\theta(z) = \theta_0(x, y) \sin(\frac{\pi z}{L})$

If  $\theta_0 \ll 1$  remains small, the equation of motion of  $\theta_0$  is :

$$\tau_0 \frac{d\theta_0}{dt} = \epsilon \theta_0 - \left( \kappa + \frac{\epsilon + 1}{2} \right) \theta_0^3$$

$$\tau_0 = \frac{\gamma}{\epsilon_0 \epsilon_a E_c^2} \quad U_c = \pi \sqrt{\frac{K_1}{\epsilon_0 \epsilon_a}} \quad \kappa = \frac{K_3 - K_1}{K_1} \quad \epsilon = \frac{U^2}{U_c^2} - 1$$

Correlation length in the xy plane :  $\xi_r = \frac{L}{\pi \sqrt{\epsilon}}$

(San Miguel, Phys. Rev. A, 32, 3811, 1985)

## Fréedericksz transition

- The Fréederick transition is a second order phase transition
- The order parameter is  $\theta_0(x, y)$
- The control parameter is  $\epsilon = U^2/U_c^2 - 1$
- The relaxation time is  $\tau_{relax} = \tau_o/\epsilon$
- The correlation length  $\xi_r = \frac{L}{\pi\sqrt{\epsilon}}$

## Shadowgraph image

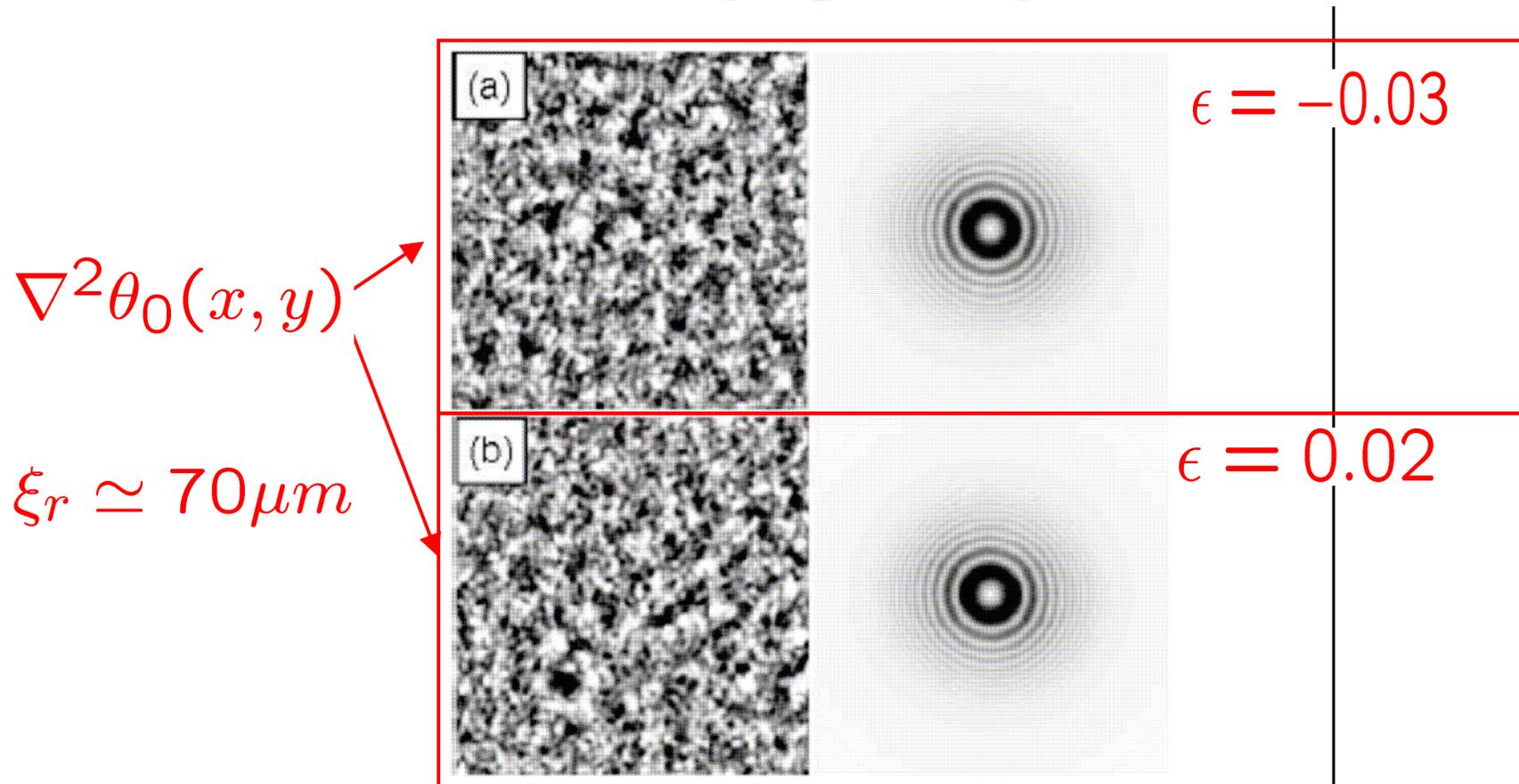


FIG. 1: Left column: shadowgraph snapshots of an area of  $1.03 \times 1.03 \text{ mm}^2$ . Right column: corresponding structure factors, averaged over 256 images. (a):  $V_0 = 3.170$ , (b):  $V_0 = 3.256$  Volt.

$$U_c = 3.22V \text{ and } L = 27 \mu m$$

from Zhou, Ahlers, arXiv: nlin/0409015v2

## Fréedericksz transition

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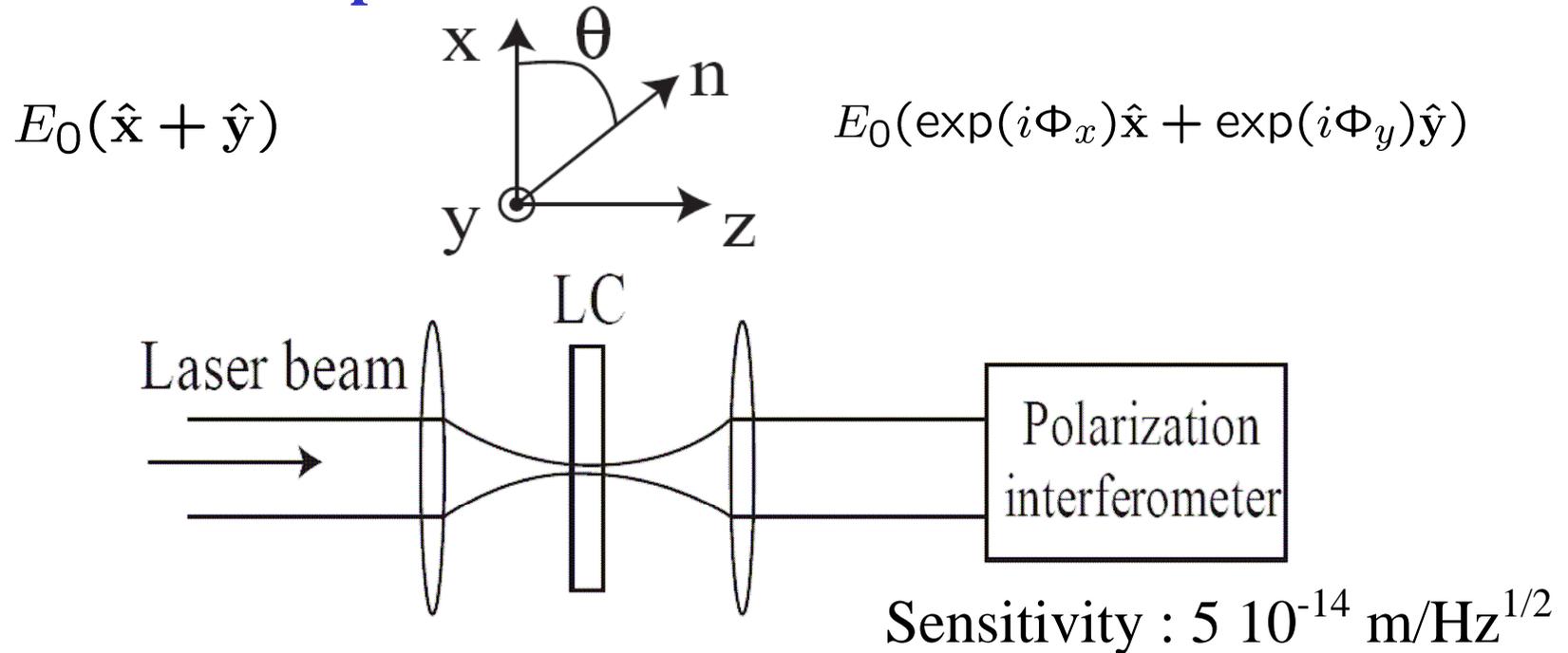
Global variable:

$$\zeta = \frac{2}{L} \int_0^L \langle (1 - n_x^2) \rangle_{xy} dz \simeq \iint_A \theta_0^2 \frac{dxdy}{A}$$

where  $A = \pi D^2/4$  and  $\langle . \rangle_{xy}$  stands for mean on  $A$ .

We measure de fluctuations of  $\zeta$   
as a function of  $D/\xi$  and  $\epsilon$

## Experimental set-up



- Two cells with different thickness:  $L = 6\mu\text{m}$  and  $L = 25\mu\text{m}$ . Surface  $S = 1\text{cm}^2$
- The liquid crystal cell is a birifrengent plate. Optical axis  $// \hat{n}$
- Measurement of the dephasing  $\Phi = \Phi_x - \Phi_y$  between the two polarisations.

$$\Phi = a + b \zeta = a + b \iint_A \theta_0^2 \frac{dxdy}{A}$$

- Laser diameter inside the cell  $38\mu\text{m}$  (limited by diffraction)

## Measure of the dephasing

The dephasing between the  $E_x$  and  $E_y$  is :

$$\Phi = \left\langle \frac{2\pi}{\lambda} \int_0^L \left( \frac{n_o n_e}{\sqrt{n_o^2 \cos(\theta)^2 + n_e^2 \sin(\theta)^2}} - n_o \right) dz \right\rangle_{xy}$$

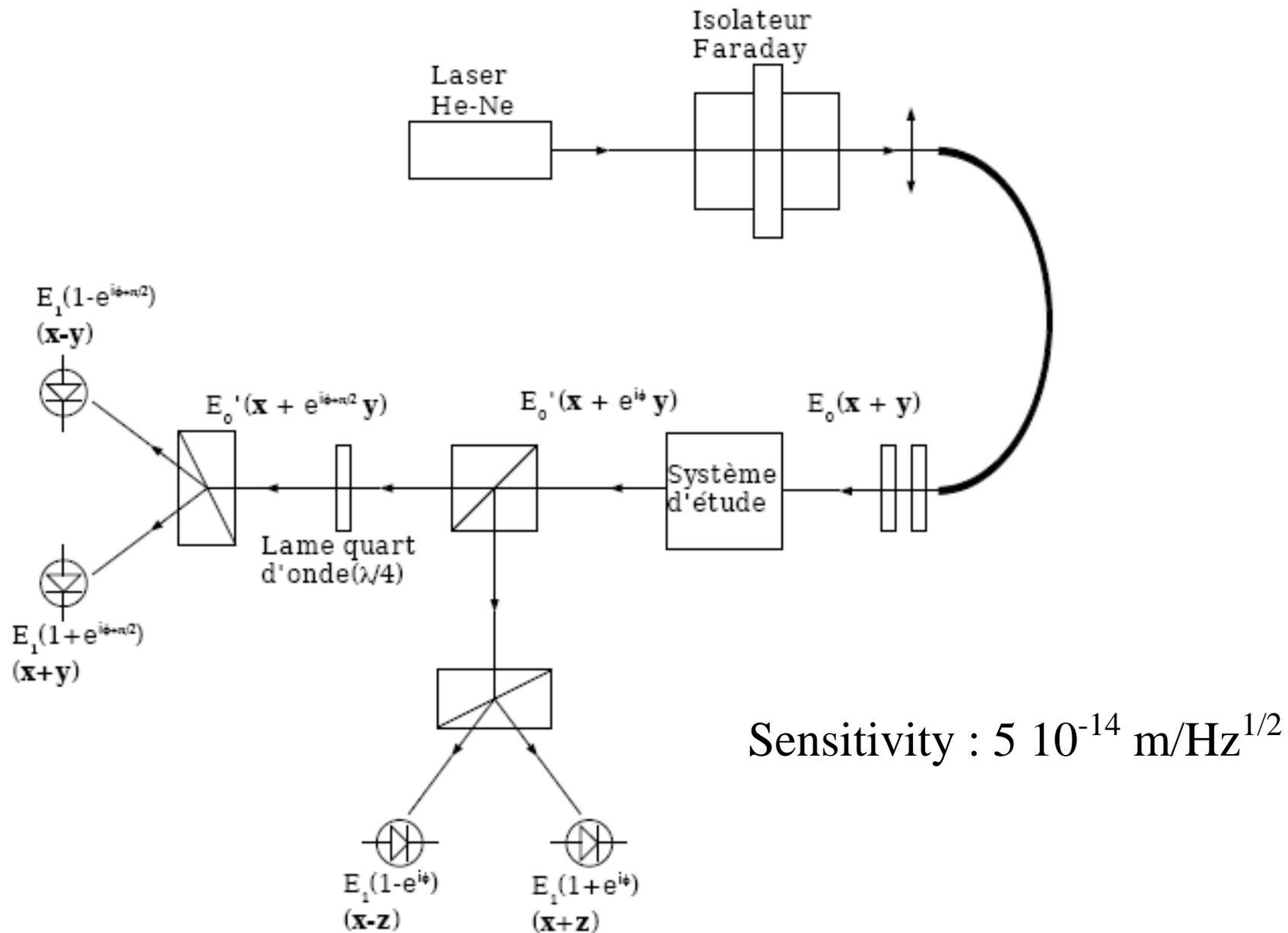
with  $(n_o, n_e)$  the two anistotropic refractive indices.

If  $\theta \ll 1$  in terms of  $\zeta$  we get:

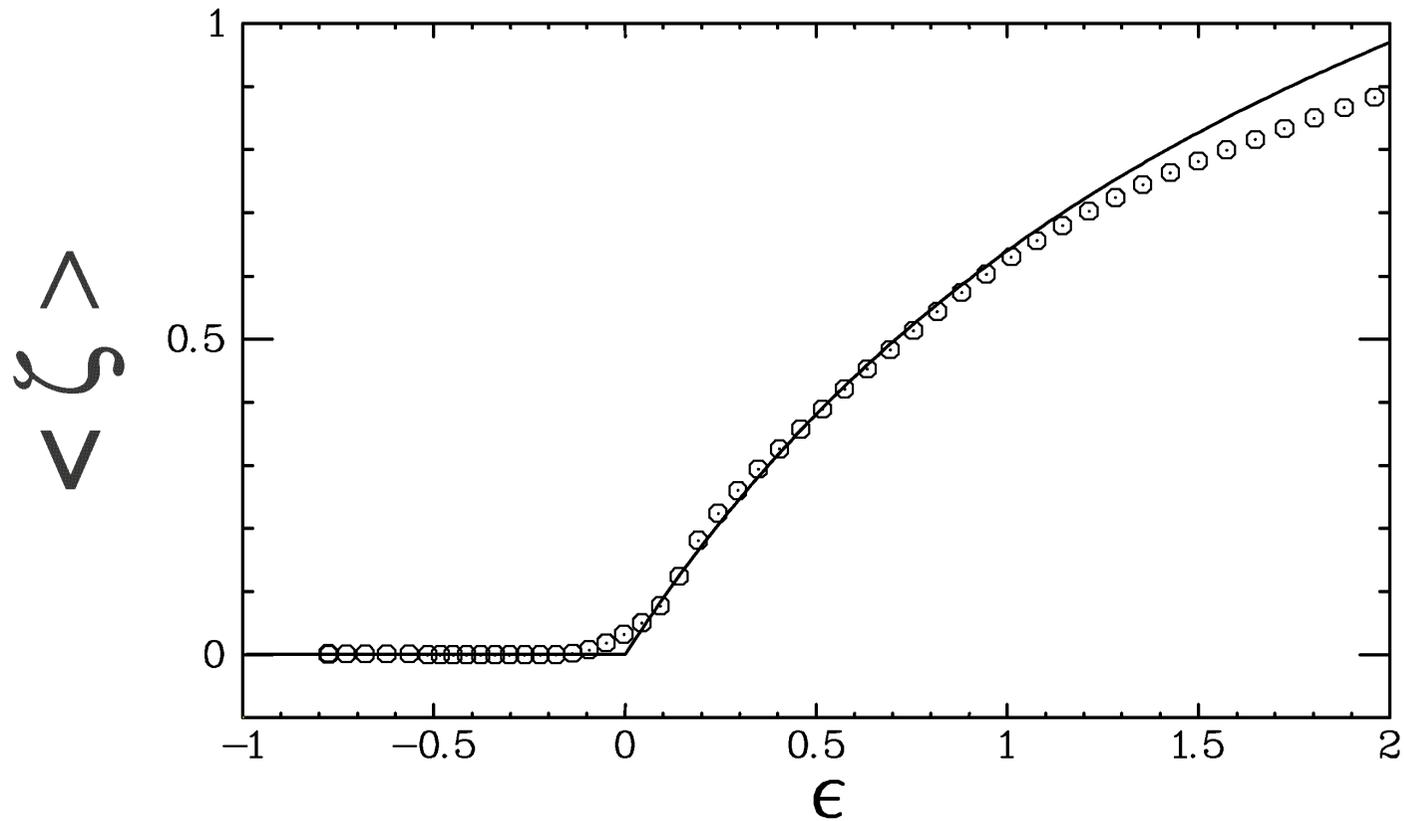
$$\Phi = \Phi_0 \left( 1 - \frac{n_e(n_e + n_o)}{4n_o^2} \zeta \right) \quad \Phi_0 \equiv \frac{2\pi}{\lambda} (n_e - n_o) L$$

$$\text{Interferometer noise : } \frac{(\Phi - \Phi_0)}{\Phi_0} \simeq 6 \cdot 10^{-8} \text{ Hz}^{-1/2}$$

## Experimental system: polarization interferometer



## Mean Amplitude of $\langle \zeta \rangle$



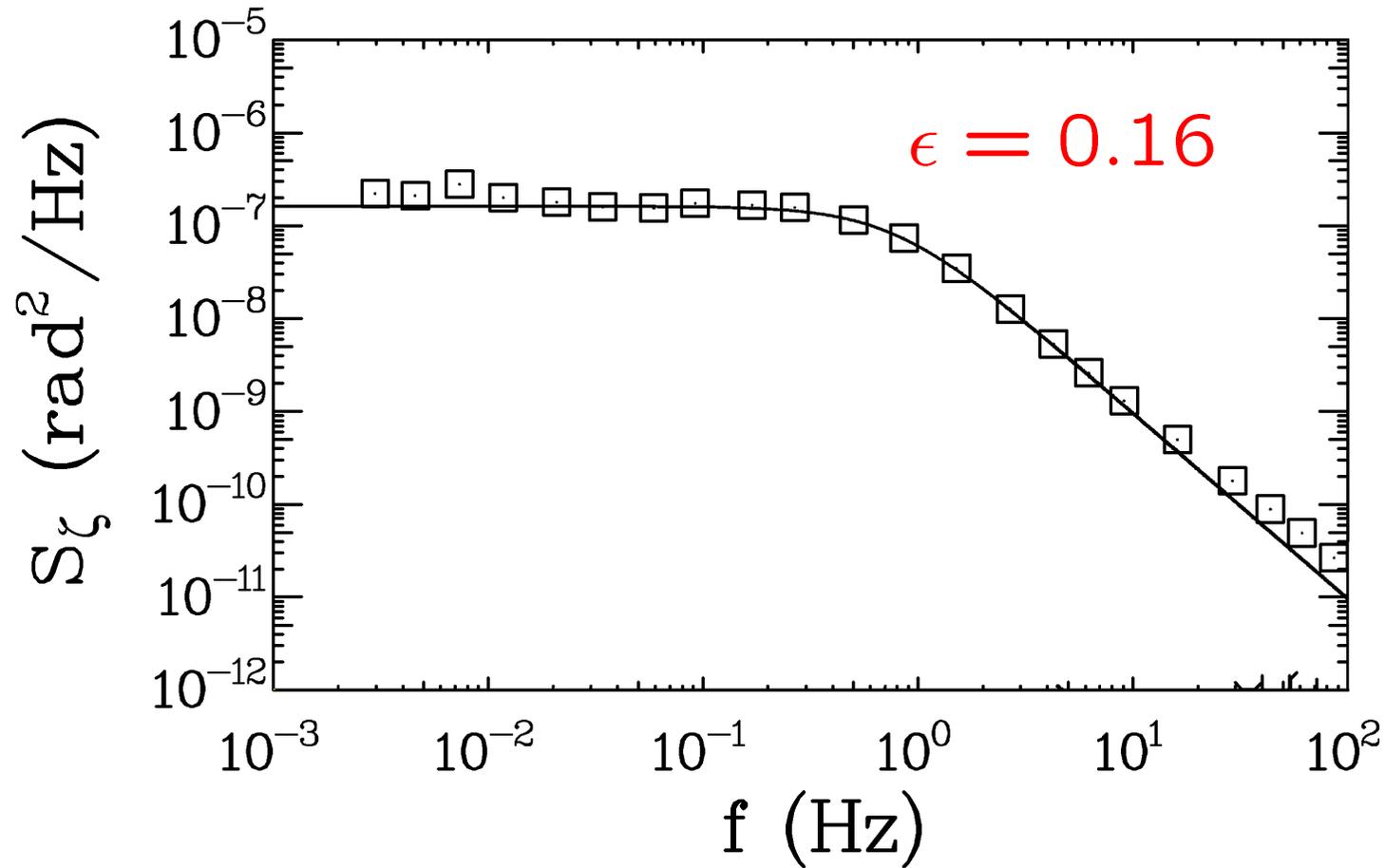
For Liquid Crystal 5CB

Computed  $V_c = 0.74V$

Measured  $V_c = 0.73V$  by  $\langle \zeta \rangle \propto \epsilon = 0$

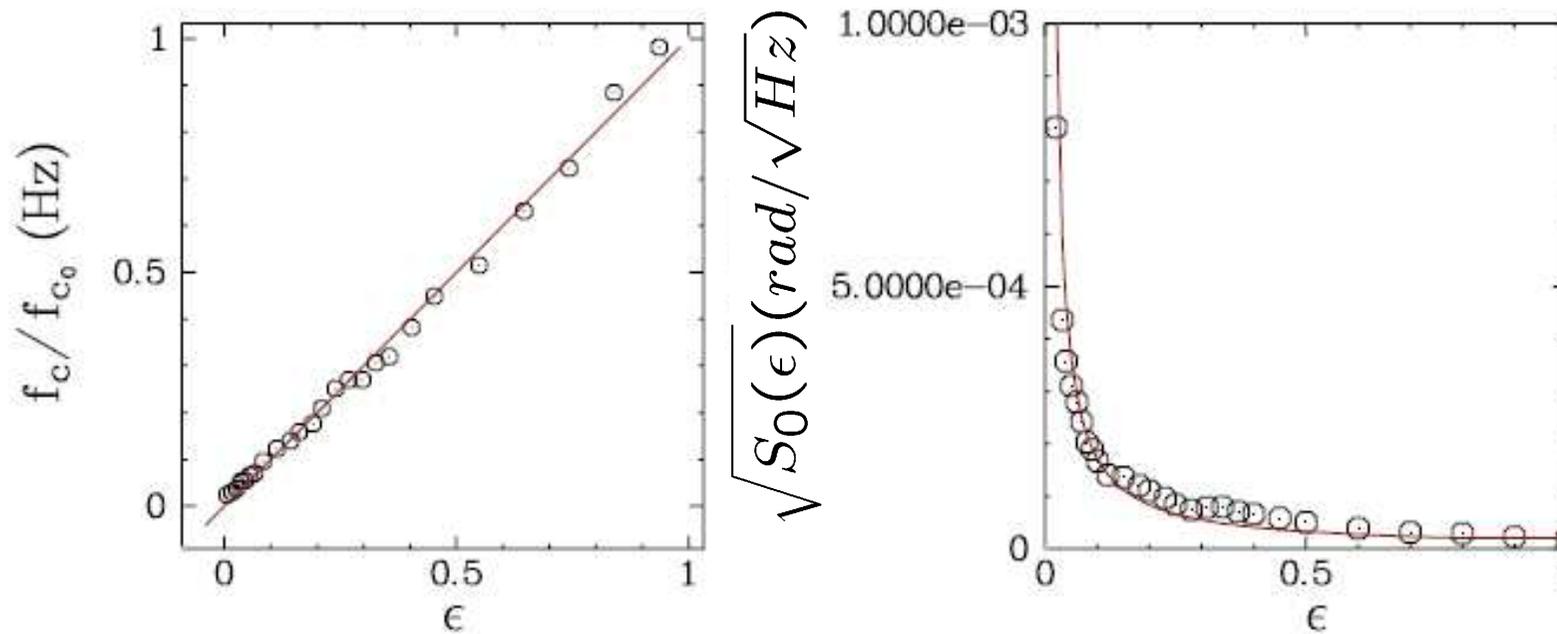
$$\langle \zeta \rangle = \frac{2\epsilon}{2k + \epsilon + 1}$$

## Fluctuation spectra of $\zeta$



$$S_\zeta(f) = \frac{S_o(\epsilon)}{1 + \left(\frac{f}{f_c(\epsilon)}\right)^2}$$

## Relaxation time as a function of $\epsilon$



$$f_c = \frac{1}{\tau_{relax}} = \epsilon \frac{1}{\tau_0}$$

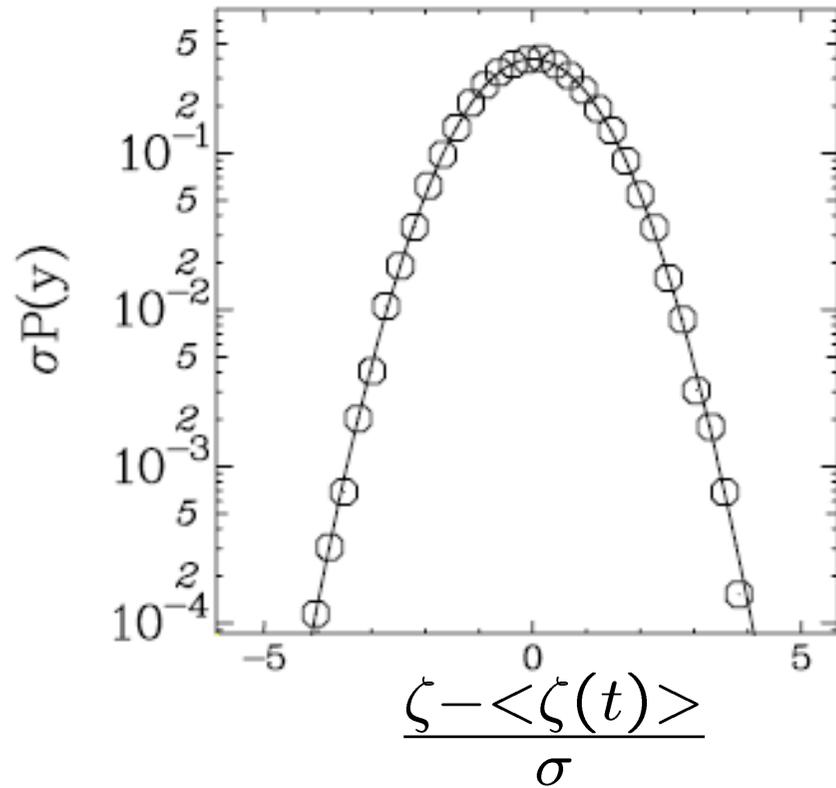
from the fit  $U_c = 0.73V$  and  $\tau_0 = 0.1s$

Computed  $\tau_0 = 0.09s$ .

Variance :  $\sigma^2 \propto S_0(\epsilon) f_c(\epsilon) \propto \epsilon^{-1}$

# PDF of $\zeta$

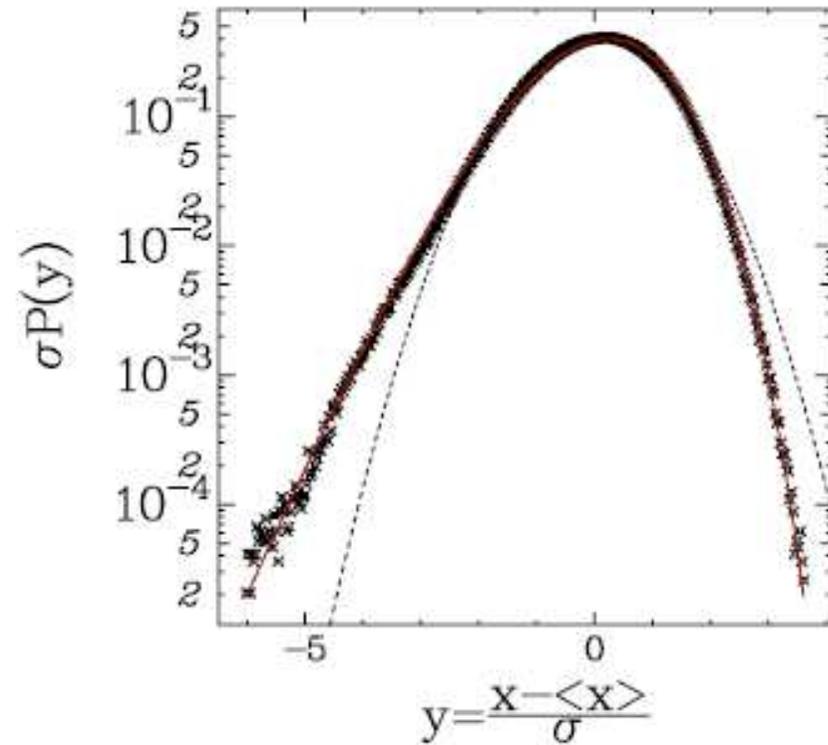
Cell width  $L=6.7 \mu m$ ,  $\tau_o \simeq 0.1s$  and  $D = 38\mu m$



$$\epsilon = 0.02$$

Gaussian fluctuations

$$\xi_r = \frac{L}{\pi\sqrt{\epsilon}} = 15\mu m$$

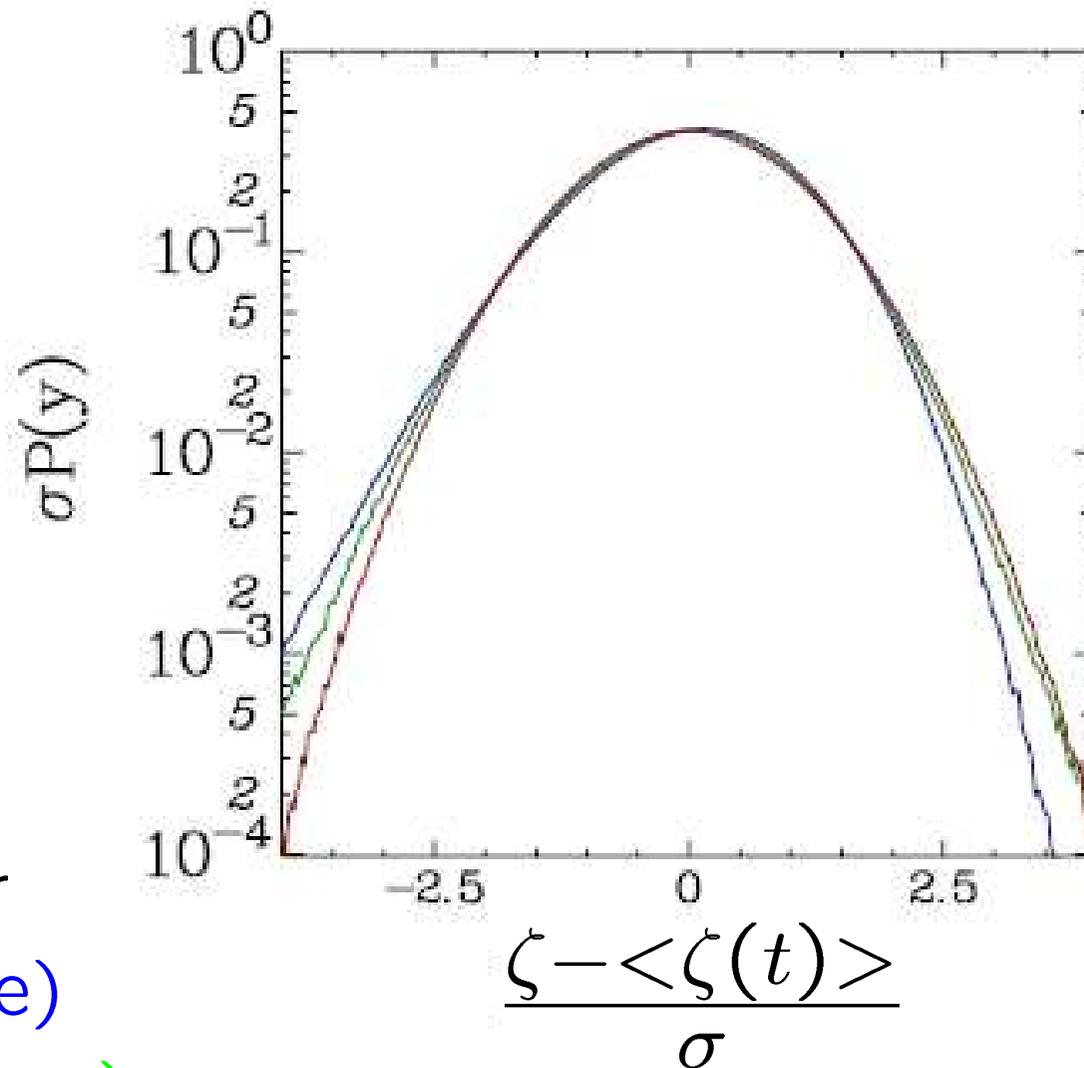


$$\epsilon \simeq 0.001$$

Non-Gaussian fluctuations

$$\xi_r = \frac{L}{\pi\sqrt{\epsilon}} = 60\mu m$$

# Low frequency influence



High-pass filter

- 1 mHz (Blue)
- 50 mHz (Green)
- 2 Hz (Red)

# Questions

- What is the interpretation for the PDF of  $\theta_0$  ?
- Why this behavior has not been observed before?

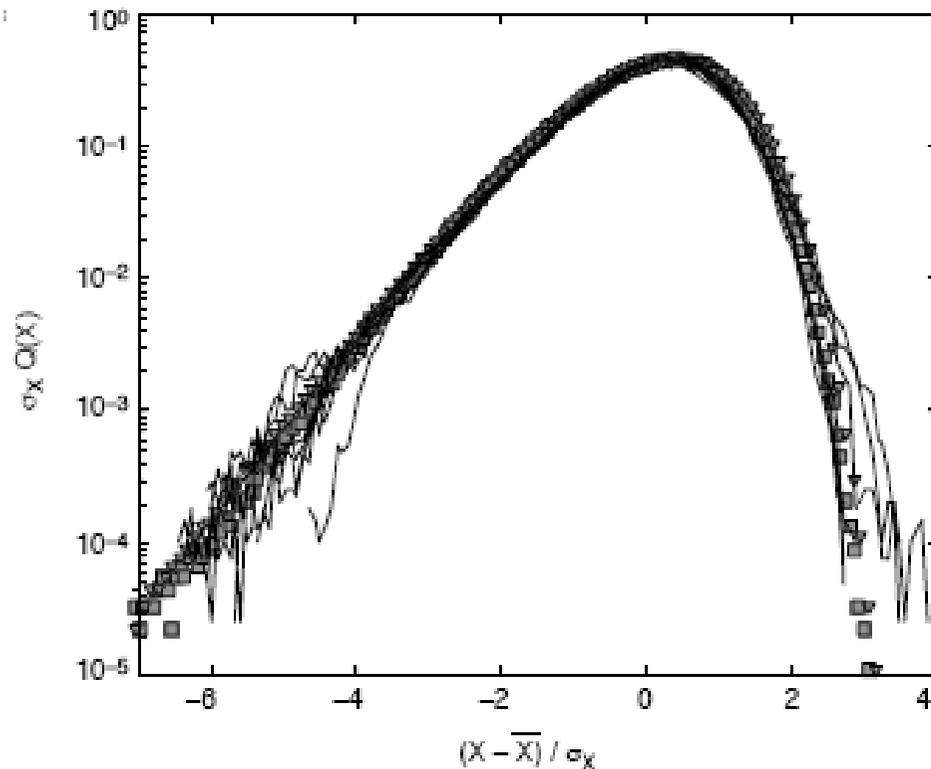
# Universality of fluctuation PDF

- In 1998 Bramwell, Holdsworth, Pinton proposed that in spatial extended systems the PDF of a global quantity  $x$  may take an universal form:

$$P(x) = K \exp\{-a [b(x - s) - \exp(b(x - s))]\}$$

BHP distribution is equivalent to Gumbel for  $a$  integer. ( Bertin 2005).

- Correlation lengths are of the order of the system size

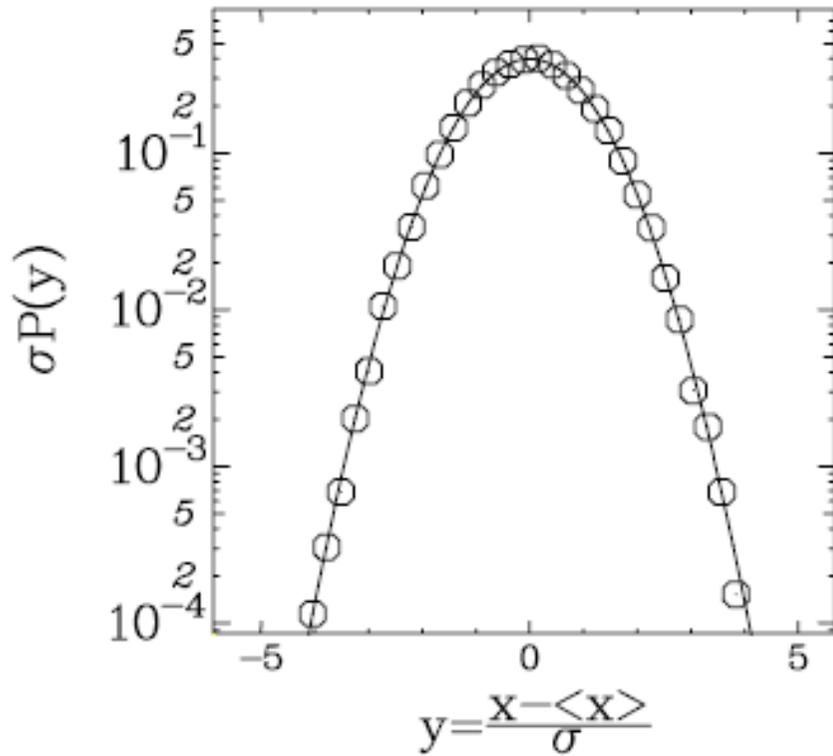


BHP has been observed in the fluctuations of :

- the injected power in an experiment of turbulence
- the magnetization in a xy model
- on the height of the Danube river

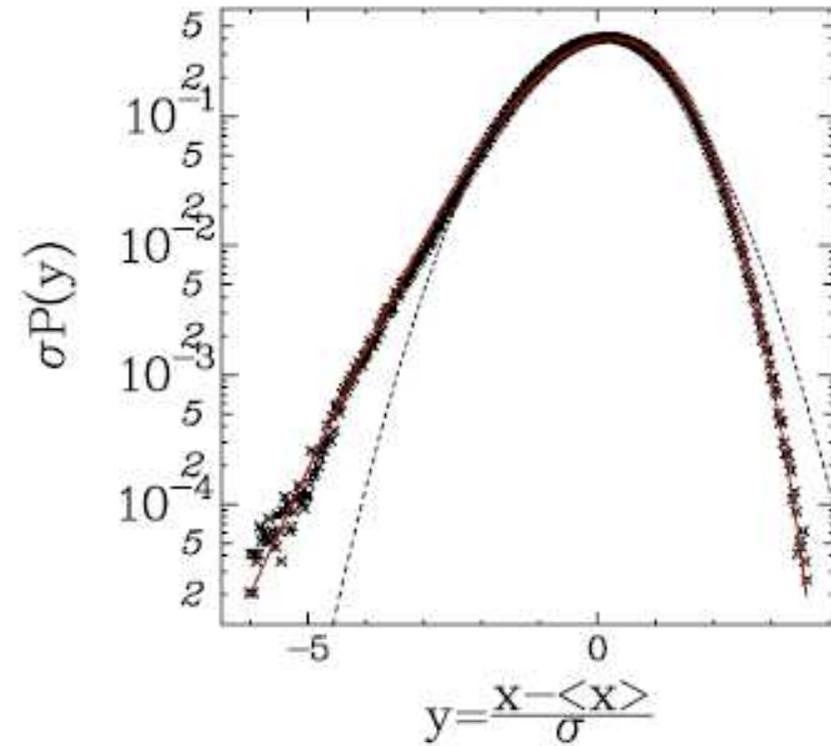
# PDF of $\zeta$

Cell with  $L=6.7 \mu\text{m}$  and  $\tau_0 \simeq 0.1\text{s}$



$$\epsilon = 0.02$$

Gaussian fluctuations



$$\epsilon \simeq 0.001$$

Non-Gaussian fluctuations

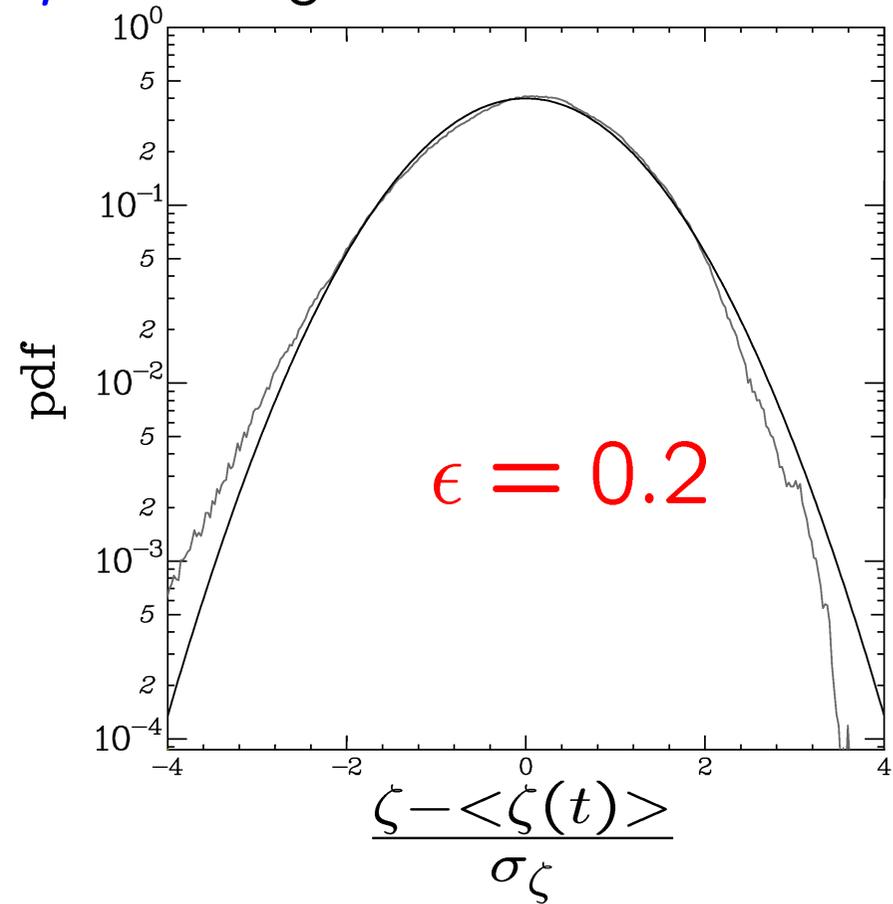
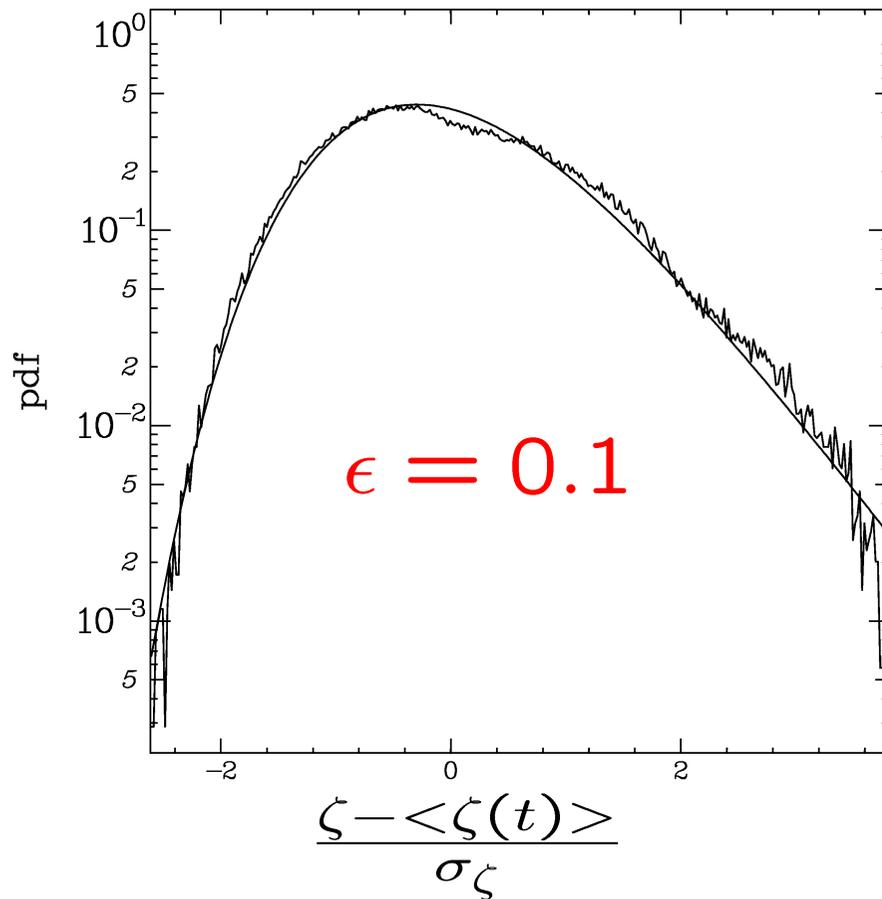
Gumbel model ( $a \simeq 6$ )

$$\xi_r = \frac{L}{\pi\sqrt{\epsilon}} = 60 \mu\text{m}$$

# PDF of $\zeta$

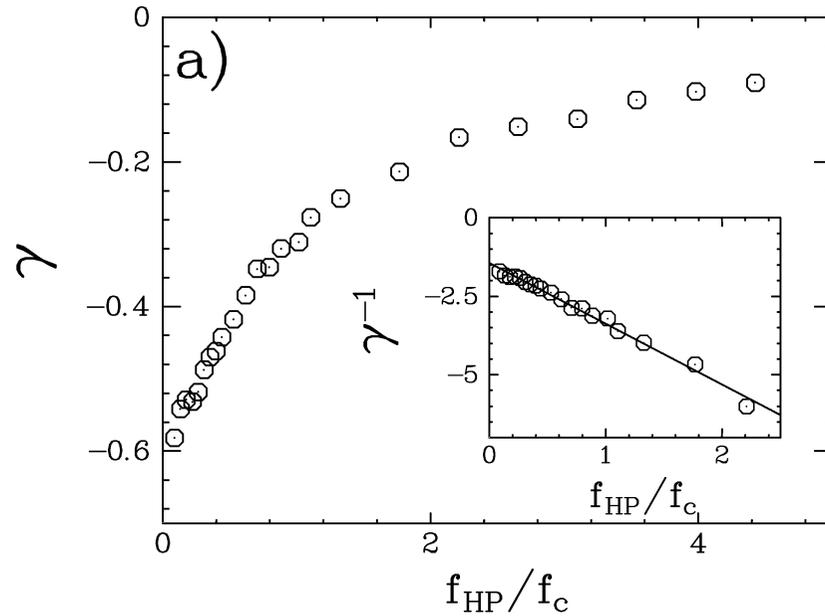
Cell with  $L = 25\mu\text{m}$

$\tau_0 \simeq 2.5\text{s}$

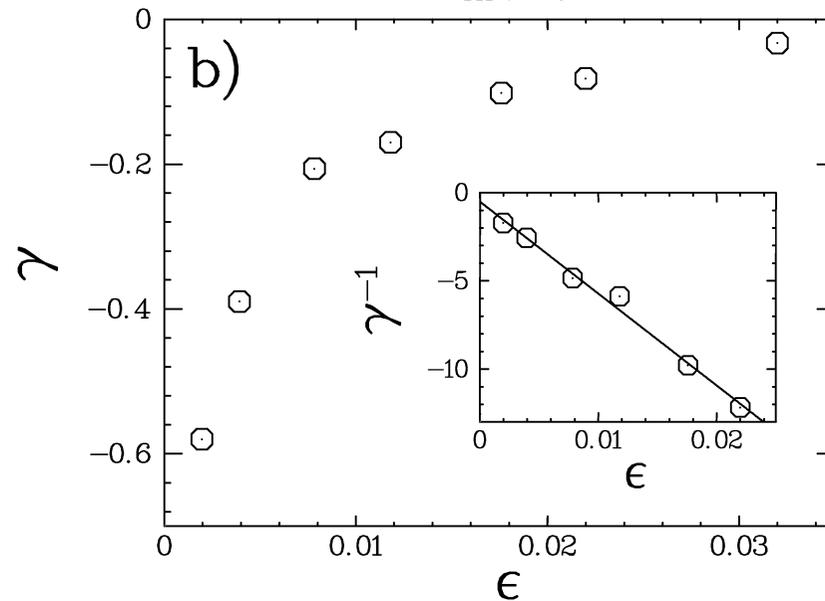


- The BHP distributions are observed when the diameter of the probe is smaller than the correlation length
- The BHP is determined by the very slow motion

# Dependence of $\gamma$ as a function of $\epsilon$ and $f_c$



$$\gamma = \langle y^3 \rangle \approx \frac{1}{\sqrt{a}}$$



# Aging at critical point

(Calabrese, Gambassi)

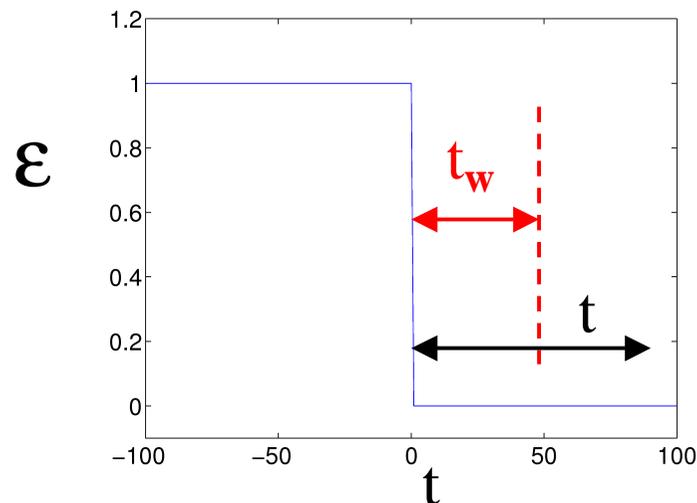
At  $t=0$  the system is rapidly quenched from  $\epsilon = \epsilon_1$  to  $\epsilon_0 = 0$ .

Within the Ginzburg-Landau approximation the dynamics of  $\theta_0$  is ruled by:

$$\tau_0 \frac{d\theta_0}{dt} = -\theta_0^3$$

whose solution is

$$\langle \theta_0(t)^2 \rangle = \frac{\tau_0}{2(t + \tau_m)} \quad \text{with} \quad \tau_m = \frac{\tau_0}{2\epsilon_1}$$



The system remains out of equilibrium forever.

Experimentally the condition  $\epsilon = 0$  is never realized

$\chi(t, t_w)$  and  $C(t, t_w)$

# Fluctuation dissipation theorem after a quench

(mean field)

We consider the fluctuations  $\delta\theta(t) = \theta_0(t) - \langle \theta_0(t) \rangle$

The correlation  $C(t, t_w) = \langle \delta\theta(t)\delta\theta(t_w) \rangle$  for  $t_w < t$

$$C(t, t_w) = \frac{(t_w + \tau_m)^4 - \tau_m^4}{2(t_w + \tau_m)^{3/2}(t + \tau_m)^{3/2}}$$

$$R(t, t_w) = \frac{\Delta\theta_0}{\Delta h} = \frac{(t_w + \tau_m)^{3/2}}{(t + \tau_m)^{3/2}} \quad \text{Response function}$$

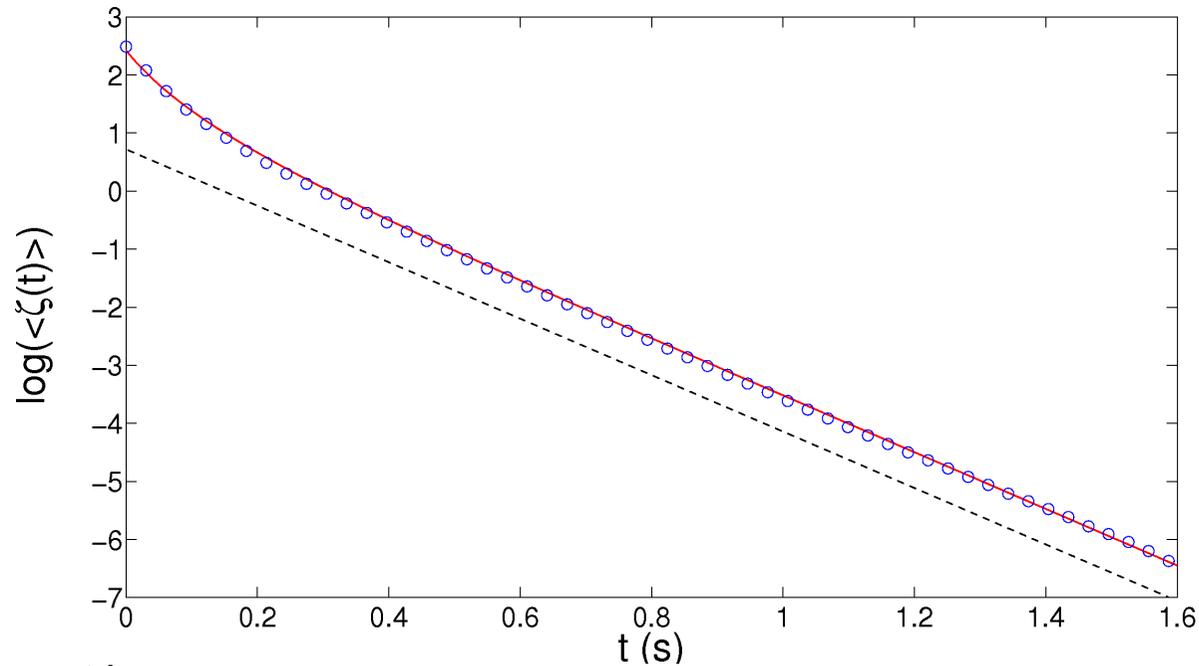
The fluctuation dissipation ratio (FDR)  $X(t, t_w) = -k_B T \frac{R(t, t_w)}{\partial_{t_w} C(t, t_w)}$

In equilibrium  $X(t, t_w) = 1$

After the quench at critical point  $X(t, t_w) < 0.8 \forall t$  and  $t_w$

# Time evolution after a quench in LC

from  $\epsilon_1 = 0.3$  to  $\epsilon_0 = 0.01$



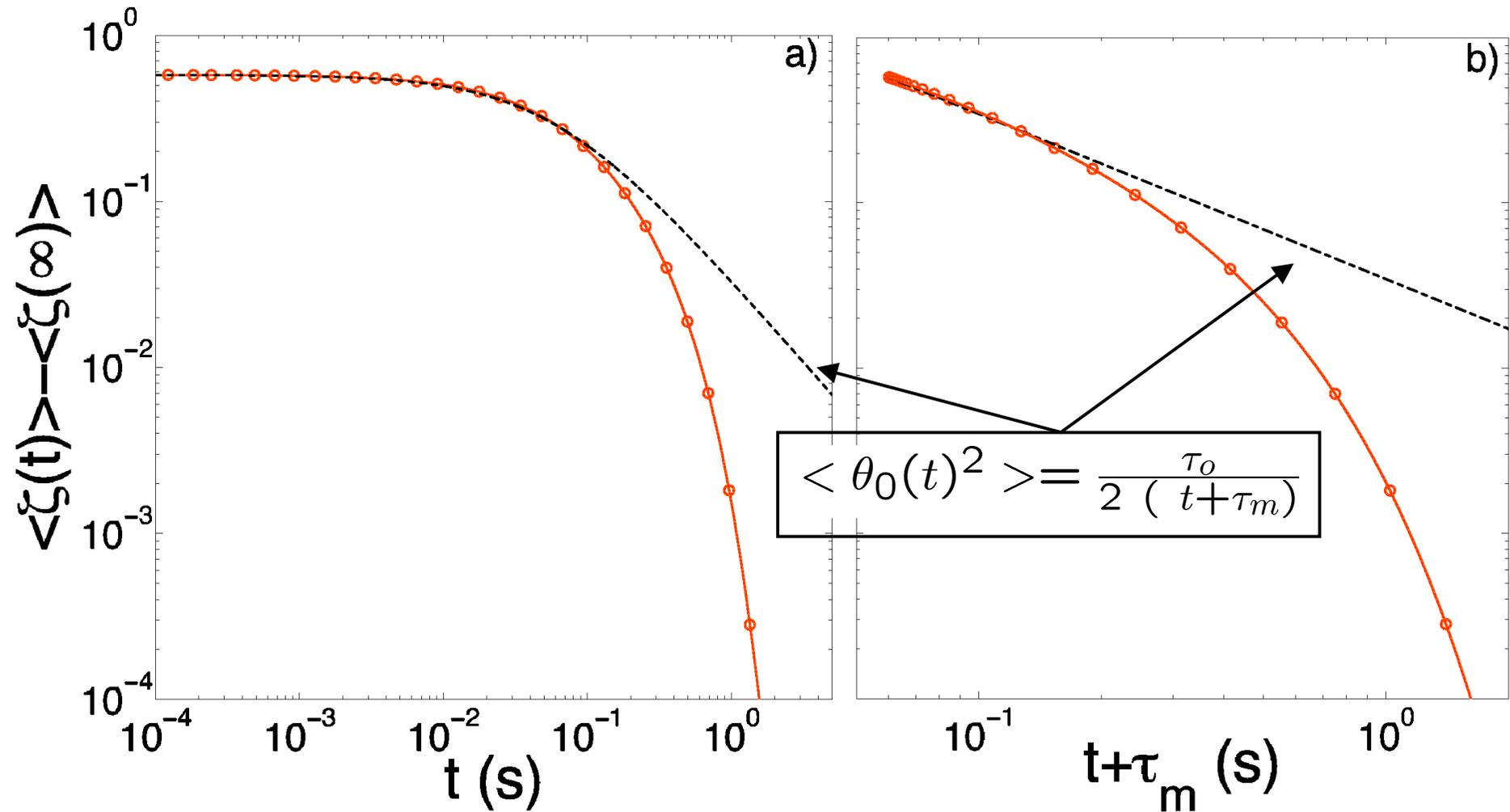
Fit function

$$\langle \zeta(t) \rangle = \frac{\langle \zeta(\infty) \rangle}{1 + \left( \frac{\langle \zeta(\infty) \rangle}{\langle \zeta(0) \rangle} - 1 \right) \exp\left(-\frac{2\epsilon_0 t}{\tau_0}\right)},$$

$$\text{with } \langle \zeta(\infty) \rangle = \frac{2\epsilon_0}{(2k + \epsilon_0 + 1)} \text{ and } \langle \zeta(0) \rangle = \frac{2\epsilon_1}{(2k + \epsilon_1 + 1)}$$

# Time evolution of $\zeta$ after a quench

Quench from  $\epsilon_1 \simeq 0.3$  to  $\epsilon_0 \simeq 0.01$   $\tau = \frac{\tau_0}{2\epsilon} \simeq 0.22s$



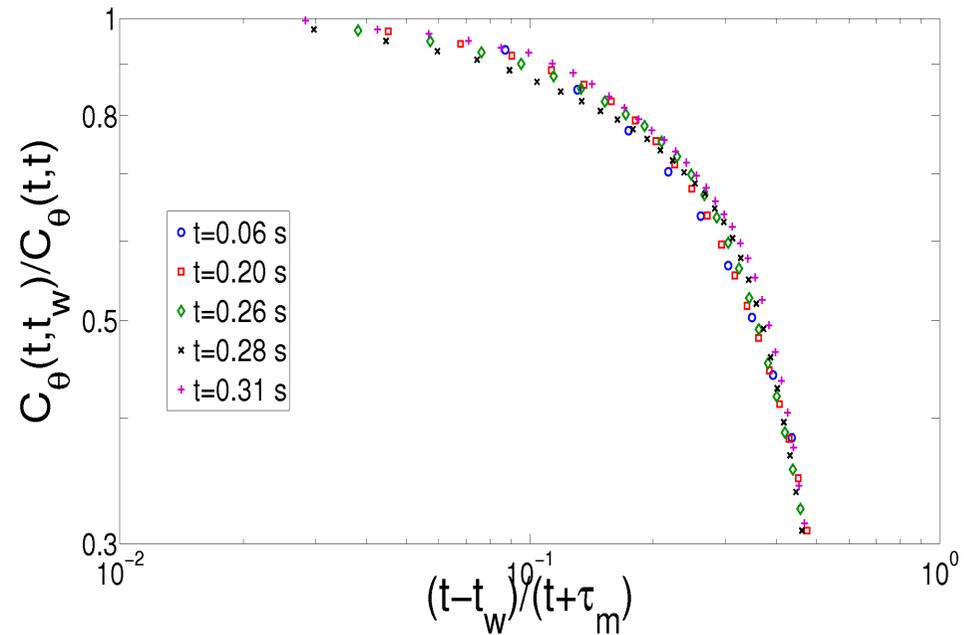
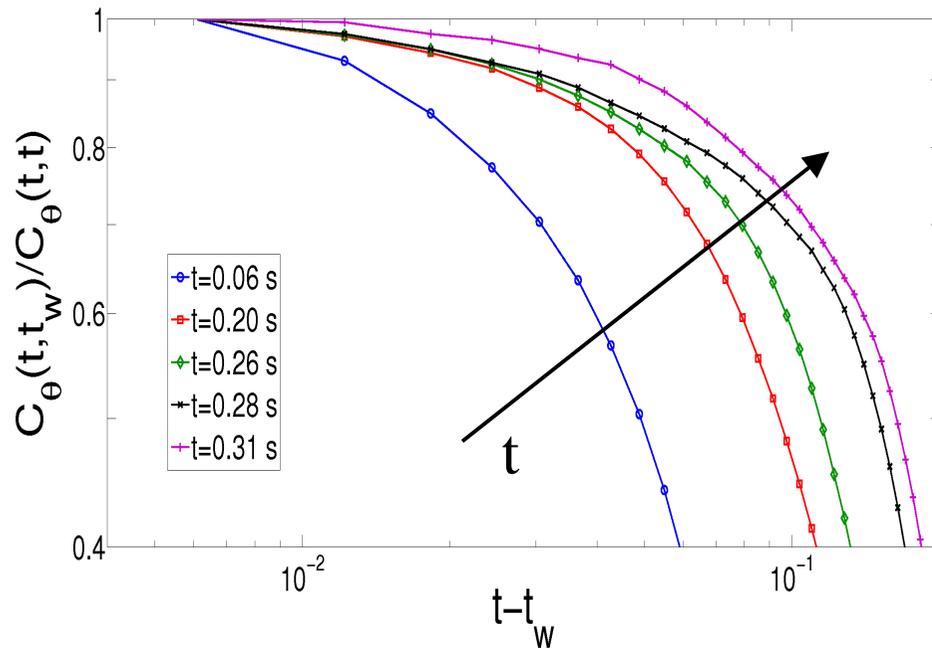
# Correlations

$$\zeta(t) = \theta_0(t)^2 = \psi_0^2(t) + 2\psi_0\delta\theta = \langle \zeta(t) \rangle + \delta\zeta(t)$$

$$\psi_0^2(t) = \langle \zeta(t) \rangle$$

$$C_\zeta(t, t_w) = \langle \delta\zeta(t)\delta\zeta(t_w) \rangle = 4\psi_0(t)\psi_0(t_w) C_\theta(t, t_w)$$

Master curve by rescaling  
 $(t - t_w) \rightarrow (t - t_w)/(t + \tau_m)$



## FDT in the LC experiment: the measure of the response function

$$\chi(t, t_w) = \frac{\langle \Delta \rangle}{\Gamma_{ext}} = \int_{t_w}^t R(t, t') dt' \quad C_\theta(t, t_w) = \langle \delta\theta(t) \delta\theta(t_w) \rangle$$

**FDR**

$$\chi(t, t_w) = \frac{1}{k_B T_{eff}(t, t_w)} (C_\theta(t, t) - C_\theta(t, t_w))$$

In equilibrium

$$T = T_{eff}$$

Which is the appropriate external torque  $\Gamma_{ext}$  for the LC ?

$$\frac{\gamma AL}{2} \frac{d\theta_0}{dt} = B [2\epsilon\theta_0 - (\kappa + \epsilon + 1) \theta_0^3] + \eta \quad \left\{ \begin{array}{l} B = A\pi^2 K_1 / 4L \\ \epsilon = \epsilon_0 + \delta\epsilon \\ \theta_0(t) = \psi_0 + \Delta(t) \end{array} \right.$$

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

$\Gamma_{ext}$

## FDT in the LC experiment:

$$\delta\theta = \frac{\delta\zeta(t)}{2\psi_0(t)} \quad \Gamma_{ext} = 4B \psi_0(t) \delta\epsilon \left(1 - \frac{\psi_0(t)^2}{2}\right)$$

Experimental test of these results (JSTAT P01033, 2009) :

- 1) Out of equilibrium, using the Transient Fluctuation Theorem
- 2) FDT in equilibrium

**In equilibrium**  $\psi_0(t) = \psi_0(t_w) = \psi_0$

$$\chi(\tau) = \frac{\langle \Delta(\tau) \rangle}{\Gamma_{ext}} = \frac{\chi_{\zeta, \delta\epsilon}}{4B \psi_0^2 \left(1 - \frac{\psi_0^2}{2}\right)}$$

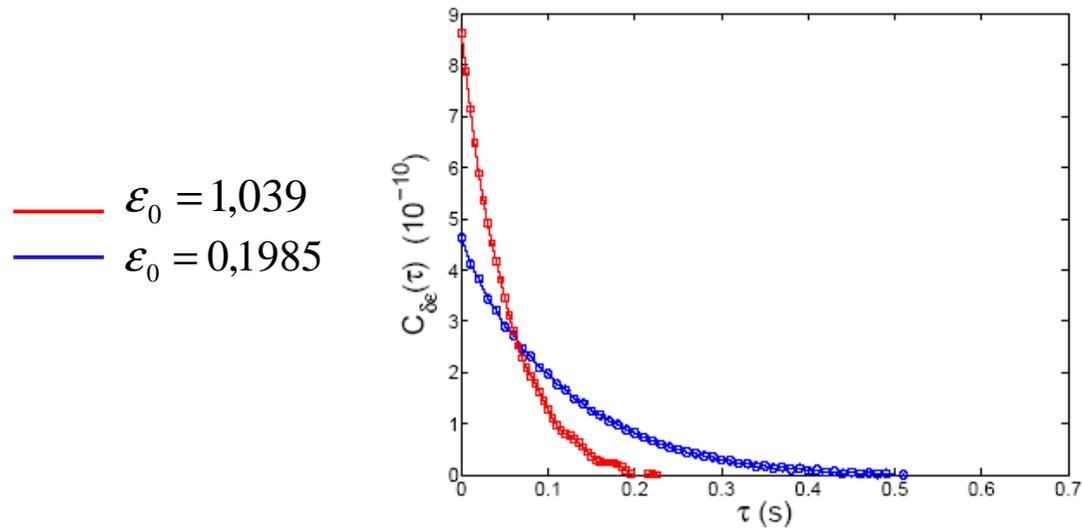
$$C_\zeta(t, t_w) = 4\psi_0^2 C_\theta(t, t_w)$$

and FDT

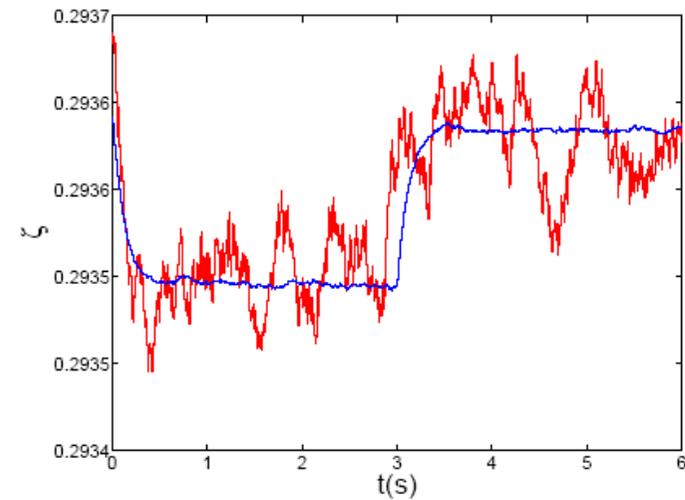
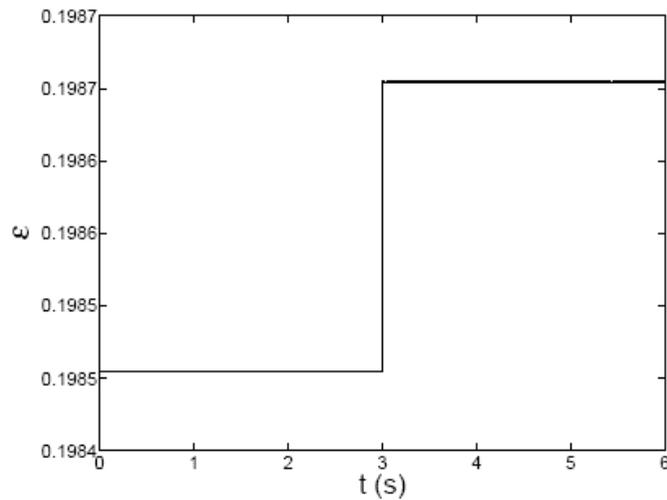
$$\frac{\chi_{\zeta, \delta\epsilon}}{B \left(1 - \frac{\langle \zeta \rangle}{2}\right)} = \frac{1}{k_B T} (C_\zeta(t, t) - C_\zeta(t, t_w))$$

with  $B = \mathcal{A}\pi^2 K_1 / 4L$

# FDT in the LC experiment in equilibrium

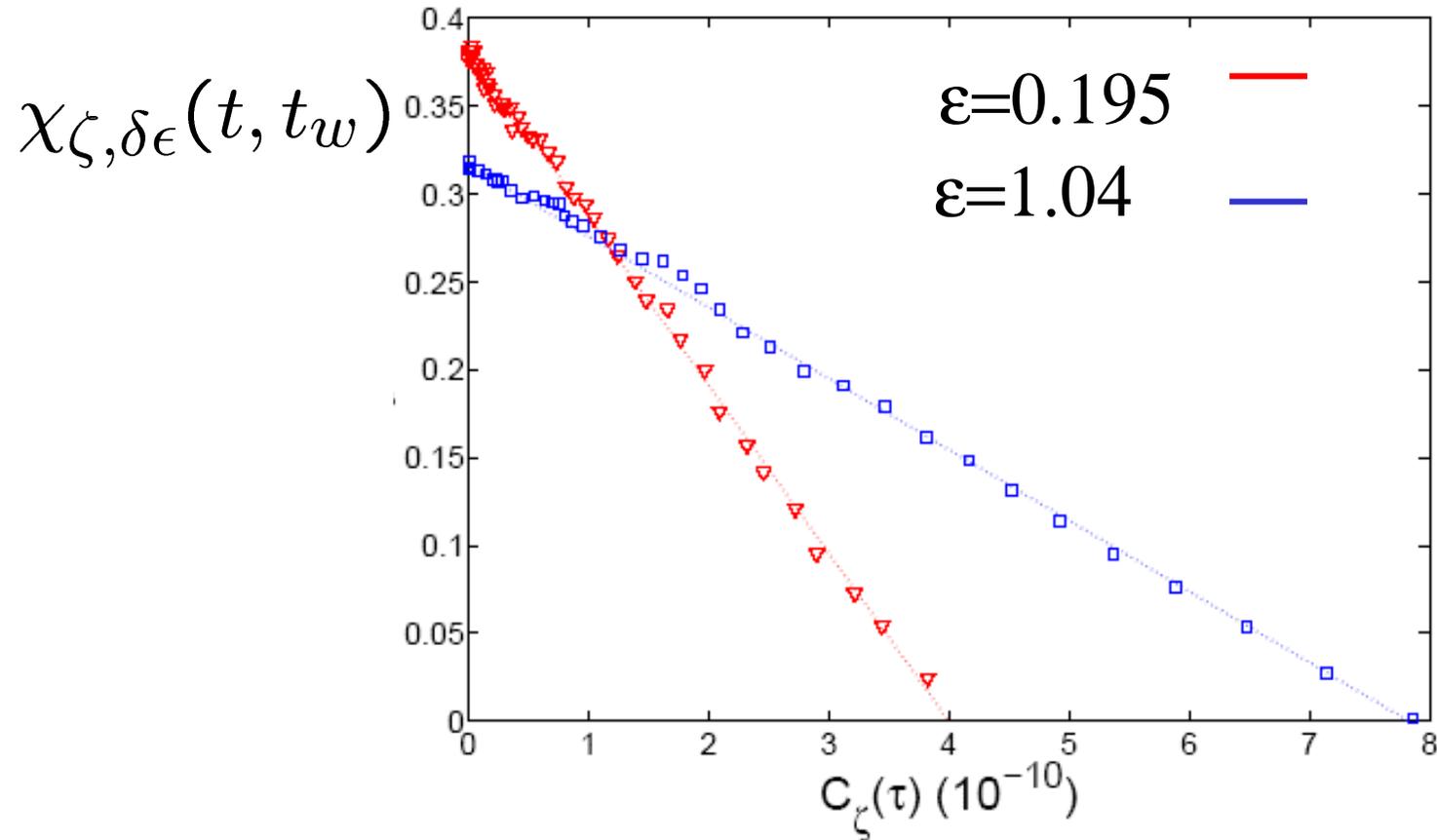


- response function



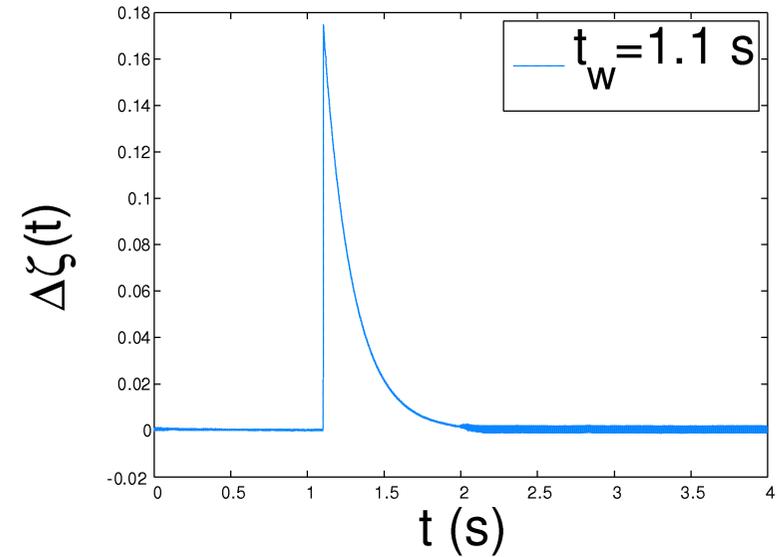
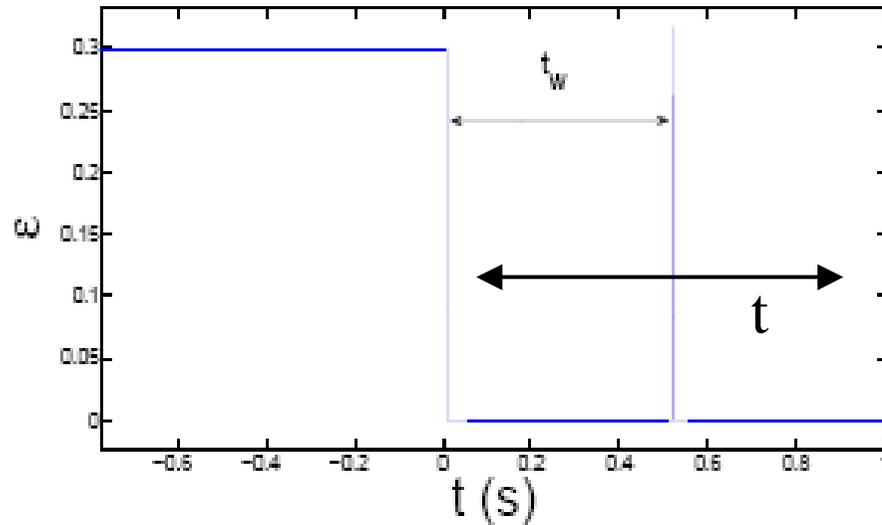
## FDT in the LC experiment in equilibrium

$$\frac{\chi_{\zeta, \delta\epsilon}}{B \left(1 - \frac{\langle \zeta \rangle}{2}\right)} = \frac{1}{k_B T} (C_\zeta(t, t) - C_\zeta(t, t_w)) \quad \text{with } B = \mathcal{A} \pi^2 K_1 / 4L$$



at  $\epsilon = 0.195$ ,  $\frac{B \left(1 - \frac{\langle \zeta \rangle}{2}\right)}{k_B T} = 8.4 \cdot 10^8$ ,  $\mathcal{A} = 2.4 \text{ mm}^2$  and  $D_0 \simeq 1.8 \text{ mm}$

# Response function during aging



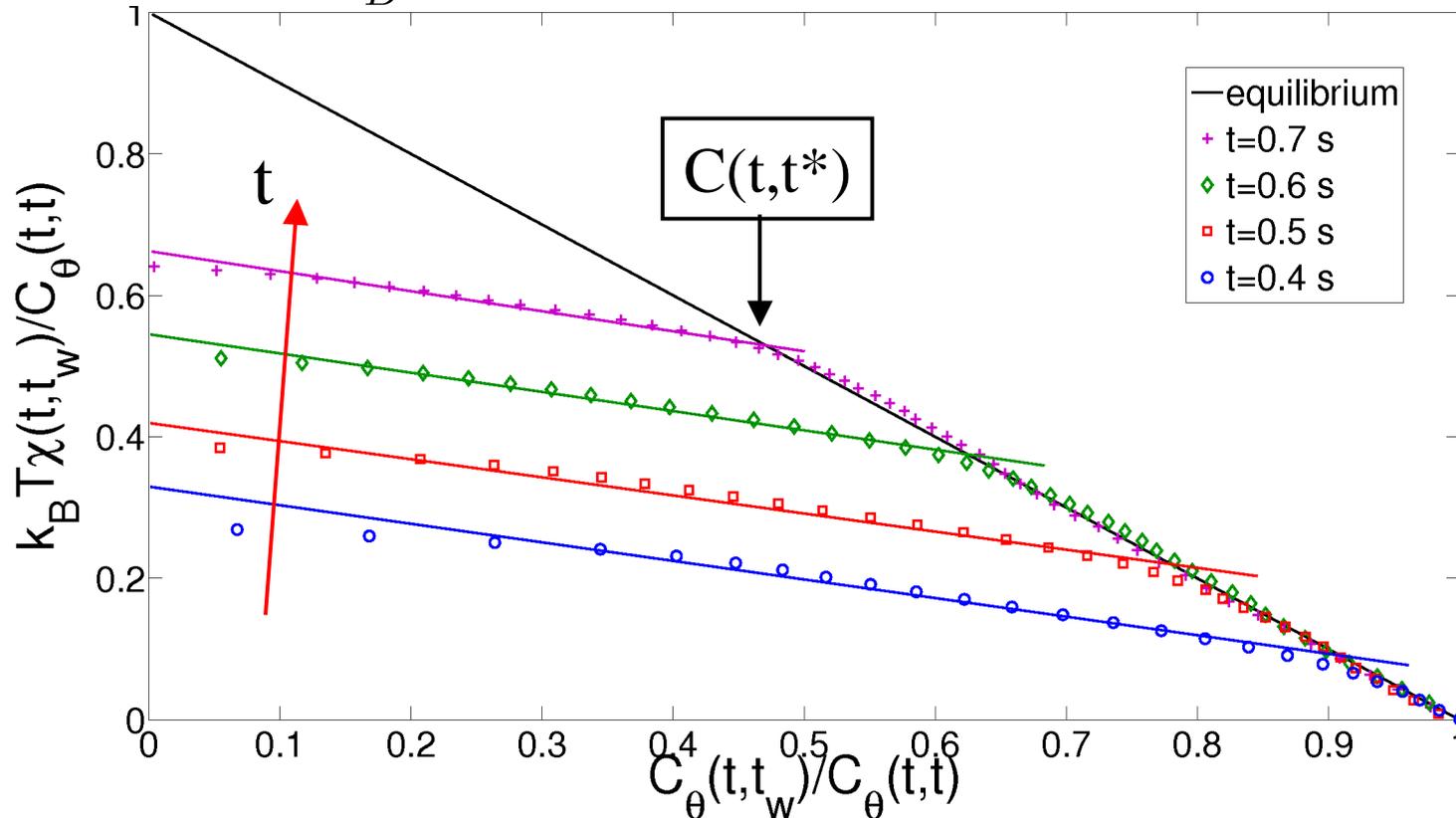
$$\Delta = \theta_0(t) - \psi_0(t) \quad \text{and} \quad \Delta = \frac{\zeta(t) - \psi_0(t)^2}{2\psi_0(t)} = \frac{\zeta(t) - \langle \zeta(t) \rangle}{2\psi_0(t)} = \frac{\Delta\zeta}{2\psi_0(t)}$$

$$R(t, t_w) = \frac{\langle \Delta(t) \rangle}{\Gamma_{ext}(t_w)} = \frac{R_{\zeta, \delta\epsilon}(t, t_w)}{4B \psi_0(t_w) \psi_0(t) \left(1 - \frac{\psi_0(t_w)^2}{2}\right)}$$

$$\chi(t, t_w) = \frac{\langle \Delta(t) \rangle}{\Gamma_{ext}} = \int_{t_w}^t R(t, t') dt'$$

# FDT out of equilibrium: fixed $t$ as a function of $t_w$

$$\chi(t, t_w) = \frac{X(t, t_w)}{k_B T} (C_\theta(t, t) - C_\theta(t, t_w)) \quad \text{with } T_{eff} = \frac{T}{X(t, t_w)}$$



for  $C(t, t_w) > C(t, t^*)$ ,  $X = 1$  and  $T_{eff} = T$

for  $C(t, t_w) < C(t, t^*)$ ,  $X \simeq 0.33$  and  $T_{eff} \simeq 3T$

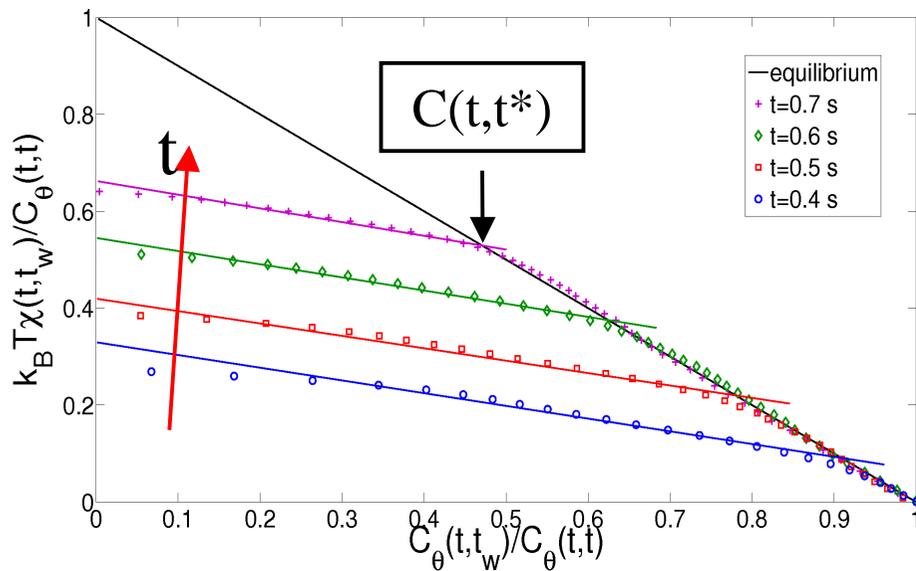
L. Cugliandolo, J. Kurchan, and L. Peliti, Phys. Rev. E 55, 3898 (1997).

D. Hérisson and M. Ocio, Phys. Rev. Lett. 88, 257202, (2002)

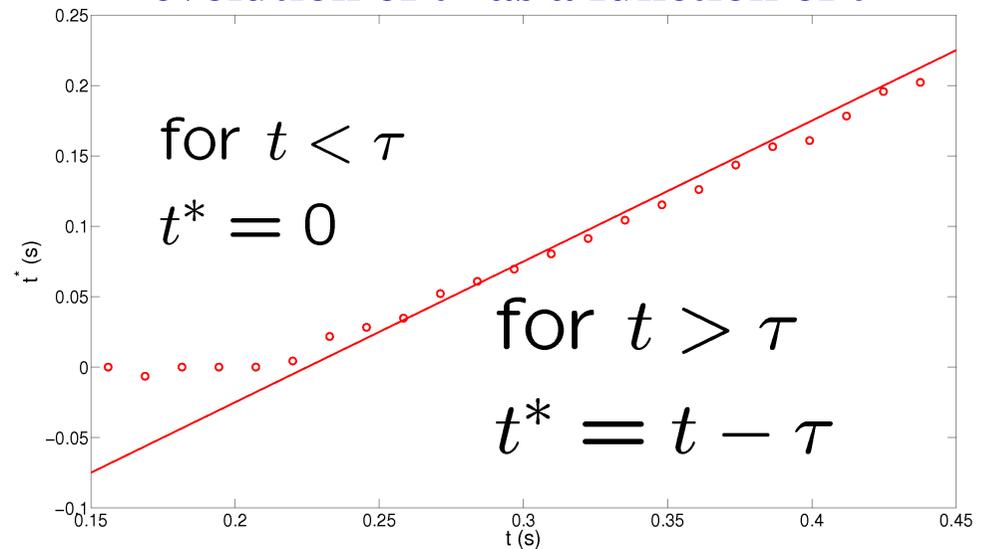
# FDT out of equilibrium: fixed $t$ as a function of $t_w$

for  $C(t, t_w) > C(t, t^*)$ ,  $X = 1$  and  $T_{eff} = T$

for  $C(t, t_w) < C(t, t^*)$ ,  $X \simeq 0.33$  and  $T_{eff} \simeq 3T$



## evolution of $t^*$ as a function of $t$



$\frac{t^*}{t} = 1 - \frac{\tau}{t}$  for  $t > \tau$  defines the length of the equilibrium interval with respect to the total time.

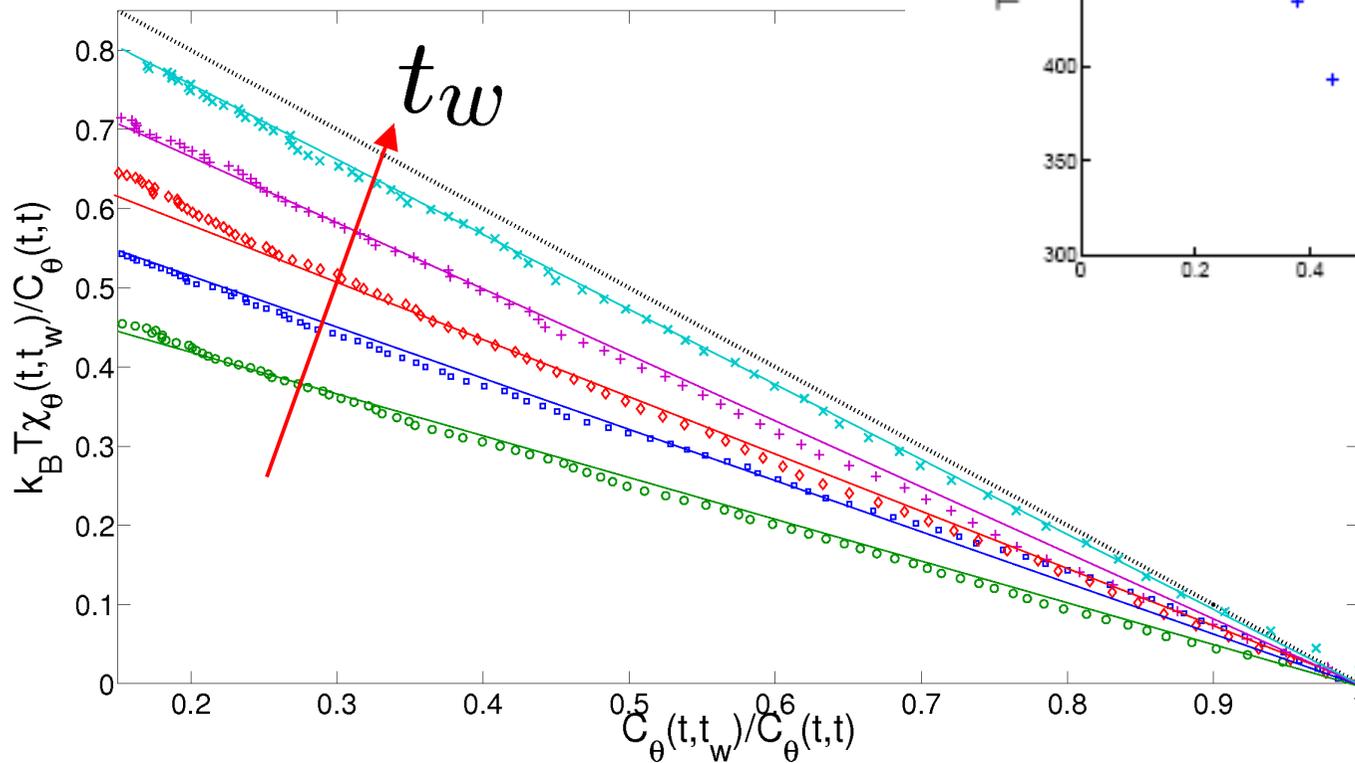
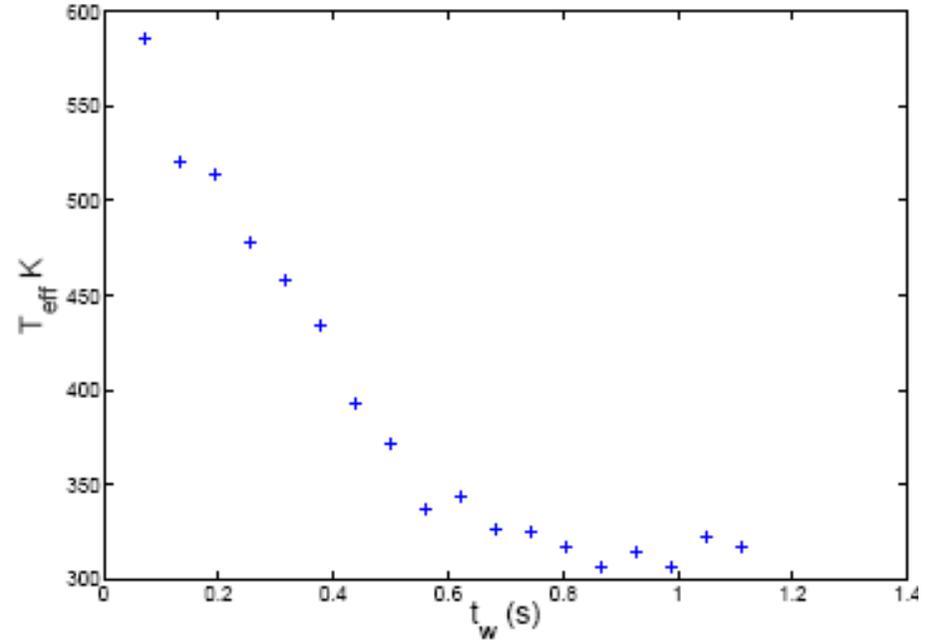
At  $\epsilon_0 = 0, \Rightarrow \tau = \infty$  : the equilibrium interval does not exist.

# FDT out of equilibrium: fixed $t_w$ as a function of $t$

$$\chi(t, t_w) = \frac{X(t, t_w)}{k_B T} (C_\theta(t, t) - C_\theta(t, t_w))$$

with  $T_{eff} = \frac{T}{X(t, t_w)}$

evolution of  $T_{eff}$  versus  $t_w$



# Conclusions

Using a liquid crystal driven by an electric field at the Fréedericksz transition we observe that :

- The probability distribution function of the order parameter fluctuations are well described by a generalized Gumbel, when the correlation length is compared to the probe size.
- After a quench close to the critical point the system presents power law decay. A rescaling similar to the one used in aging materials, produces a master curve of correlations
- FDT is violated during the decay. The observed violation depends on the procedure used to define  $t$  and  $t_w$ .
- For the “good procedure” an asymptotic temperature can be defined, which is not the one computed from mean field.