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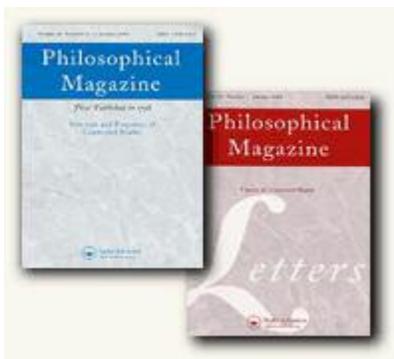
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Finite size effects and aging near a critical point

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Special Issue

Finite size effects and aging near a critical point

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(*Mai 2008*)

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. We show that near the critical value of the field the spatially averaged order parameter has a generalized Gumbel distribution instead of a Gaussian one. The latter is recovered away from the critical point. We also show that after a quench at the critical point the system presents aging properties.

Keywords: Non Gaussian Statistics, Gumbel, Aging, Critical Point, Fluctuations
Dissipations Relations, Finite Size.

1. Introduction

In a second order phase transition, when the control parameter approaches its critical value, the correlation length ξ and the relaxation time τ_r of the order parameter fluctuations diverge. In this context, there are two very interesting phenomena which have been theoretically predicted. The first is an equilibrium property and it appears when ξ , because of the critical divergency, becomes of the order of the measuring region size D . It has been shown that in such a case the probability density function (PDF) of a global variable of the system is not a Gaussian PDF but has a different shape close to a Gumbel one [1–3]. The second phenomenon is an out of equilibrium one and it is related to the divergency of τ_r . It appears when the system is quenched to the critical point by suddenly changing the control parameter. In this case, it has been theoretically predicted that the system presents several properties usually observed in aging materials such as glasses, spin glasses and colloids [4, 5]. In this paper we will present several experimental results that confirm those theoretical predictions briefly summarized in sections 1.1 and 1.2.

1.1. *Distribution of a global quantity close to a critical point*

The fluctuations of global quantities of a system formed by many degrees of freedom have very often a Gaussian distribution. This result is a consequence of the central limit theorem, which is based on the hypothesis that the system under consideration may be decomposed into many uncorrelated domains. However if this hypothesis

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is not satisfied then the PDF of global quantities may take a different form. This is the typical situation when ξ becomes of the order of D . In this case when only the fluctuations of a few coherent domains are measured, it has been derived [2] that the PDF, $P_a(\zeta)$, of a global quantity ζ takes under certain conditions a form which is very well approximated by :

$$P_a(\zeta) = \frac{a^a b_a}{\Gamma(a)} \exp\{-a [b_a(\zeta - s_a) - \exp(-b_a(\zeta - s_a))]\}. \quad (1)$$

where $\Gamma(a)$ is the Gamma function. The only free parameter of $P_a(\zeta)$ is a because b_a and s_a are fixed by the mean $\langle \zeta \rangle$ and the variance σ_ζ^2 of ζ . Furthermore a is determined by the skewness. This distribution $P_a(\zeta)$, named the generalized Gumbel distribution (GG), is for a integer the PDF of the fluctuations of the a^{th} largest value for an ensemble of N random and identically distributed numbers. Instead the interpretation of a non integer is less clear and has been discussed in ref. [3]. It has been shown that the GG appears in many different physical systems where finite size effects are important; however many of these systems are mainly theoretical models [1–3]. In section 2) of this article we will show that close to the critical point of a second order phase transition, the PDF of a spatially averaged order parameter takes the GG form when the correlation length is comparable to the size of the measuring region. The Gaussian distribution is recovered when the system is driven away from the critical point. We also stress that the deviation to the Gaussian PDF are produced by very slow frequencies.

1.2. Aging at critical point

Colloids and glasses may present extremely slow relaxations (often not exponential) towards equilibrium which some time may last much more than any reasonable observation time, in other words they age. This aging is often characterized by power law decay of correlation functions and by the violation of the Fluctuation Dissipation Theorem (FDT) [6]. It has been recently shown that such a kind of behavior is not ubiquitous of aging materials but it may appear in a second order phase transition after a quench to the critical point because of the divergency of the relaxation time [4, 5]. The simplest way to understand this point is to consider the mean-field approximation for a second order phase transition in which θ_0 is the order parameter and ϵ the reduced control parameter :

$$\tau_0 \frac{d\theta_0}{dt} = \epsilon \theta_0 - \alpha \theta_0^3 \quad (2)$$

where τ_0 is a characteristic time and α a parameter which depends on the physical properties of the system. If at $t = 0$ the control parameter is suddenly changed from $\epsilon_0 > 0$ to $\epsilon = 0$ then it can be easily shown that the dynamics of θ_0 is :

$$\langle \theta_0^2 \rangle = \frac{\tau_0}{2 \alpha (t + \frac{\tau_0}{2\epsilon_0})} \quad (3)$$

where $\langle . \rangle$ stand for ensemble average. For very large time t the relaxation of θ_0 is a power law, *i.e.* exactly at the critical point the dynamics never relaxes [5]. During this aging the correlation function $C(t, t - \tau) = \langle \delta\theta_0(t)\delta\theta_0(t - \tau) \rangle$ of the fluctuations $\delta\theta_0 = \theta_0 - \langle \theta_0(t) \rangle$ is a function of both t and τ , with $0 < \tau < t$. It has been shown that $C(t, \tau)$ has not a simple exponential relaxation [5]. The response $\chi(t, t - \tau)$ of the system to an external perturbation is also a complex

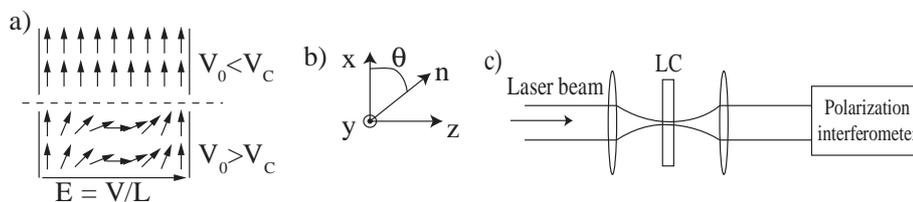


Figure 1. a) The geometry of Fréedericksz transition : director configuration for $V_0 < V_c$ and director configuration for $V_0 > V_c$. b) Definition of angular displacement θ of one nematic \vec{n} . c) Experimental setup. A polarized laser beam is focused into the LC cell and a polarization interferometer measures the phase shift between the ordinary an extraordinary rays [9, 12].

function of t and τ . It turns out that the fluctuation dissipation ratio $X(t, t - \tau) = k_B T \chi(t, t - \tau) / (-\partial_\tau C(t, t - \tau))$ (k_B being the Boltzmann constant and T the temperature) is not a constant as in equilibrium where $X = 1$. In other words, at $\epsilon = 0$ the Fluctuation Dissipation Theorem does not hold as in several aging systems [6]. Experimentally it is of course impossible to drive the system exactly at $\epsilon = 0$ therefore there will be exponential corrections to these power laws. In section 4) of this article we will show that in spite of the presence of these experimental corrections these aging effects can be experimentally observed close to the critical point of a second order phase transition.

2. Experimental set-up

The theoretical prediction shortly summarized in sections 1.1 and 1.2 have been studied using the Fréedericksz transition of a liquid crystal (LC) submitted to an electric field \vec{E} [7, 8]. In this system the measured global variable ζ is the spatially averaged alignment of the LC molecules, whose local direction of alignment is defined by the unit vector \vec{n} . The LC is confined between two parallel glass plates at a distance L (see fig. 1). The inner surfaces of the confining plates have transparent electrodes, used to apply the electric field. Furthermore the plate surfaces, are coated by a thin layer of polymer mechanically rubbed in one direction. This surface treatment causes the alignment of the LC molecules in a unique direction parallel to the surface (planar alignment), *i.e.* all the molecules have the same director parallel to x -axis and $\vec{n} = (1, 0, 0)$ (see fig. 1). The LC is submitted to an electric field perpendicular to the plates. To avoid the electrical polarization of the LC, the electric field has a zero mean value which is obtained by applying a sinusoidal voltage V at a frequency of 1 kHz between the electrodes : $V = \sqrt{2}V_0 \cos(2\pi \cdot 1000 \cdot t)$ [7, 8]. When V_0 exceeds a critical value V_c the planar state becomes unstable and the LC molecules, except those anchored to the glass surfaces, try to align parallel to the field, *i.e.* the director, away from the confining plates, acquires a component parallel to the applied electric field (z -axis) (see fig. 1a)). This is the Fréedericksz transition which is a structural transformation whose properties are those of a second order phase transition [7, 8]. For V_0 close to V_c the motion of the director is characterized by its angular displacement θ in xz -plane (fig. 1b)), whose space-time dependence has the following form : $\theta = \theta_0(x, y, t) \sin(\frac{\pi z}{L})$ [7, 8, 10]. If θ_0 remains small then its dynamics is described by the Ginzburg-Landau equation (eq.2) and one expects mean-field critical phenomena [7, 8, 10], in which θ_0 is the order parameter and $\epsilon = \frac{V_0^2}{V_c^2} - 1$ is the reduced control parameter. The global variable of interest is the spatially averaged alignment of the molecules, precisely $\zeta = \frac{2}{L} \int_0^L \langle (1 - n_x^2) \rangle_{xy} dz \simeq \iint_A \theta_0^2 dx dy / A$, where $A = \pi D^2 / 4$ is the area of the measuring region of diameter D in the (x, y) plane and $\langle . \rangle_{xy}$ stands for

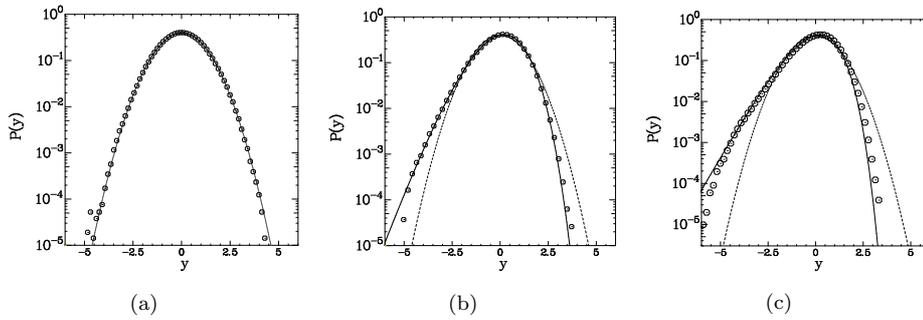


Figure 2. a),b),c) PDF of $y = \frac{\zeta - \langle \zeta \rangle}{\sigma}$ at $\epsilon \sim 0.16, 4 \cdot 10^{-3}$, and $2 \cdot 10^{-3}$ respectively. Dashed line is a Gaussian fit. In b) and c) the continuous lines are the GG distributions with $a = 6.6$, and 2.95 respectively.

mean on A [9]. As ζ is a global variable of this system, its fluctuations, induced by the thermal fluctuations of θ_0 , depend on the ratio between D and the correlation length ξ of θ_0 . The angle θ_0 is a fluctuating quantity whose correlation length and correlation time are respectively: $\xi = L(\pi\sqrt{\epsilon})^{-1}$ and $\tau_r = \tau_0/\epsilon$ where τ_0 is a characteristic time which depends on the LC properties and L^2 [7, 8, 10]. The variable ζ is measured using the experimental set-up shown in fig. 1c). The confocal alinement of the two lenses is very important because the focal length of the lenses determines the diameter D of the laser beam inside the cell. We have performed experiments in cells with three different thickness $L = 25\mu m, L = 20\mu m$ and $L = 6.7\mu m$. The results reported here are those of the thinner cell. The cells are filled by a LC having a positive dielectric anisotropy ϵ_a (p-pentyl-cyanobiphenyl, 5CB, produced by Merck). For this LC $V_c = 0.720$ V and $\tau_o = 55$ ms in the cell with $L = 6.7 \mu m$. We first check the accuracy of our experimental setup by measuring the average $\langle \zeta \rangle$ of the global variable ζ and compare it to the results of a mean-field theory (eq. 2). The experimental results are in very good agreement with theoretical predictions based on the Ginzburg-Landau equation using the physical properties of this LC without adjustable parameters. This excludes the presence of the weak anchoring effects described in literature [8]. The inverse $1/\tau_r$ of the measured relaxation time decreases with ϵ with a linear behavior as predicted by the Ginzburg-Landau model, i.e. $1/\tau_r = \epsilon/\tau_o$, where the value of τ_0 agrees with that obtained from the LC parameters [9].

3. Finite size effects on the probability distribution functions

Many aspects of the director fluctuations, such as power spectra and correlation lengths, at Fréedericksz transition have been widely studied both theoretically [7, 8, 10] and experimentally [11]. Here we focus on the statistical properties of ζ as a function of the ratio $N_{eff} = D/\xi$. We consider the normalized order parameter : $y = \frac{\zeta - \langle \zeta \rangle}{\sigma}$ where σ^2 is the variance of ζ . The probability density functions of y are plotted in fig. 2 for three different values of ϵ . We find that far from the critical value ($\epsilon = 0.16$) the distribution is Gaussian (fig. 2a)). In contrast, for a value of ϵ closer to 0, typically $\epsilon \sim 2 \cdot 10^{-3}$, the PDF of fluctuations of ζ are not Gaussian as it is clear from fig. 2 c). In figure 2b), we plot the distribution of ζ for an intermediate value of ϵ . The exponential tail becomes more pronounced when ϵ decreases. We want now to compare this distribution to a GG (eq. (1)). The value of the free parameter a is given by the skewness of the fluctuations: $\gamma \sim -1/\sqrt{a}$. We obtain $a = 2.95$ at $\epsilon \sim 2 \cdot 10^{-3}$, $a = 6.6$ at $\epsilon \sim 4 \cdot 10^{-3}$. Using these values in eq. (1), we get the PDFs plotted in fig 2 as continuous lines which agree quite well with the

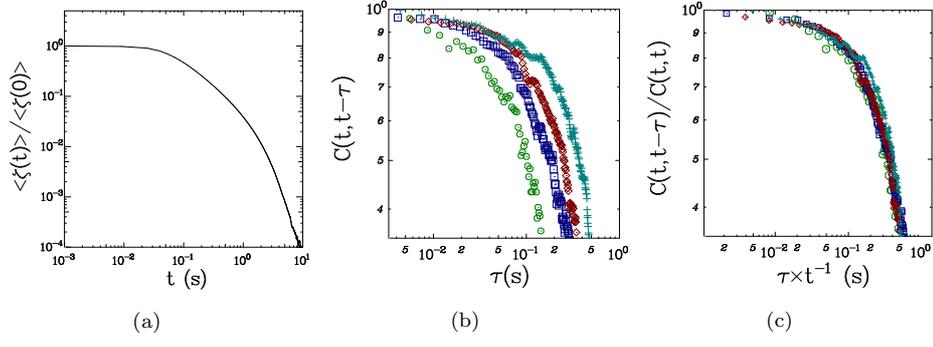


Figure 3. a) Mean response $\langle \zeta \rangle$ of the system after a quench. b) Correlation functions of the fluctuations of ζ measured at different times t after the quench as a function of τ . c) The same correlations as in a) but as a function of τ/t . The correlation functions collapse on a master curve in this case.

experimental distributions. The observation of the GG for ϵ very close to 0 confirm the theoretical predictions outlined in section 1.1. The deviation to the gaussian is mainly given by the slow modes indeed by high-pass filtering the time evolution of ζ acquired at $\epsilon = 2 \cdot 10^{-3}$. A Gaussian behavior is retrieved for $f_{HP} 10/\tau_r$. These experimental results indicate that the slow modes are responsible for the non-gaussian PDF of ζ . Let us now consider the correlation length ξ of θ_0 which has to be compared to the diameter of the measuring volume D . At $\epsilon = 0.002$, we find $\xi = 47 \mu\text{m}$, that is $\xi \sim D/3$. In other words the laser detects the fluctuations of only a few coherent domains and, in agreement with the theoretical predictions these fluctuations have the GG distribution (see section 1.1). We observe that γ goes to zero for increasing ϵ and the Gaussian behavior is retrieved for $\epsilon 0.03$. The inverse of γ is linear in ϵ , that is $\gamma^{-1} = -\sqrt{a} = p + q\epsilon = p + \tilde{q}N_{\text{eff}}^2$ as for the theoretical model (see ref.[9] for details).

4. Quench at the critical point

We now consider the dynamic of ζ as a function of time after a quench at $\epsilon \simeq 0$ from an initial value $\epsilon_o \simeq 1$. The fact that the control parameter is the electric field allows one to have extremely fast quenches. The typical mean response of the system after the quench is plotted in fig. 3a) as a function of the time t after the quench. The behavior of ζ is very close to the one predicted by eq.3. It remains constant for a very long time and then slowly relaxes. However as the relaxation time is very long the system ages for a very long time of about 4 order of magnitude after quench. This curve is obtained by repeating the experiment several times. Then we determine the mean time dependence $\langle \zeta(t) \rangle$ (plotted in fig. 3 a) and the fluctuations $\delta\zeta(t) = \zeta(t) - \langle \zeta(t) \rangle$ around this mean. The correlation functions $C(t, t - \tau) = \langle \delta\zeta(t)\delta\zeta(t - \tau) \rangle$ are also obtained as ensemble average on the different quenches because it is not possible to average in time as the statistics is not stationary. The computed correlations are plotted as function of τ for different t in fig. 3. We see that the decay of the correlations changes as a function of t . However it is possible to superpose all the correlation functions if they are plotted as a function of τ/t . This is a typical behavior of correlation function which is observed in many numerical models of aging materials and for example in the scaling of response function in spin glasses. In our system with a rather complex procedure [13], too complicate to be explained in such a short note, we measure the response function of ζ to an external perturbation. By measuring the response of the system to an external perturbation one can also determine the Fluctuation

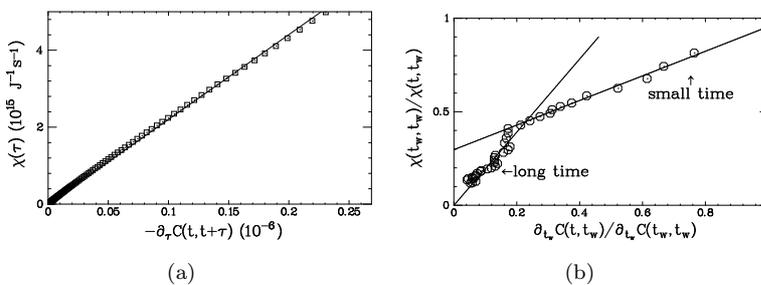


Figure 4. Response function versus the time derivative of the correlation functions of the fluctuations of ζ . a) $\epsilon = 0.01$ in equilibrium. b) $\epsilon = 0.003$ at $t_w = 0.5$ s.

Dissipation Ratio $X(t, t - \tau)$. In fig. 4 we plot $\chi(t, t - \tau)$ versus $-\partial_\tau C(t, t - \tau)$ in equilibrium (fig. 4a) and after a quench (fig. 4b). We see that in equilibrium the FDR is constant and equal 1 whereas the change of slope in fig. 4b) indicates that the Fluctuation Dissipation Theorem is violated during this aging dynamics at critical point. This effect has been predicted theoretically but never observed in a real system close to a critical point.

5. Conclusions

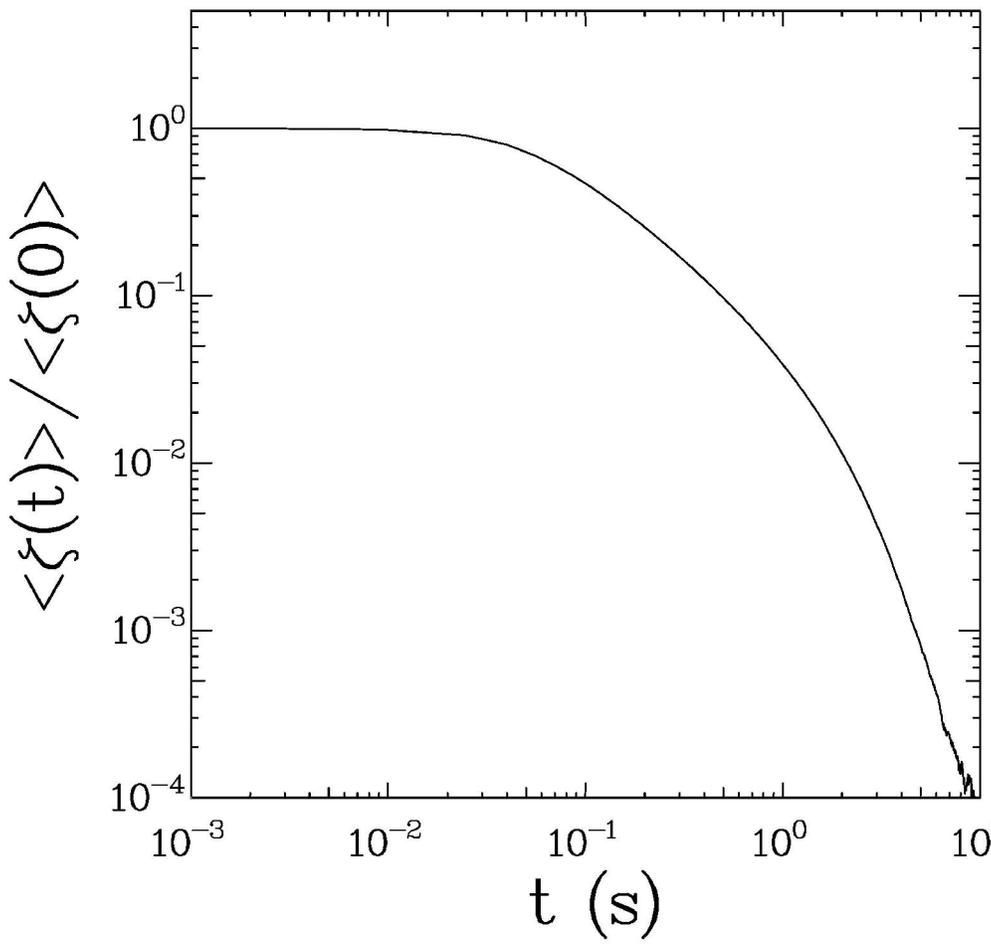
In conclusion, we have experimentally shown, using the Fréedericksz transition of a LC, that in a second order phase transition the fluctuations of a spatially extended quantity have a GG distribution if the coherence length is of the order of the size of the measuring area. Furthermore we have also shown that in this LC transition it is also possible to observe after a fast quench at the critical point many properties which are similar to those observed during the aging of glasses and colloids. This study on aging near a critical point, which will be presented in more details in a longer report, is extremely useful to fix the limits of the experimental set-up when measuring fluctuations in aging materials.

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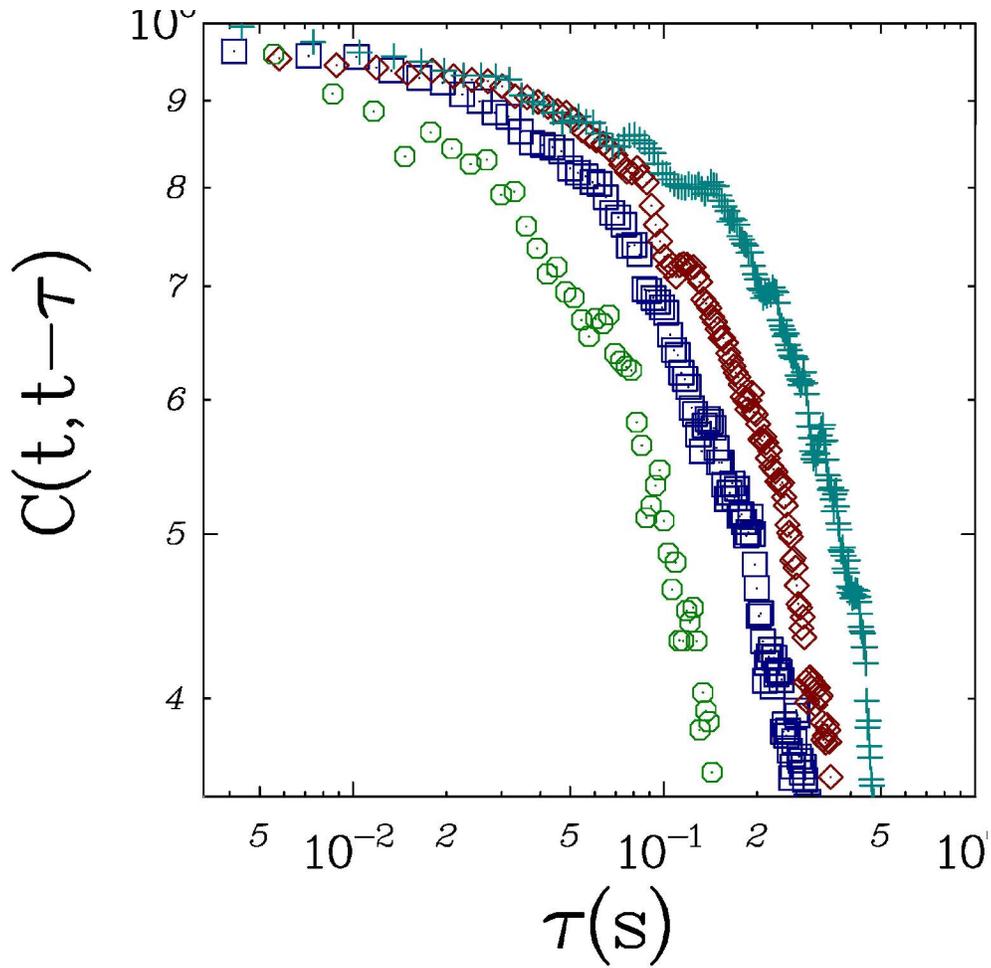
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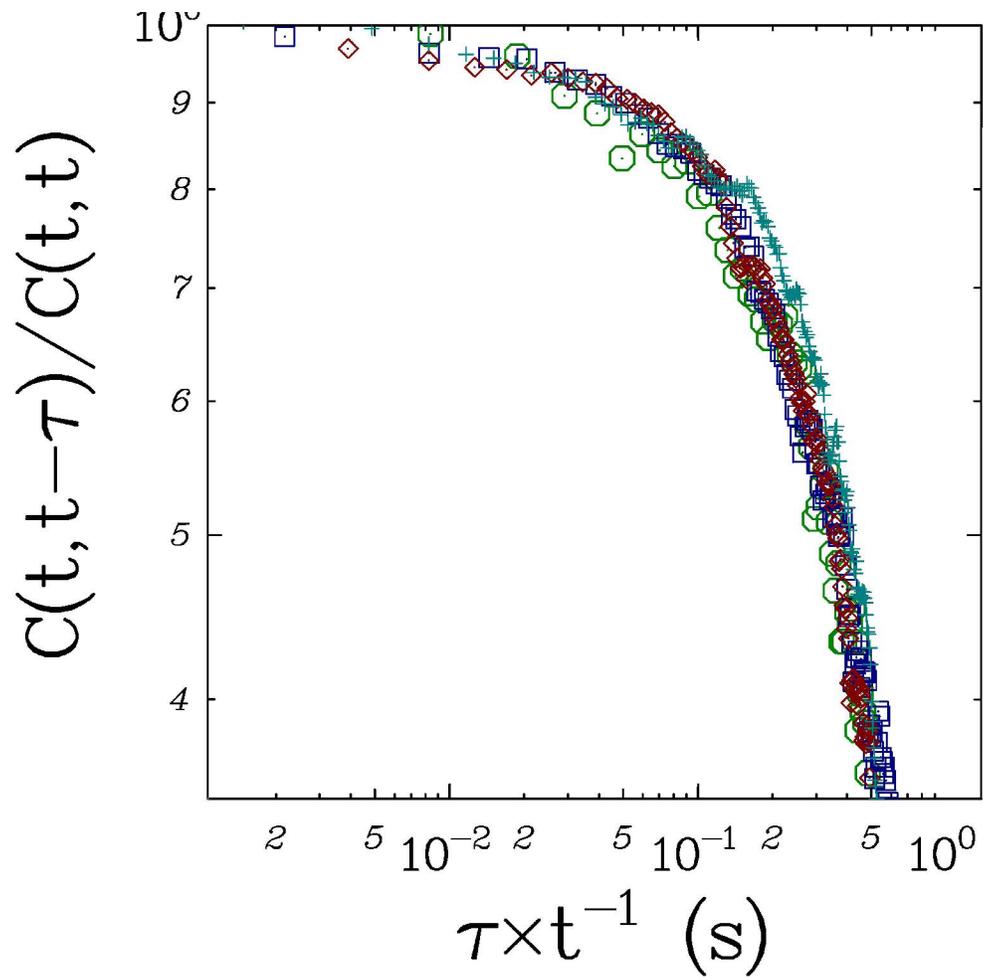


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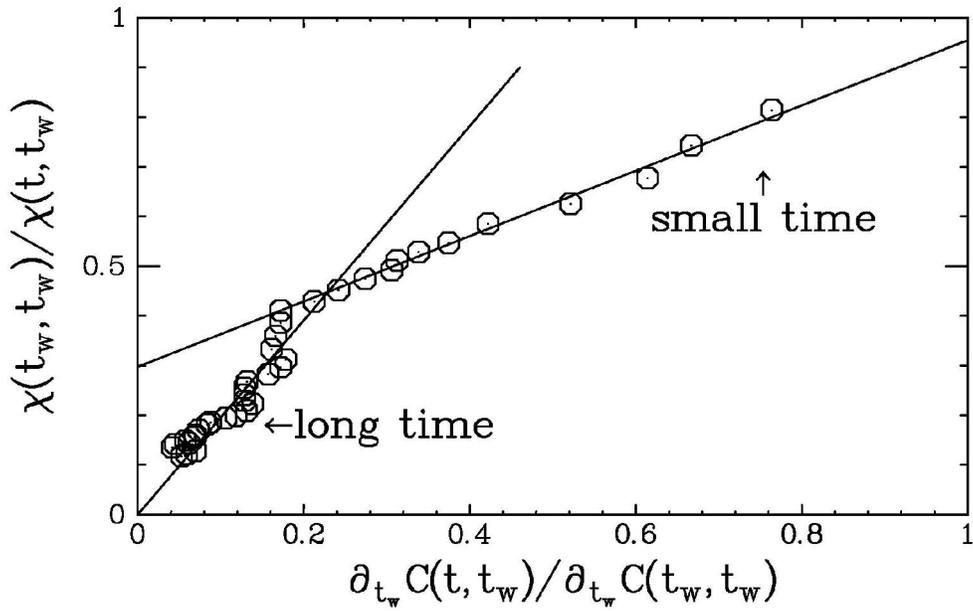
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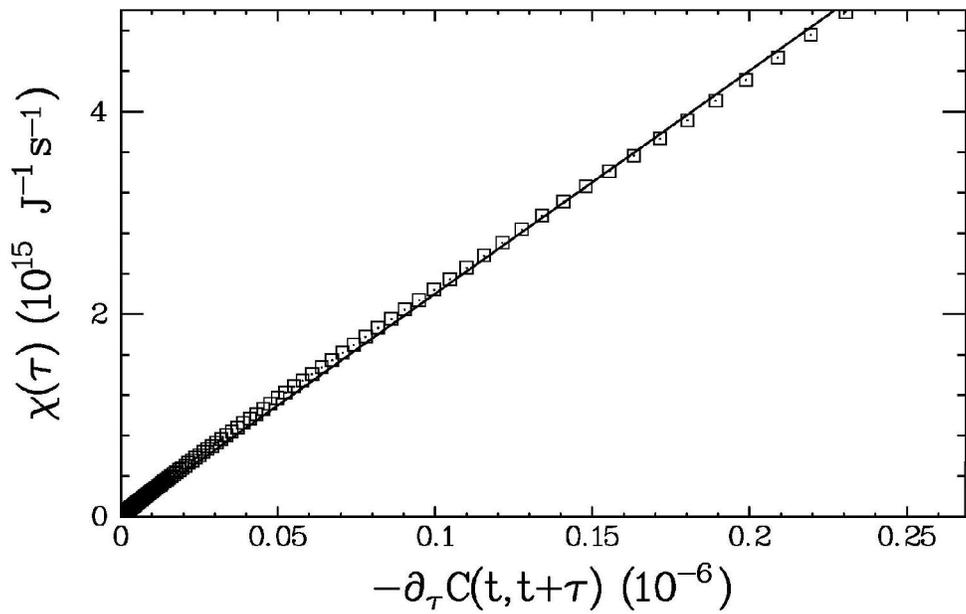


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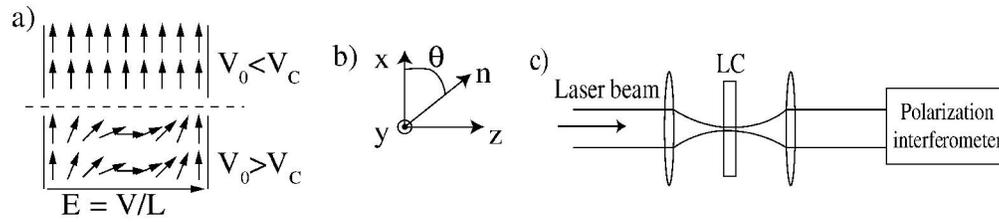
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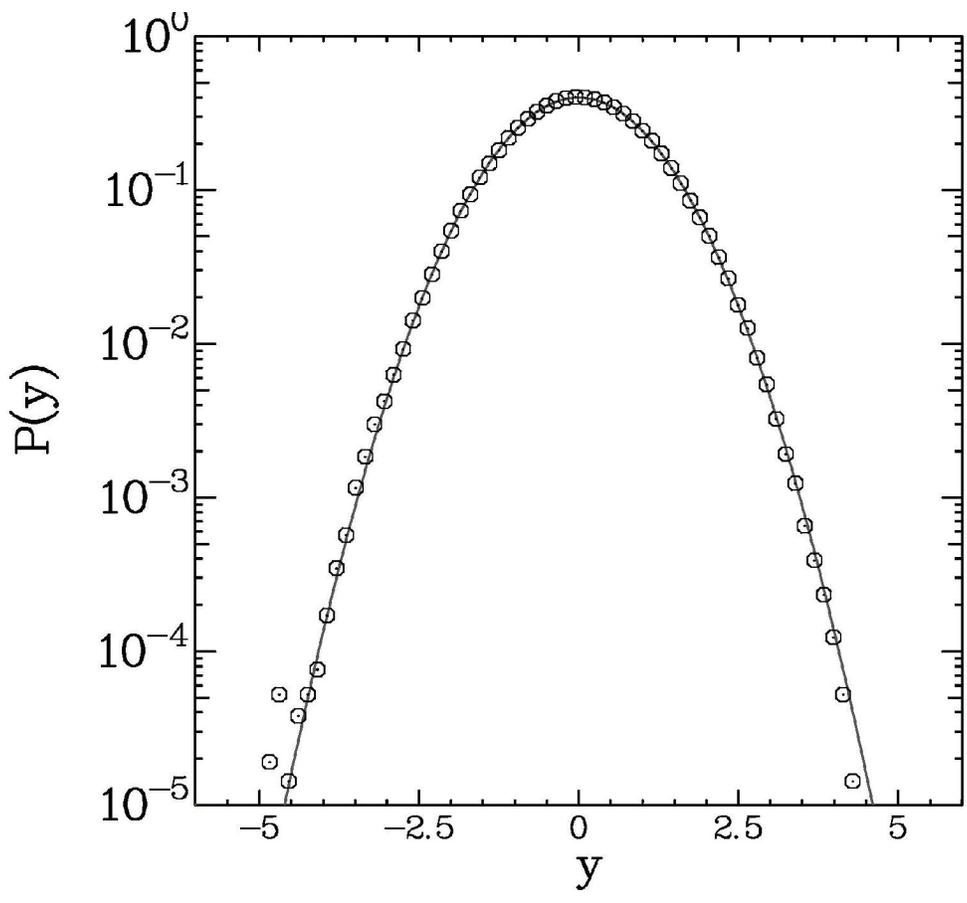


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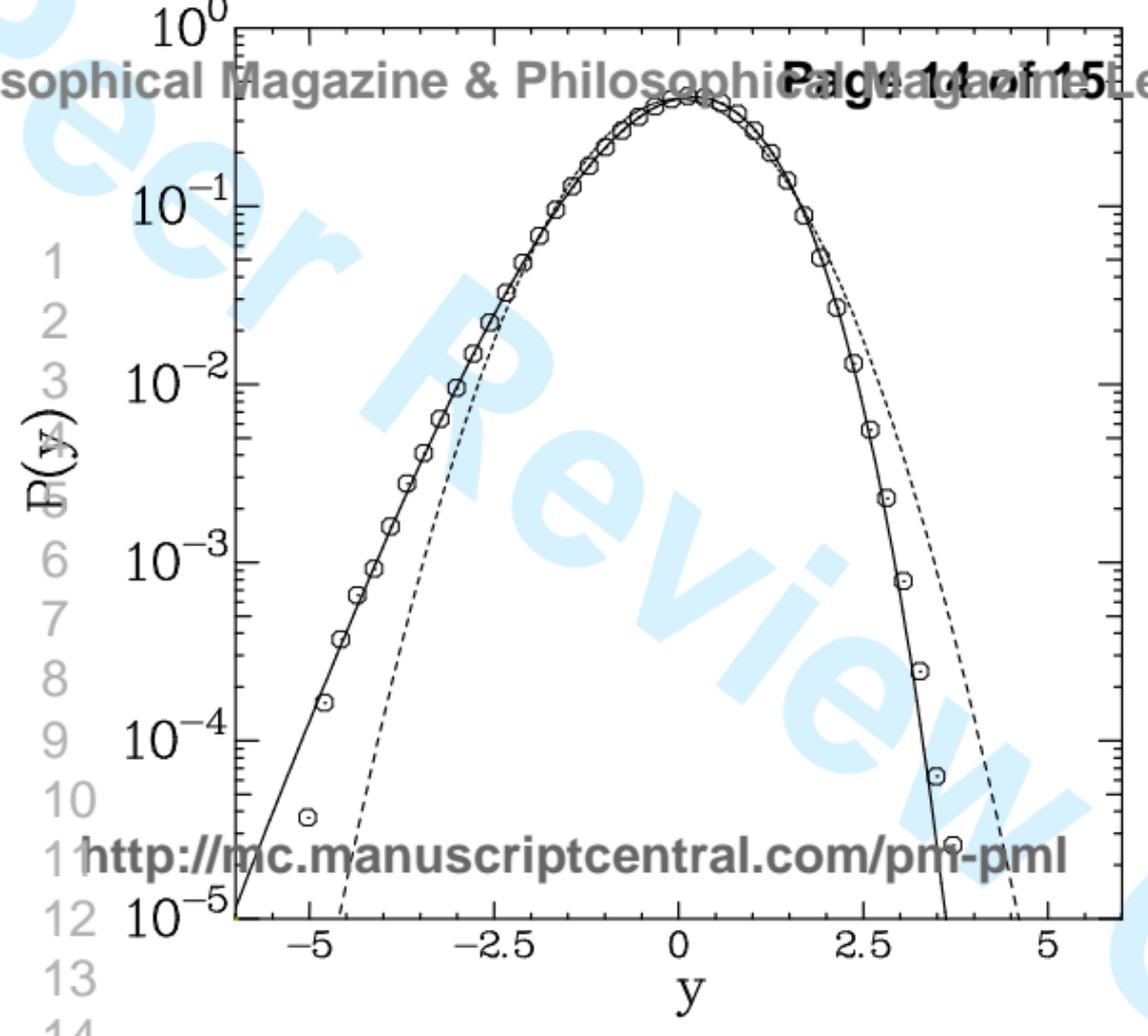
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