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# Experimental evidence of non-Gaussian fluctuations near a critical point.

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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. The relevance of slow modes is pointed out. The parameter of generalized Gumbel is related to the effective number of degrees of freedom.

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The fluctuations of global quantities of a system formed by many degrees of freedom have very often a Gaussian probability density function (PDF). 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 is not satisfied then the PDF of global quantities may take a different form. A few years ago it has been proposed [1, 2, 3, 4, 5, 6] that in spatially extended systems, where the correlation lengths are of the order of the system size, the PDF,  $P_a(\chi)$ , of a global quantity  $\chi$  takes under certain conditions [6] a form which is very well approximated by :

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

The only free parameter of  $P_a(\chi)$  is  $a$  because  $b_a$  and  $s_a$  are fixed by the mean  $\langle\chi\rangle$  and the variance  $\sigma_\chi^2$  of  $\chi$  :

$$b_a = \frac{1}{\sigma_\chi} \sqrt{\frac{d^2 \ln \Gamma(a)}{da^2}}, \quad s_a = \langle\chi\rangle + \frac{1}{b_a} \left( \ln a - \frac{d \ln \Gamma(a)}{da} \right) \quad (2)$$

where  $\Gamma(a)$  is the Gamma function. This distribution  $P_a(\chi)$ , named the generalized Gumbel distribution (GG), is for  $a$  integer the PDF of the fluctuations of the  $a^{\text{th}}$  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. [7]. For  $a = \pi/2$ , the distribution  $P_a$  is approximately the BHP distribution (from Bramwell, Holdsworth, Pinton). It has been shown in ref. [5, 7] that the GG appears in many different physical systems where finite size effects are important. An example of these non Gaussian fluctuations is the magnetization of the two dimensional XY model which presents a Kosterlitz-Thouless transition as a function of temperature. When the control parameter is close to the critical value, the correlation length of the system diverges and when it becomes of the order of the system size, then the PDF of the fluctuations of the magnetization has a GG form instead of the Gaussian one [1]. Several other examples where the GG gives

good fits of the PDF of the fluctuations of global parameters are : the magnetization in Ising model close to the critical temperature, the energy dissipated in the forest fire model, the density of relaxing sites in granular media models and the power injected in a turbulent flow and in electroconvection [1, 2, 3, 4, 5, 6, 7, 8]. Except for the two last examples which use experimental data all of the other mentioned results are obtained on theoretical models. Therefore it is of paramount importance to check whether the above mentioned theoretical predictions on GG can be observed experimentally in other phase transitions. We report in this letter the first experimental evidence 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.

In our experiment, these properties of global variables have been studied using the Fréedericksz transition of a liquid crystal (LC) submitted to an electric field  $\vec{E}$  [9, 10]. In this system the measured global variable  $\chi$  is the spatially averaged alignment of the LC molecules, whose local direction of alignment is defined by the unit vector  $\vec{n}$ . Let us first recall the general properties of the Fréedericksz transition. The system under consideration is a LC confined between two parallel glass plates at a distance  $L$  (see fig. 1). The inner surfaces of the confining plates have transparent ITO 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) [11]. 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 volt-

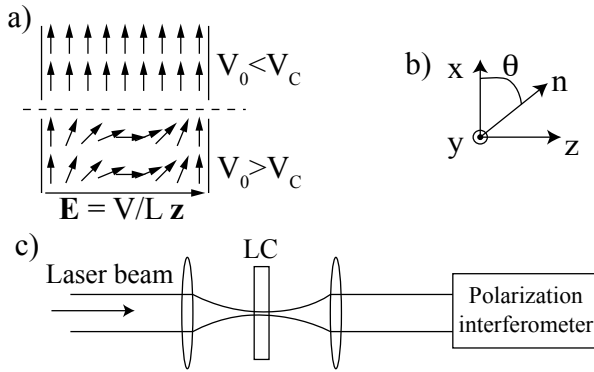


FIG. 1: a) The geometry of Fréedericksz transition : director configuration for  $V_0 < V_c$  and director configuration for  $V_0 > V_c$ . b) Definition of angular displacement  $\theta$  of one nematic  $\vec{n}$ . c) Experimental setup. A polarized laser beam is focused into the LC cell and a polarization interferometer measures the phase shift  $\Phi$  between the ordinary and extraordinary rays [17].

age  $V$  at a frequency of 1 kHz between the ITO electrodes, i.e.  $V = \sqrt{2}V_0 \cos(2\pi \cdot 1000 \cdot t)$  [9, 10]. 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 [9, 10]. For  $V_0$  close to  $V_c$  the motion of the director is characterized by its angular displacement  $\theta$  in  $xz$ -plane (fig. 1b)), whose space-time dependence has the following form :  $\theta = \theta_0(x, y, t) \sin(\frac{\pi z}{L})$  [9, 10, 12]. If  $\theta_0$  remains small then its dynamics is described by a Ginzburg-Landau equation and one expects mean-field critical phenomena [9, 10, 12], in which  $\theta_0$  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  $\chi = \frac{2}{L} \int_0^L \langle (1 - n_x^2) \rangle_{xy} dz \simeq \iint_A \theta_0^2 dx dy / A$ , where  $A = \pi D^2 / 4$  is the area of the measuring region of diameter  $D$  in the  $(x, y)$  plane and  $\langle \cdot \rangle_{xy}$  stands for mean on  $A$ . As  $\chi$  is a global variable of this system, its fluctuations, induced by the thermal fluctuations of  $\theta_0$ , depend on the ratio between  $D$  and the correlation length  $\xi$  of  $\theta_0$ . The angle  $\theta_0$  is a fluctuating quantity whose correlation length and correlation time are respectively:  $\xi = L(\pi\sqrt{\epsilon})^{-1}$  and  $\tau = \tau_0/\epsilon$  where  $\tau_0$  is a characteristic time which depends on the LC properties and  $L^2$  [9, 10, 12]. Many aspects of the director fluctuations, such as power spectra and correlation lengths, at Fréedericksz transition have been widely studied both theoretically [9, 10, 12] and experimentally [13, 14, 15, 16]. However the statistical properties of the spatially averaged director fluctuations have never been characterized as a function of the ratio  $N_{eff} = D/\xi$ .

As this ratio is the key parameter of our study, we have performed the experiment 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 mainly those of the thinner cell and a detailed comparison with those of the others will be the aim of a longer paper. The cells are filled by a LC having a positive dielectric anisotropy  $\epsilon_a$  (p-pentylcyanobiphenyl, 5CB, produced by Merck). For this LC  $V_c = 0.720$  V and  $\tau_0 = 55$  ms in the cell with  $L = 6.7\mu m$ .

Let us describe now how  $\chi$  has been measured. The deformation of the director field produces an anisotropy of the refractive index of the LC cell. This optical anisotropy can be precisely estimated by measuring the optical path difference  $\Phi$  between a light beam crossing the cell linearly polarized along  $x$ -axis (ordinary ray) and another beam crossing the cell polarized along the  $y$ -axis (extraordinary ray). The experimental set-up employed is schematically shown in fig. 1c). The beam is produced by a stabilized He-Ne laser ( $\lambda = 632.8$  nm) and focused into the liquid crystal cell by a converging lens (focal length  $f = 160$  mm). A second lens with the same focal length is placed after the cell to have a confocal optical system, which insures that inside the cell the laser beam is parallel and has a diameter  $D$  of about  $125\mu m$ . The beam is normal to the cell and linearly polarized at  $45^\circ$  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 polarisation interferometer [17]. After some algebra the phase shift  $\Phi$  is given by :

$$\Phi = \left\langle \frac{2\pi}{\lambda} \int_0^L \left( \frac{n_o n_e}{\sqrt{n_o^2 \cos^2(\theta)^2 + n_e^2 \sin^2(\theta)^2}} - n_o \right) dz \right\rangle_{xy} \quad (3)$$

with  $(n_o, n_e)$  the two anisotropic refractive indices [9, 10]. In term of  $\chi$ , we get :

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

The phase  $\Phi$ , measured by the interferometer, is acquired with a resolution of 24 bits at a sampling rate of 1024 Hz. The instrumental noise of the apparatus [17] is three orders of magnitude smaller than the amplitude  $\delta\Phi$  of the fluctuations of  $\Phi$  induced by the thermal fluctuations of  $\chi$ .

We first check the accuracy of our experimental setup by measuring the time-average  $\langle \chi \rangle$  of the global variable  $\chi$  and compare it to the results of a mean-field theory. In figure 2, we plot the measured  $\langle \chi \rangle$  versus the control parameter  $\epsilon$ .  $\langle \chi \rangle$  vanishes for  $\epsilon < 0$  and increases for  $\epsilon > 0$ . 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 pres-

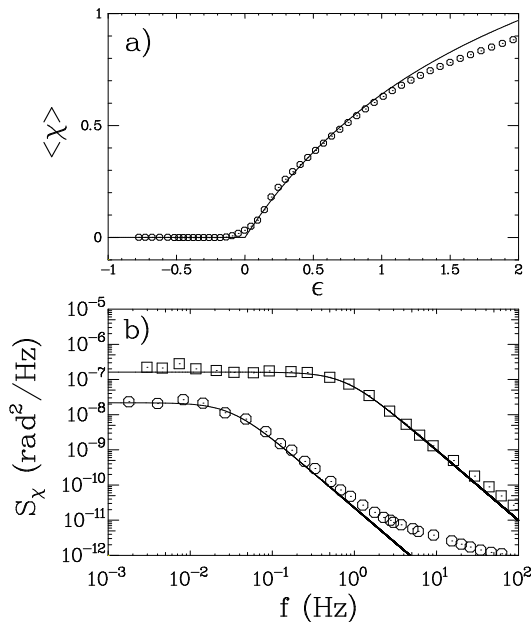


FIG. 2: a) Average value of the the global variable  $\chi$  as a function of  $\epsilon$  ( $\circ$ ). Continuous line is a theoretical prediction based on the Ginzburg-Landau equation for  $\theta_0$  using the values of this LC, with no adjustable parameters. b) Power spectrum  $S_\chi$  and the Lorentzian fit (continuous line) measured at  $\epsilon = 2 \cdot 10^{-3}$  ( $\circ$ ) and at  $\epsilon = 0.16$  ( $\square$ ).

ence of the weak anchoring effects described in literature [10, 11]. We observe that the model is valid even for large values of  $\epsilon$ . The rounding near the transition is a finite size effect because in our experiment  $N_{eff} \simeq 1$  for  $\epsilon \simeq 0$ . Indeed cells with the larger  $L$  show even a more pronounced rounding effect close to the transition.

To shed some light on the dynamics of the fluctuations, we first measure the power spectral density  $S_\chi$  of  $\chi$ . As the slow thermal drift of the interferometer may perturb the statistics of the acquired signals,  $\chi$  is high-pass filtered at 2 mHz. The power spectrum  $S_\chi$ , measured at  $\epsilon = 0.16$  and  $\epsilon = 0.002$ , are plotted in figure 2. They can be fitted by a Lorentzian for  $\epsilon > 0$  :

$$S_\chi = \frac{S_0(\epsilon)}{1 + (f/f_c(\epsilon))^2} \quad (5)$$

$S_0(\epsilon)$  represents the amplitude of fluctuations and  $f_c(\epsilon)$  is proportional to the inverse of the relaxation time  $\tau(\epsilon)$  of  $\theta_0$  ( $f_c = (\pi\tau)^{-1}$ ). This form is the same found by Galatola for light-scattering measurements [14, 15] but we have increased the resolution at low frequencies of about three orders of magnitude. The values of  $S_0$  and  $f_c$  are obviously dependent on  $\epsilon$  and its sign. For  $\epsilon < 0$ ,  $S_\chi$  is the sum of two Lorentzian functions with two cut-off frequencies. Each frequency corresponds to a relaxation of the director of the LC in two different directions. The lowest frequency, which corresponds to  $\theta$ , depends on  $\epsilon$  contrary to the other frequency. The cut-off frequency

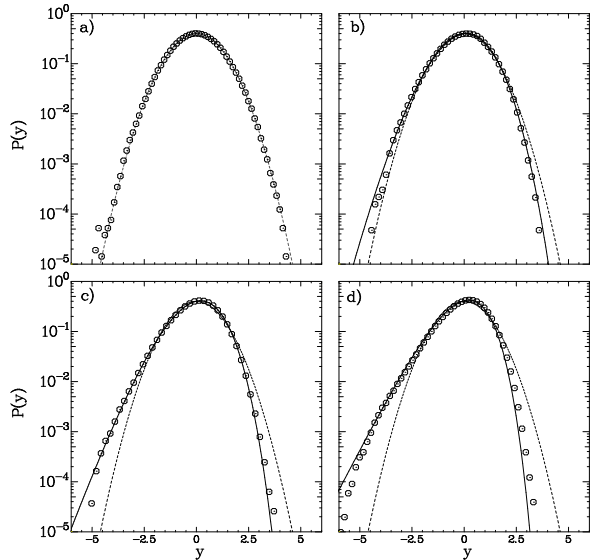


FIG. 3: a),b),c),d) PDF of  $y = \frac{\chi - \langle \chi \rangle}{\sigma}$  at  $\epsilon \sim 0.16$ ,  $8 \cdot 10^{-3}$ ,  $4 \cdot 10^{-3}$ , and  $2 \cdot 10^{-3}$  respectively. Dashed line is a Gaussian fit. In b), c) and d) the continuous lines are the GG distributions with  $a = 23.5$ , 6.6, and 2.95 respectively.

$f_c$  decreases with  $\epsilon$  with a linear behavior as predicted by the Ginzburg-Landau model, i.e.  $1/\tau = \epsilon/\tau_0$ , where the value of  $\tau_0$  agrees with that obtained from the LC parameters. The amplitude  $S_0$  has a complex dependence on  $\epsilon$ . This dependence, which can be understood on the basis of the Ginzburg-Landau model, is not relevant for the results presented in this letter and will be discussed in a longer report.

We now turn to the main point of this letter that is the statistical description of the fluctuations of  $\chi$ . We consider the normalized order parameter :  $y = \frac{\chi - \langle \chi \rangle}{\sigma}$  where  $\sigma^2$  is the variance of  $\chi$ . The probability density functions of  $y$  are plotted in Fig. 3 for three different values of  $\epsilon$ . We find that far from the critical value ( $\epsilon = 0.16$ ) the distribution is Gaussian (Fig. 3a)). In contrast, for a value of  $\epsilon$  closer to 0, typically  $\epsilon \sim 2 \cdot 10^{-3}$ , the PDF of fluctuations of  $\chi$  are not Gaussian as it is clear from fig.3 d). In figure 3b) and c), we plot the distribution of  $\chi$  for two intermediate values of  $\epsilon$ . The exponential tail becomes more pronounced when  $\epsilon$  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 [4] :

$$\gamma = \langle y^3 \rangle = -\left(\frac{d^3 \ln \Gamma(a)}{da^3}\right) / \left(\frac{d^4 \ln \Gamma(a)}{da^4}\right)^{3/2} \sim -1/\sqrt{a} \quad (6)$$

We obtain  $a = 2.95$  at  $\epsilon \sim 2 \cdot 10^{-3}$ ,  $a = 6.6$  at  $\epsilon \sim 4 \cdot 10^{-3}$  and  $a = 23.5$  at  $\epsilon \sim 8 \cdot 10^{-3}$ . Using these values in eq. (1), we get the PDFs plotted in fig 3 as continuous lines which agree quite well with the experimental distributions. The observation of the GG for  $\epsilon$  very close to 0 is the main result of this letter. One may wonder

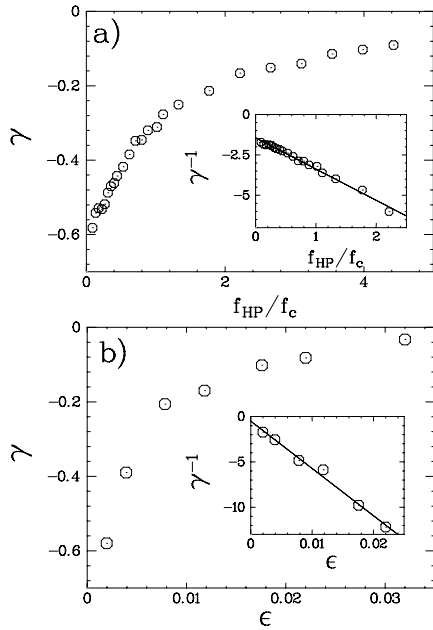


FIG. 4: a) The skewness  $\gamma$  of the fluctuations at  $\epsilon = 2 \cdot 10^{-3}$  is plotted as a function of  $f_{\text{HP}}$ . Inset:  $\gamma^{-1}$  is linear in  $f_{\text{HP}}/f_c$ . b)  $\gamma$  as a function of  $\epsilon$ . Inset:  $\gamma^{-1}$  is linear in  $\epsilon$ .

why the GG is observed in our experiment and not in other experiments on phase transitions. To answer to this question, let us first consider the slow modes of  $\chi$  whose relevance for the GG distribution has been pointed out in ref. [5]. To confirm this point the time evolution of  $\chi$  acquired at  $\epsilon = 2 \cdot 10^{-3}$  is high-passed filtered at various cut-off frequencies  $f_{\text{HP}}$ . The skewness  $\gamma$  of the filtered signal is plotted as a function of  $f_{\text{HP}}$  (figure 4a)). When  $f_{\text{HP}}$  is increased, we see that the skewness decreases ( $\gamma^{-1}$  is linear in  $f_{\text{HP}}$ ). A Gaussian behavior is retrieved for  $f_{\text{HP}} > 10f_c \simeq 0.1$  Hz. These experimental results indicate that the slow modes, with frequency lower than  $f_c$ , are responsible for the non-gaussian PDF of this global parameter. Previous experiments on Fréedericksz transition did not have a sufficient resolution at low frequencies and they erased this effect. Let us now consider the correlation length  $\xi$  of  $\theta_0$  in the plane  $(x, y)$ . This correlation length has to be compared with the diameter of the measuring volume, that, in our experiment, is determined by the laser beam diameter  $D$  inside the cell. 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. The effective number of degrees of freedom of the system is related to the ratio  $N_{\text{eff}} = D/\xi \propto \sqrt{\epsilon}$ . In figure 4b), we plot the values of the skewness as a function of  $\epsilon$ . We observe that  $\gamma$  goes to zero for increasing  $\epsilon$  and the Gaussian behavior is retrieved for  $\epsilon > 0.03$ . The inverse of  $\gamma$  is linear in  $\epsilon$ , that is  $\gamma^{-1} = -\sqrt{a} = p + q\epsilon = p + \tilde{q}N_{\text{eff}}^2$ . We

measure  $p = -0.51$  and  $q = -521$ . Thus the free parameter  $a$  of the GG is a measure of the effective number of degrees of freedom as underlined in ref. [4, 18]. For the magnetization of the two dimensional XY model it has been found that:  $\gamma^{-1} \sim -\sqrt{a} \sim -\sqrt{\frac{2}{\pi}} \left[ 1 + \frac{1}{2} \left( \frac{N_{\text{eff}}}{2\pi} \right)^2 \right]$ .

The dependence of  $\sqrt{a}$  on  $N_{\text{eff}}$  is the same than in our experiment but the coefficients depend on the system. As the  $\xi$  is proportional to the cell thickness, we have verified that for cells having larger  $L$ , the GG is obtained for larger values of  $\epsilon$ . This is indeed the case and the detailed description of the results of the other cells will be the subject of a long article.

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 size of the measuring area. The slow modes, corresponding to large scales, are responsible for this non-Gaussian behavior. This observation confirms several theoretical predictions on GG, which have never been observed before in an experiment on a phase transition.

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