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 the generalized Gumbel distribution 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 was proposed [1–6] 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 χ takes under certain conditions [6] a form which is very well approximated by:

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

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 σ_χ^2 of χ:

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

where $\Gamma(a)$ is the Gamma function. This distribution $P_a(\chi)$, named the generalized Gumbel distribution (GG), is for a an integer the PDF of the fluctuations of the ath largest value for an ensemble of N random and identically distributed numbers. Instead, the interpretation of a non-integer a is less clear and has been discussed in Ref. [7]. For $a = \pi/2$, the distribution P_a is approximately the Bramwell-Holdsworth-Pinton distribution. It has been shown in Refs. [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 [1–8]. 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 deviations to the Gaussian PDF are produced by very slow frequencies.

In our experiment, these properties of global variables have been studied by 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 χ 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 indium-tin-oxide (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 of the molecules have the same director parallel to the x axis and $\vec{n} = (1, 0, 0)$ (see Fig. 1) [11]. The LC is submitted to an electric field

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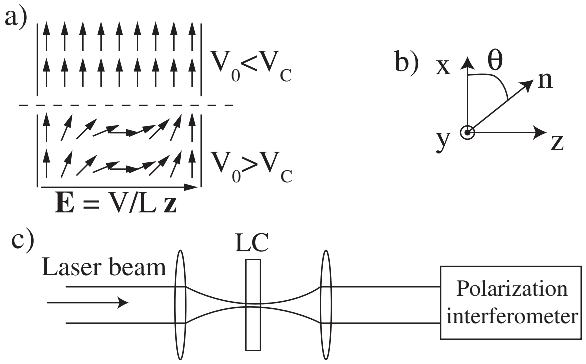

FIG. 1. (a) The geometry of the Fréedericksz transition: director configuration for $V_0 < V_c$ and director configuration for $V_0 > V_c$. (b) Definition of angular displacement θ of one nematic $\vec{n}$. (c) Experimental setup. A polarized laser beam is focused into the LC cell, and a polarization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

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 ITO electrodes, i.e., $V = \sqrt{2}V_0 \cos(2\pi \times 1000t)$ [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. 1(a)]. 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 θ in the xz plane [Fig. 1(b)], whose space-time dependence has the following form: $\theta = \theta_0(x, y, t)\sin(\frac{\pi z}{L})$ [9,10,12]. If θ_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 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 $\chi = \frac{2}{L} \times \int_0^L \langle(1 - n_x^2)\rangle_{xy}\, dz \simeq \iint_A \theta_0^2 dxdy/A$, 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 mean on A. As χ 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 $\tau = \tau_0/\epsilon$, where τ_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 the Fréedericksz transition have been widely studied both theoretically [9,10,12] and experimentally [13–16]. However, the statistical properties of the spatially averaged director fluctuations have never been characterized as a function of the ratio $N_{\rm eff} = D/\xi$. As this ratio is the key parameter of our study, we have performed the experiment in cells with three different thicknesses: $L = 25\ \mu$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 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.

Let us describe now how χ 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 Φ between a light beam crossing the cell linearly polarized along the x axis (ordinary ray) and another beam crossing the cell polarized along the y axis (extraordinary ray). The experimental setup employed is schematically shown in Fig. 1(c). 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 ensures that inside the cell the laser beam is parallel and has a diameter D of about 125 μm. The beam is normal 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 [17]. After some algebra, the phase shift Φ is given by:

$$\Phi = \left\langle \frac{2\pi}{\lambda} \int_0^L \left(\frac{n_o n_e}{\sqrt{n_0^2 \cos(\theta)^2 + n_e^2 \sin(\theta)^2}} - n_0 \right) dz \right\rangle_{xy}, \quad (3)$$

with (n_o, n_e) the two anistotropic refractive indices [9,10]. In terms of χ, we get

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

The phase Φ, 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 3 orders of magnitude smaller than the amplitude $\delta\Phi$ of the fluctuations of Φ induced by the thermal fluctuations of χ.

We first check the accuracy of our experimental setup by measuring the time average $\langle\chi\rangle$ of the global variable χ and compare it to the results of a mean-field theory. In Fig. 2, we plot the measured $\langle\chi\rangle$ versus the control parameter ϵ. $\langle\chi\rangle$ 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 presence of the weak anchoring effects described in literature [10,11]. We observe that the model is valid even for large values of ϵ.

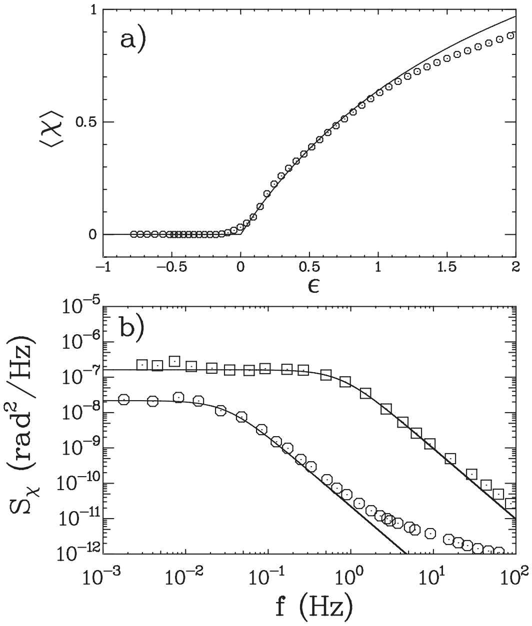

FIG. 2. (a) Average value of the global variable χ as a function of ϵ (\circ). The continuous line is a theoretical prediction based on the Ginzburg-Landau equation for θ_0 using the values of this LC, with no adjustable parameters. (b) Power spectrum S_χ and the Lorentzian fit (continuous line) measured at $\epsilon = 2 \times 10^{-3}$ (\circ) and at $\epsilon = 0.16$ (□).

The rounding near the transition is a finite size effect because in our experiment $N_{\rm eff} \simeq 1$ for $\epsilon \simeq 0$. Indeed, cells with the larger L show an even 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_χ of χ. As the slow thermal drift of the interferometer may perturb the statistics of the acquired signals, χ is high-pass filtered at 2 mHz. The power spectra S_χ, measured at $\epsilon = 0.16$ and $\epsilon = 0.002$, are plotted in Fig. 2. They can be fitted by a Lorentzian for $\epsilon > 0$:

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

$S_0(\epsilon)$ represents the amplitude of fluctuations, and $f_c(\epsilon)$ is proportional to the inverse of the relaxation time $\tau(\epsilon)$ of θ_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 3 orders of magnitude. The values of S_0 and f_c are obviously dependent on ϵ and its sign. For $\epsilon < 0$, S_χ is the sum of two Lorentzian functions with two cutoff frequencies. Each frequency corresponds to a relaxation of the director of the LC in two different directions. The lowest frequency, which corresponds to θ, depends on ϵ contrary to the other frequency. The cutoff frequency f_c decreases with ϵ with a linear behavior as predicted by the Ginzburg-Landau model, i.e., $1/\tau = \epsilon/\tau_o$, where the value of τ_0 agrees with that obtained from the LC parameters. The amplitude S_0 has a complex dependence on ϵ. 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 χ. We consider the normalized order parameter: $y = \frac{\chi - \langle\chi\rangle}{\sigma}$, where σ^2 is the variance of χ. The probability density functions of y are plotted in Fig. 3 for three different values of ϵ. We find that, far from the critical value ($\epsilon = 0.16$), the distribution is Gaussian [Fig. 3(a)]. In contrast, for a value of ϵ closer to 0, typically $\epsilon \sim 2 \times 10^{-3}$, the PDF of fluctuations of χ are not Gaussian as is clear from Fig. 3(d). In Figs. 3(b) and 3(c), we plot the distribution of χ for two intermediate values 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 [4]:

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

We obtain $a = 2.95$ at $\epsilon \sim 2 \times 10^{-3}$, $a = 6.6$ at $\epsilon \sim 4 \times 10^{-3}$, and $a = 23.5$ at $\epsilon \sim 8 \times 10^{-3}$. By 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 ϵ very close to 0 is the main result of this Letter. One may wonder why the GG is observed in our experiment and not in other experiments on phase transitions. To answer this question, let us first consider the slow modes of χ whose relevance for the GG

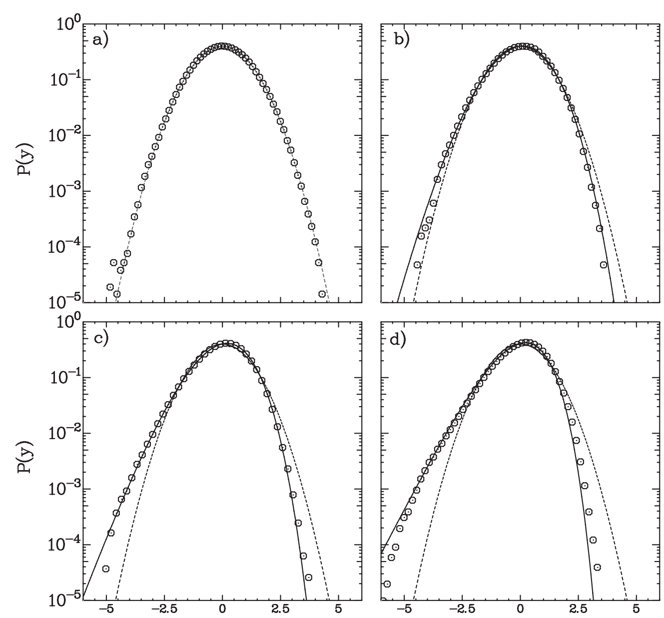

FIG. 3. (a)–(d) PDF of $y = \frac{\chi - \langle\chi\rangle}{\sigma}$ at $\epsilon \sim 0.16$, 8×10^{-3}, 4×10^{-3}, and 2×10^{-3}, respectively. The dashed line is a Gaussian fit. In (b)–(d), the continuous lines are the GG distributions with $a = 23.5$, 6.6, and 2.95, respectively.

distribution has been pointed out in Ref. [5]. To confirm this point, the time evolution of χ acquired at $\epsilon = 2 \times 10^{-3}$ is high-passed filtered at various cutoff frequencies $f_{\rm HP}$. The skewness γ of the filtered signal is plotted as a function of $f_{\rm HP}$ [Fig. 4(a)]. When $f_{\rm HP}$ is increased, we see that the skewness decreases (γ^{-1} is linear in $f_{\rm HP}$). A Gaussian behavior is retrieved for $f_{\rm HP} > 10 f_c \simeq 0.1$ 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 the Fréedericksz transition did not have a sufficient resolution at low frequencies, and they erased this effect. Let us now consider the correlation length ξ of θ_0 in the plane (x, y). This correlation length has to be compared with the diameter of the measuring volume, which, in our experiment, is determined by the laser beam diameter D inside the cell. At $\epsilon = 0.002$, we find $\xi = 47$ μm, 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 $N_{\rm eff} = D/\xi \propto \sqrt{\epsilon}$. In Fig. 4(b), we plot the values of the skewness as a function of ϵ. 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, $\gamma^{-1} = -\sqrt{a} = p + q\epsilon = p + \tilde{q} N_{\rm eff}^2$. 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 Refs. [4,18]. For the magnetization of the two-dimensional XY model, it has been found that $\gamma^{-1} \sim -\sqrt{a} \sim -\sqrt{\frac{2}{\pi}}[1 + \frac{1}{2}(\frac{N_{\rm eff}}{2\pi})^2]$. The dependence of $\sqrt{a}$ on $N_{\rm eff}$ is the same as in our experiment, but the coefficients depend on the system. As the ξ is proportional to the cell thickness, we have verified that, for cells having larger L, the GG is obtained for larger values of ϵ. This is indeed the case, and the detailed description of the results of the other cells will be the subject of a long article.

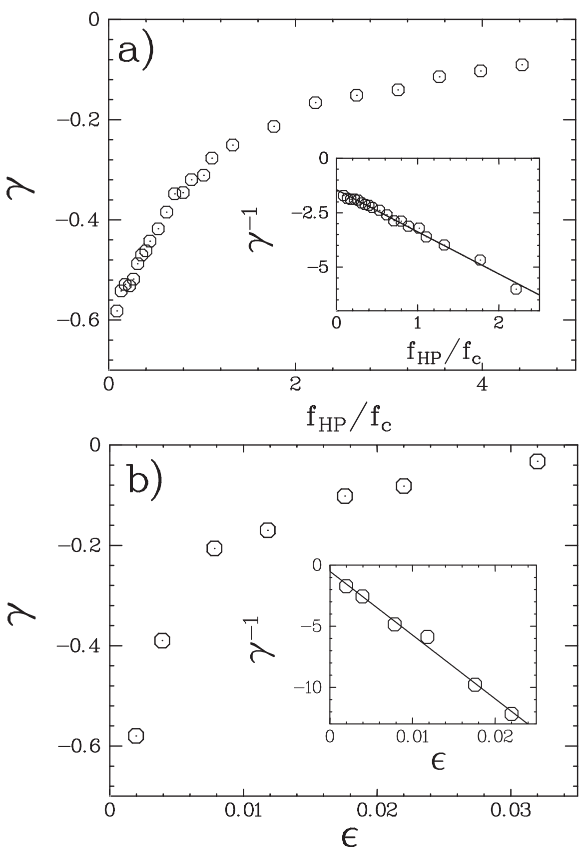

FIG. 4. (a) The skewness γ of the fluctuations at $\epsilon = 2 \times 10^{-3}$ is plotted as a function of $f_{\rm HP}$. Inset: γ^{-1} is linear in $f_{\rm HP}/f_c$. (b) γ as a function of ϵ. Inset: γ^{-1} is linear in ϵ.

In conclusion, we have experimentally shown, by 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. 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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