Light depolarization effects during the Fréedericksz transition in nematic liquid crystals

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Abstract: This work is aimed to the photopolarimetric characterization of the disorder evolution occurring in homeotropically aligned nematic liquid crystal films during the electrically induced Fréedericksz transition. The molecular director dynamics and the transversal reorientation modes are investigated by the analysis of the depolarization of the light beam emerging from the sample. Our measurements reveal unexpected depolarization effects at the transition, which we interpret in terms of director field unhomogeneity and defects creation.

OCIS codes: (120.5410) Polarimetry; (160.3710) Liquid Crystals; (290.5855) Scattering and Polarization.

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1. Introduction

One of the most intensively studied phenomena in the physics of liquid crystals is the Fréedericksz transition [1], which consists in the elastic distortion of a homeotropically or planarly aligned nematic liquid-crystal film under the action of an external either magnetic or electric field. Under this circumstances, two torques determine the behavior of a nematic liquid crystal (NLC): the elastic torque within the material tries to align the NLC director along the easy direction as determined by the boundary conditions; the torque caused by the external field tends to align the NLC molecules either parallel ($\Delta\varepsilon>0$) or orthogonal ($\Delta\varepsilon<0$) to the field. If only one of these torques occurs we can have a uniform NLC orientation, on the contrary in the situation we are considering there is a competition between them, therefore above a threshold value V_{TH} of the applied voltage V the uniformity may be partially destroyed, the initial alignment due to the anchoring disappears in the bulk and the Fréedericksz transition occurs [1,2].

The application of the bifurcation theory to a variety of problems in physics and applied mathematics has led to a more complete understanding of how complicated non-linear behaviors arise in these systems [3]. The Fréedericksz transition in a planarly aligned cell was considered as an example of symmetry breaking (pitchfork) bifurcation [4] where the symmetry being broken is a mirror-like (anti)symmetry about the mid-plane.

An other important subject in liquid crystal science is the formation of defects: defects consist in the local breaking of the symmetry of an ordered medium. Unlike dislocations, which break the translational symmetry, disclinations break the rotational symmetry and they are typical of media having continuous symmetries, as liquid crystals (for more details, see, e.g., [5]). We point out that a phase transition represents a global symmetry breaking, on the contrary in defects we have a local symmetry breaking. We recall that the Fréedericksz transition is a second order transition, i.e., it is continuous [1]. That is in first order transition if the system is in a stable state and the bifurcation parameter is varied, the state could become unstable giving rise to the nucleation of other stationary stable states, which are largely different from the initial one. In second order transition the stable state, that becomes unstable, leads to the appearance of stationary stable states that are similar to the unstable one (see [6] and its citations). Then it is difficult to hypothesize defects formation in this type of transition, though A. Buka and L. Kramer observed a transient pattern during the splay Fréedericksz transition in a planarly aligned nematic film [7]. This pattern originates at high voltages from the field distortion owing to the strong anisotropies (dielectric and/or conductive). Defect-antidefect patterns can be easily observed in homeotropic NLC samples after the Fréedericksz transition.

In this paper the ellipsometric study of the Fréedericksz transition in homeotropically aligned nematic liquid crystal samples is reported. During the transition we observed an unexpected light depolarization of the transmitted light beam, that can be understood by means simple concepts of phase transitions and defect theories.

2. Experimental

Upon applying an alternate electric voltage (1000 Hz) to a nematic liquid crystal film (M7 nematic mixture [8]) confined between two transparent electrodes (glass slides coated by an ITO layer), being 25 μm the cell thickness, each ITO electrode was coated by a surfactant (DMOAP) to induce a homeotropic alignment. The pretilt angle is zero. The M7 parameters values reported in the literature [8] are the following: the rotational viscosity is γ_1=41cP; the

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dielectric anisotropy is $\Delta\varepsilon$=-0.31 and the bend elastic costant is $K_{33} = 8.1 \cdot 10^{-7}$ dyne. Because M7 has a negative dielectric anisotropy ($\varepsilon_{\parallel} < \varepsilon_{\perp}$), the nematic molecular director tends to align itself perpendicular to the electric field and, above a certain threshold voltage, the Fréedericksz transition occurs. We characterized the transient regime by analyzing the polarization of the transmitted light by the same experimental set-up described in our previous article [9]. The light emitted by the laser (λ=632.6 nm) was focused on the cell (0.5mW/0.008mm^2). The polarization state of the light transmitted by the sample was determined by a Four Detector Photopolarimeter (FDP), which simultaneously measures the Stokes parameters [10] of the electromagnetic wave.

The FDP is described in reference [10] in particular the instrument calibration has been executed as reported in references [11-13].

The measurement of the Stokes parameters S_i not only provides information on any parameter characterizing the polarized light, for example the ellipticity and the azimuthal angle Θ of the major axis of the ellipse of polarization, but also the degree of polarization P of the light beam can be determined.

Let us recall some relations:

$$\Theta = \frac{1}{2}\arctan\left(\frac{S_2}{S_1}\right),$$
$$e = \tan\left[\frac{1}{2}\arcsin\left(\frac{S_3}{\left(S_1^2 + S_2^2 + S_3^2\right)^{1/2}}\right)\right], \qquad (1)$$
$$P = \sqrt{\sum_{k=1}^{3}\left(\frac{S_K}{S_0}\right)^2}.$$

P ranges from zero (completely unpolarized light) to unity (totally polarized light) and it assumes any intermediate values for partially polarized light.

In all the experiments reported here, the samples have been shined with left-handed circular polarized light and the temperature of the sample was kept at (25±1)°C.

3. Light Depolarization effects during the Fréedericksz transition

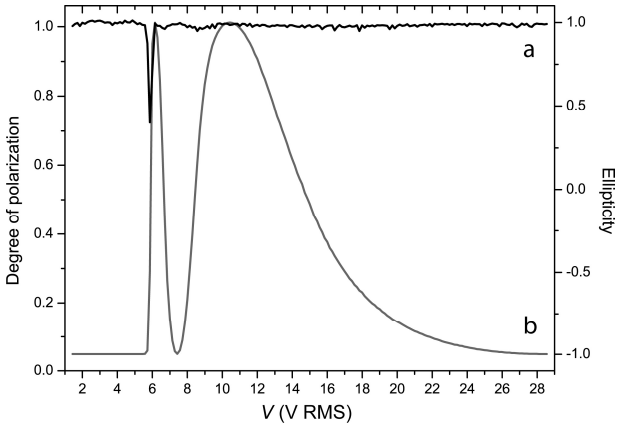

Fig. 1. Degree of polarization (curve a) and ellipticity (curve b) vs. the applied voltage V, the impinging light beam was left-handed circularly polarized.

In Fig. 1 we report both the ellipticity and the degree of polarization vs. the driving voltage. Below the threshold the system persists in its initial state, no depolarization occurs (P=1) and

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the transmitted light remains left-handed circularly polarized (e=-1). As soon as the applied voltage exceeds V_{TH} = 5.4V RMS the Fréedericksz transition occurs, in fact the reorientation of the NLC molecular director is revealed by ellipticity oscillations in the range [-1÷1], we stress that the degree of polarization is almost constant below and above the transition, but it shows an unexpected deep minimum at the transition.

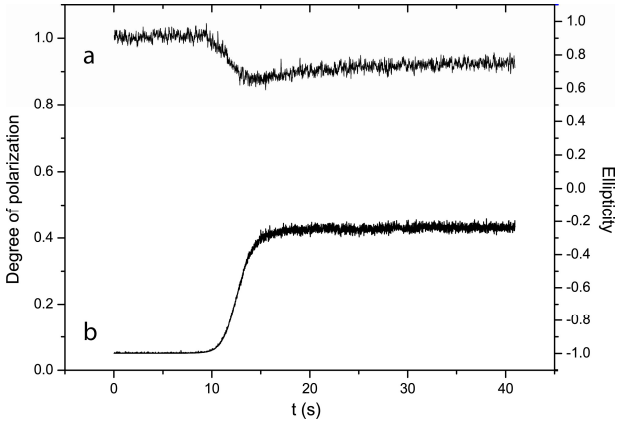

Fig. 2. Time behavior of both the degree of polarization (curve a) and the ellipticity (curve b), the applied voltage was kept fixed at 5.6 V RMS, the impinging light beam was left-handed circularly polarized.

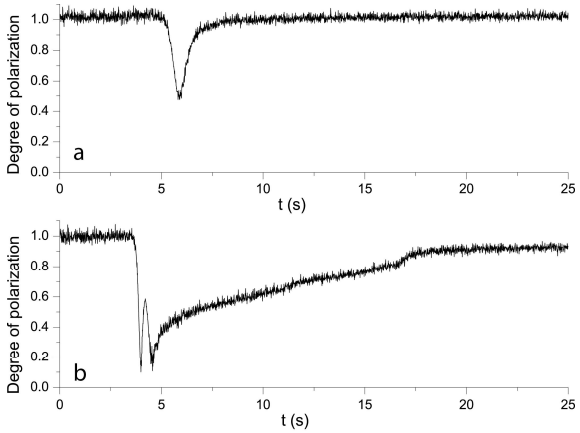

Fig. 3. Time behavior of the degree of polarization *P* for two different values of the applied voltage: 6 V RMS (a), 7 V RMS (b).

In Fig. 2 we report the time behavior of both the degree of polarization and the ellipticity meanwhile an alternate voltage (5.6V RMS) was applied to the sample. At the Fréedericksz transition, owing to the molecular director reorientation, the ellipticity increase from e=-1 to about e=-0.2, but also the degree of polarization decreases and then it increases. In Fig. 3 we report the degree of polarization time behavior for two other value of the applied voltage (6V RMS and 7V RMS), nevertheless the *P* behavior appears quantitatively different, *P* always tends to return to its initial value.

In order to make a comparison between homeotropic and planar configurations we performed the same observations on planar sandwich type cells filled by BL001 by Merck, in fact a positive dielectric anisotropy NLC must be used to observe the Fréedericksz transition in planar cells.

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In Fig. 4 we report the time behavior of the degree of polarization (a) and ellipticity (b) when the applied voltage is switched between 0 and 2.56V RMS (in this case V_{TH} ~1V RMS). As soon as the applied voltage *V* is turned on the Fréedericksz transition occurs, in fact the molecular director reorientation processes are clearly revealed by the ellipticity variations, which can be observed also when *V* is turned off and the system returns to the initial planar orientation .

We want to emphasize that for planarly oriented NLC cells we did not observe any light depolarization effect, either during the Fréedericksz transition or during the transient towards the planar state.

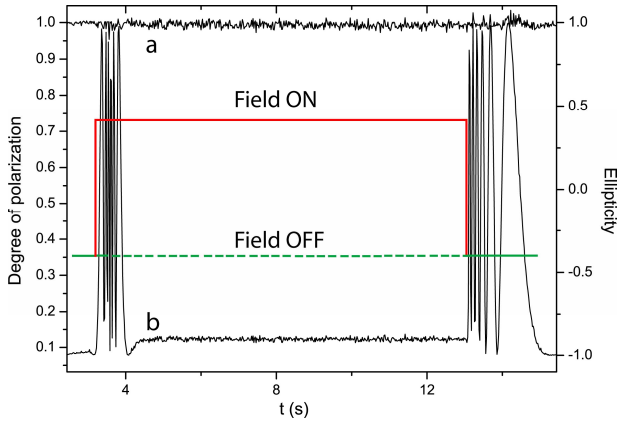

Fig. 4. Time behavior of the degree of polarization (curve a) and ellipticity (curve b) for a planar BL001 cell. The applied voltage was switched between 0 and 2.56V RMS. The oscillations of the ellipticity denote the reorientation dynamics at the OFF→ON and the ON→OFF transients respectively, no depolarization effects were observed.

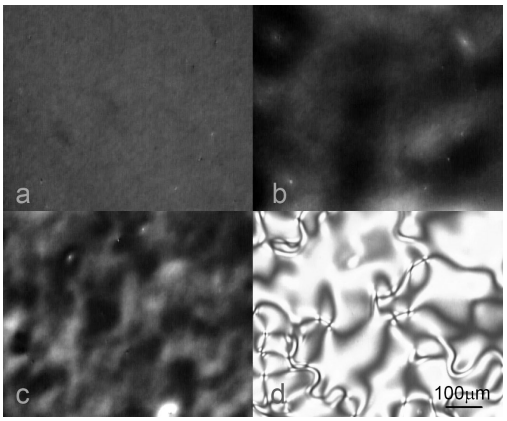

Fig. 5. Observations by orthoscopic optical microscopy of the Fréedericksz transition in a homeotropic cell at different applied voltage: a) in absence of field; b) 5.4 V RMS c) 9 V RMS just after the Fréedericksz transition d) 9 V RMS when the defect-antidefect pattern is formed.

These observations point out a substantial difference between the homeotropic and the planar cases: during the Fréedericksz transition the molecular director of a planarly aligned sample undergoes a reorientation between two fixed direction, it reorients “in plane” from planar to normal. This is a transition where the initial and the final directions of the director are established by the geometry of the system. In the homeotropic case only the initial director orientation is fixed by the boundaries, during the transition the director can bend in any

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directions around the normal to the cell surfaces. Actually, the azimuthal angle φ is a further degree of freedom that we don't have for planar orientation, this degeneracy is an example of breaking (pitchfork) bifurcation [4].

Within the illuminated volume the molecular director can reorient locally with any different φ bringing to different director orientation domains, we can suppose that the external electric voltage amplifies same transversal orientational mode, which wave vector depends on the intensity of the external field, in particular the more the electric field is intense the greater are the amplified wave vectors (see Fig. 5). Interacting with the anisotropic sample the electric field components of the incident light wave undergo to casual local phase displacements, therefore it becomes partially unpolarized. In our case the light depolarization mechanism differs from that treated in references [14,15] where depolarization occurs through scattering processes, further investigations about this aspect are in progress. Summarizing, depolarization reflects the mesoscopic spatial disorder of the director field during the early stages of the Freédericksz transition, subsequently the viscoelastic relaxation of director distortion towards a uniform orientation causes *P* to increase again.

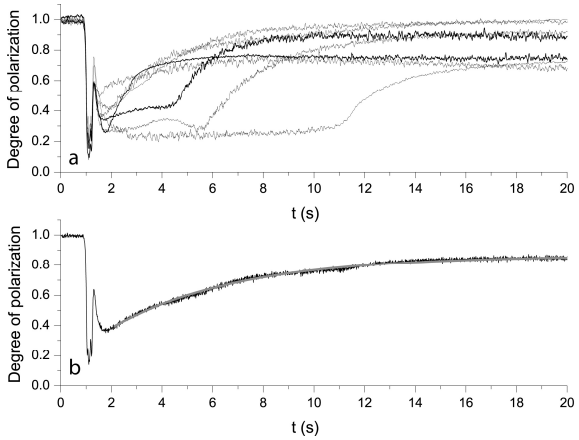

Fig. 6. a) Degree of polarization measured by the repetition of the same observation (12 V RMS). b) Average of the measures showed in a) together with the exponential fit of its long time behavior $P = P_0 + Ae^{-\frac{t}{T}}$. Fitted parameters: P_0=0.87, A=-0.47, T=5.0s.

Anyway our description does not account the presence of two minima in Fig. 6, thus to get more insight into the phenomenon we have repeated the polarimetric measurements observing simultaneously the sample by an optical polarizing microscope. In Fig. 6a the results of several experimental runs at 12 VRMS is reported: all the runs exhibit a similar behavior with two equally spaced minima during the early stages of the transition, then the slow relaxation process can follow different paths. The average of these runs (Fig. 6b) exhibit a most regular asymptotic behavior, which we have fitted by an exponential decay to determine the relaxation time T = 5 sec. Collating the measurement of *P* and the microscope observations we can infer the following statements:

- As soon as the initial homeotropic orientation is perturbed in the bulk, a change of the transmitted light ellipticity can be observed, but the polarization degree remains unchanged (P=1), in fact the sample appears homogeneous and brighter at the optical microscope.
- When mesoscopic domains with different director orientation appear in the sample, *P* promptly decreases reaching its first relative minimum (P_{MIN}), after which both the enlargement of the orientation domains and the homogenization causes *P* to suddenly increases again. This transient is always accompanied by the creation of a defect-antidefect

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pattern, as soon as defects are completely formed (see Fig. 5d) *P* reaches the second minimum.

- Finally, the relaxation of the director field and the defects annihilation leads *P* in average to increase again.

In conclusion the observed light depolarization effects have two origins: the formation of domains of different director orientation which acts as a Cornu pseudodepolarizer [16] and the presence of defects. The number of defects can vary within the shined region of the sample giving rise to different P behaviors in Fig. 6a. The behavior showed in Fig. 6b corresponds to the situation in which defect-antidefect pattern is homogeneously distributed in the sample and then it regularly disappears.

We can estimate the typical size of the above mentioned orientational domains by measuring the characteristic time of the relaxation processes. We suppose that the domains director relaxation time τ is twice the time Δt elapsed between the application of the external field and occurrence of the first *P* minimum.

In Fig. 7a we report Δt as functions of the reduced voltage $\varepsilon = V^2/V_{TH}^2 - 1$, being V the applied voltage and V_{TH} the threshold voltage. We have performed a statistical analysis of Δt at different voltages, the mean value and the standard deviation are reported. We have fitted this curve with an exponential decay disregarding the first experimental points because in this case V is really near V_{TH} and the formation of defect could occur before the formation of domains. In according with [17] and its citations we suppose that the domains director relaxation time is:

$$\tau \simeq 2\Delta t \simeq \gamma_1 \mathrm{D}^2 / K_{33}\pi^2, \tag{2}$$

where D is the domain size, γ_1 denotes the rotational viscosity and K_{33} is the bend elastic modulus, we note that the applied voltage dependence is introduced via D, i.e larger the voltage smaller the domain size D. Then, introducing the M7 material constants [8], we can approximately estimate D for different values of τ, i.e. ε, therefore in Fig. 7b we show D as function of ε. As we can see the domains size ranges from 5 μm to 1 μm and it shrinks at largest ε.

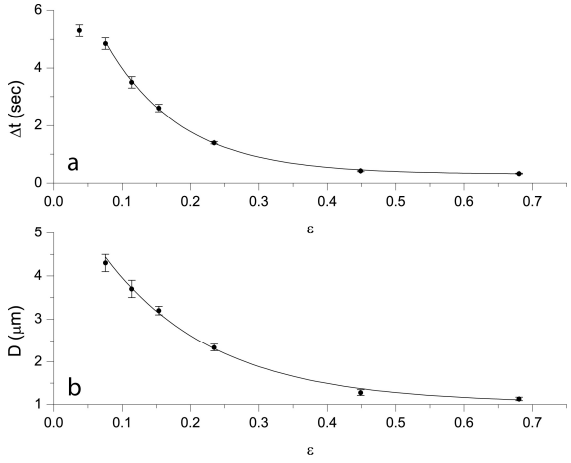

Fig. 7. a) Δt (defined in the text) vs $\varepsilon = V^2/V_{TH}^2 - 1$ and the exponential fit (solid line) $\Delta t = \Delta t_0 + Ae^{-\frac{\varepsilon}{\varepsilon_1}}$, fitted parameters: Δt_0=0.30s, A=9.2s, ε_1=0.11. b) The same for the domains size $\mathrm{D}=\mathrm{D}_\infty + Be^{-\frac{\varepsilon}{\varepsilon_2}}$, fitted parameters: D_∞=1.03μm, B=5.4μm, ε_2=0.16.

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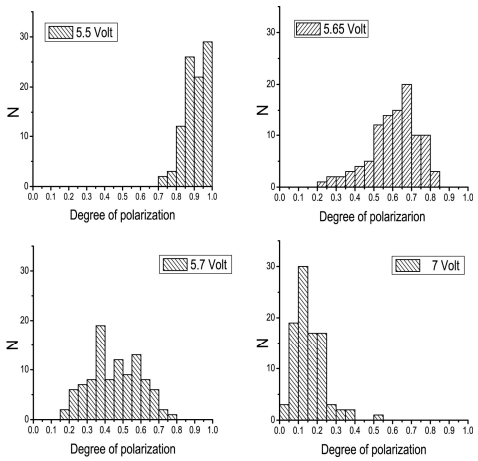

Fig. 8. Histograms of P_{MIN} at different voltage.

In Fig. 8 we reports the experimental statistical distributions of P_{MIN} at different applied voltage values, we have estimated their first and second moments fitting them by Poisson distributions. In Fig. 9 we report the voltage behavior of the mean value of P_{MIN}. The plot in Fig. 9 shows an exponential decay to the value $P_\infty = 0.16 \pm 0.03$. Since P_{MIN} is always not zero, we suppose that there are not statistically independent mesoscopic regions, i.e. we are dealing with an inertial system [18,19].

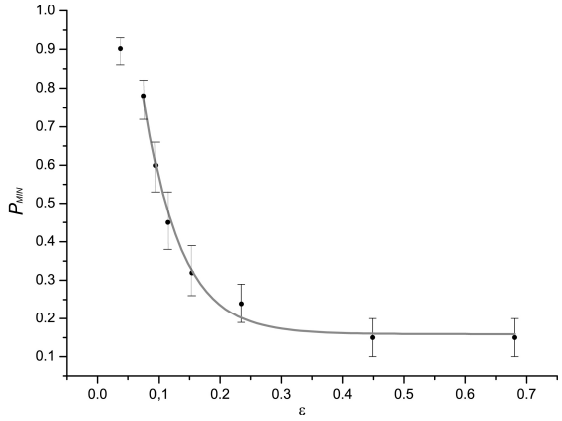

Fig. 9. Mean values of P_{MIN} as function of the reduced voltage ε, together with the exponential fit (solid line) $P_{\min} = P_\infty + P_0 e^{-\frac{\varepsilon}{\varepsilon_3}}$, fitted parameters: P_∞=0.16, P_0=2.2, ε_3=0.06.

Finally, some speculations are needed in order to discuss the light depolarization in presence of defects. Firstly, we observe that light depolarization can be observed even if no defects are inside the illuminated volume. In fact, optical microscope observations confirm that only few defects appear in the sample just above V_{TH} (see Fig. 2), so that the average distance among them is larger then the size of the illuminated region (about 100 μm). Therefore we can expect on average not more than one defect inside the shined regions of the sample. On the other hand the more the defect number increases the more the light is depolarized. So we could suppose that the presence of some defects could act as the boundary

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of a cavity selecting a few orientation modes which propagates through a visco-elastic coupling. This situation could generate weak-defects (with no energetic dense core and zero winding number), which increase both scattering and depolarization of light [20]. In addition the larger is *V* the more the defect density and the light depolarization increase. Actually the presence of far defect-antidefect pairs (that can survive for minutes) slows down the relaxation process which would allows *P* to return to 1.

4. Conclusions

In this work we have reported the characterization of disorder evolution that occurs during the Fréedericksz transition in homeotropically aligned NLC cells. The study was carried out by measuring the Stokes parameters of the light transmitted by the sample during the transient. We focused our attention on the time behavior of the degree of polarization which reveals interesting characteristics of the disorder due to domains of different director orientation on a mesoscopic scale.

We find that there is a substantial difference between the homeotropic and the planar cases of initial configuration of the nematic film. The depolarization effects during the Fréedericksz transition occur only in the homeotropic case. In the planar case the orientational director dynamics is established by the geometry of the system, which unambiguously fixes the initial and the final directions of the director. In the homeotropic case only the initial director orientation is fixed (perpendicular to the cell plates), than, during the transition, the director is free to reorient in all the directions around the initial one. This is a further degree of freedom which is not present in the planar case. This symmetry breaking produces a local director orientation which is different in the various points of the cell. As a consequence the wave front of the transmitted light undergoes a local phase displacement and we have light depolarization. The average size of this domains is about 5 μm for ε=0.07 and 1 μm for ε=0.7.

Moreover there is evidence of other depolarization effects in presence of the defects. We observe that the depolarization does not occur when the defects disappear for the defect-antidefect annihilation mechanism. So we could suppose that the presence of defects could select some modes of the orientational perturbation. Therefore we have a director orientation pattern that continuously varies and weak-defects might appear.

These characteristics could appertain to the majority of the phase transitions in which a similar symmetry breaking is involved, depending on the correlation length of the phase a disorder transient in the studied field can be produced and, consequently, the distribution of the degrees of freedom of the energy (elastic in the case of nematic) can be not uniform and singular. Moreover the formation of defects could cause amplification of some modes in the system, perturbation waves, that persist as long as defects persist.

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