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# Spirals in cholesteric liquid crystals: A possible nucleation initiator

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## Abstract

The helicoidal structure of the cholesteric liquid crystal phase can be untwisted by the electric field effect. We have shown that spiral-shaped structures could nucleate at the threshold of the field-induced homeotropic state from filaments (linked to the twisted state) embedded in a homeotropic matrix (untwisted cholesteric). In this article, we describe and study what are the mechanisms involved in this phenomenon. In particular, we study to what extent the characteristics suggest a nucleation mechanism based on the propagation from the surfaces of distortions in the distribution of molecules director. These deformations propagate through the homeotropic matrix adjacent to the filaments.

## 1. Introduction

The unwinding of the helicoidal structure of a cholesteric liquid crystal by an electric field can be observed in the case of a positive dielectric anisotropy value and a field direction perpendicular to the helix axis [1]. This cholesteric to homeotropic nematic transition has formed the subject of numerous studies in the seventies, notably on account of potential applications in display devices [2].

Recently, we have shown that field-induced rotating spirals could germinate from a configuration of filaments, linked to the cholesteric phase, embedded in a homeotropic matrix [3-5]. In this article, we study to what extent the characteristics of the phenomenon suggest a nucleation mechanism based on the propagation of surface deformations via the homeotropic matrix adjacent to

the filaments. Our initial postulate is as follows: the chiral doping of nematic liquid crystal (5CB) does not annihilate mechanisms present in the pure material but rather transposes some of them in electro-optic effects intrinsic to the chiral state. The observations are made at the threshold of the homeotropic state (i.e. molecules parallel to the electric field direction). Therefore, the system is marked by the coexistence of two states from which we must take intrinsic characteristics into account: a homeotropic state (untwisted cholesteric state) for the majority of molecules and a chirality subsisting under the shape of filaments. From this configuration spiral patterns are nucleating and growing. The characteristics suggest a parallel with electro-optic effects already present in the series of nematic cyanobiphenyls, especially 5CB, one compound of the binary mixture (for 91% in weight).

The article is organized as follows. In Section 2, we give the experimental conditions. In Section 3, we briefly recall the nucleation and growth characteristics of spiral patterns in cholesterics when the helicoidal structure is unwound by an AC electric

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field. In Section 4, and preliminarily to the discussion, we introduce the electro-optic effects present in nematic cyanobiphenyls. We then discuss, in Section 5, what are the different mechanisms involved in the phenomenon. In particular, we emphasize the analogies which exist between the flexo-electric character of electro-optic effects present in nematics and the behaviour of field-induced spirals in cholesterics.

## 2. Experimental

The study is made in the case of a mixture of a liquid crystal side-chain copolymer (cyclic polysiloxane with two types of mesogens: cholesteric and nematic) and the 5CB nematic liquid crystal (also called K15) [3]. The polymer is cholesteric between 50 and 200 °C. The handedness of the helicity is left. The pitch is equal to 290 nm (measured near the glassy transition) and the reflection wavelength (at normal incidence) corresponds to the blue of the visible spectrum (450 nm).

The copolymer concentration is equal to 9% (in weight). This concentration corresponds to a large pitch cholesteric ( $\sim 3 \mu\text{m}$ ) at ambient temperature—temperature of the study—and up to 42 °C. The texture is of fingerprint-type. The sample is contained between two glass plates covered with a transparent conducting film of ITO (Indium Tin Oxide) for applying an electric field. The cell gap is about 3.5  $\mu\text{m}$ . Thus, the thickness to natural pitch ratio (confinement ratio) is near to 1; the cell geometry is a confined geometry. The dielectric anisotropy value of the mixture is +7. The electric field direction is perpendicular to the helicoidal axis which is, therefore, parallel to the cell walls. These conditions lead to an unwinding of the cholesteric structure by the electric field effect due to the dielectric coupling.

## 3. Field-induced spirals in confined cholesterics near the untwisted state

The cholesteric liquid crystal phase presents a helicoidal structure. The helix unwinding by an electric field can be notably observed in the case of a cholesteric liquid crystal with a positive dielectric

anisotropy and the field direction perpendicular to the helicoidal axis [1]. Rotating spiral patterns induced by an AC electric field can germinate in a texture exhibiting filaments, linked to the twisted state, embedded in a homeotropic matrix, in which molecules are aligned according to the field direction, identical to the optic axis. Fig. 1(left) describes the sequence of a spiral growth for different times at fixed frequency. Spirals can be left- or right-handed. The phenomenon has a critical character: at fixed frequency  $f$ , spiral patterns arise for a voltage  $V$  greater than some critical value.  $V$  is typically between 75% of  $V_{\text{th}}$ , which is the threshold voltage corresponding to the complete untwisting of the cholesteric helix and  $V_{\text{th}}$  [3]. The nucleation always begins from a fixed point which is a filament extremity. The frequency value plays a leading part for the nucleation time: if spirals nucleate in less than one minute for frequencies less than 50 Hz, longer times—more than one hour—are required as soon as the frequency exceeds 1 kHz [4]. On growth, the transverse speed of a whorl quickly decreases when frequency increases: typically from 6 to 0.3  $\mu\text{m/s}$  when the frequency goes from 10 to 100 Hz [4, 5]. Spirals are the last structures to resist to the homeotropic state propagating when the voltage value reaches the threshold value  $V_{\text{th}}$ . For a sufficiently long time (frequency-dependent), the texture is formed by interconnected spirals (Fig. 1(right)). The confinement state is important: for confinement ratios greater than one, the spiral nucleation is perturbed by convective instabilities which parasite and conceal the observation [4, 5]. The density of spiral is less important when the cholesteric structure is not constrained, the germination being more liable to miscarry. The phenomenon persists when charge injection by the electrodes is avoided (SiO layers are deposited on the conductive glass plates). A treatment of cell walls for inducing a strong homeotropic orientation modifies the dynamic behaviour and one for inducing a planar anchoring annihilates the nucleation [5].

## 4. Electro-optic effects in nematic cyanobiphenyls

Nematic liquid crystals can be homogeneously oriented, by treating the two plate boundaries, in

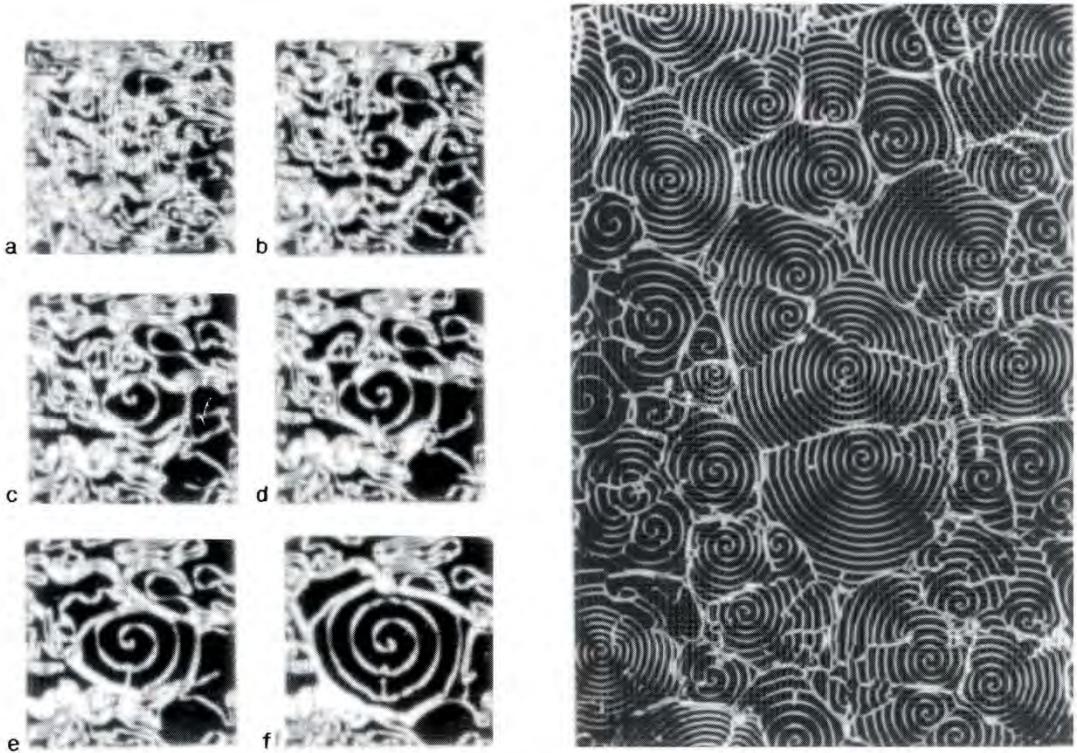


Fig. 1. (left) Nucleation and growth of a spiral pattern from a confined cholesteric liquid crystal submitted to an AC electric field.  $V = 4.2$  V,  $f = 50$  Hz. The observations are made by polarized-light microscopy between crossed polarizers. The field direction is perpendicular to the plane (glass plates). Photographs are taken for different times: (a)  $t = 0$  s, the texture exhibits disordered filaments, linked to the cholesteric phase, embedded in a dark matrix (molecules are aligned according to the field direction and, thus, the optic axis); (b)  $t = 3$  s, nucleation begins from an extremity of a filament which is a fixed point; (c)  $t = 6$  s, the filament incurves itself and forms a whorl; (d)  $t = 9$  s, growth progresses; (e)  $t = 12$  s; (f)  $t = 18$  s. (right) Spiral texture: for a sufficiently long time (frequency dependent), the space is totally recovered by spiral patterns embedded in a homeotropic matrix.

order to exhibit a single-crystal texture. By its anisotropic action, an external electric (or magnetic) field can induce distortions in the mean direction of molecules. Consequently, it can induce texture instabilities.

The effects associated to instabilities in nematics have given rise to numerous studies, especially in  $n$ CB cyanobiphenyls as 5CB. The interest of such a molecule consists in the important response to an electric field and the experimental geometry of the cell. Indeed: (i) it offers great values for surface polarization  $P_s$ ; (ii) the polarization-induced tilt of the director  $\mathbf{n}$  can be easily detected by optical

methods; (iii) with a positive anisotropy of the dielectric permittivity  $\Delta\epsilon$  as of the electrical conductivity  $\Delta\sigma$  and with a vertical external electric field ( $E \parallel \mathbf{n}$ ), Freedericksz or Carr-Helfrich effects [1] are eliminated, i.e. it avoids a reorientation of  $\mathbf{n}$  because of the dielectric or anisotropic electro-hydrodynamic torques; (iv) in the initial state (homeotropic, i.e. molecules perpendicular to the substrates) the cell, observed under a polarizing microscope, is totally dark (since the optic axis of the nematic liquid crystal coincides with the microscope axis) which makes it possible to detect the changes in orientation of  $\mathbf{n}$ ; (v) the amphiphatic

nature of 5CB, with its capacity to adopt different orientations on hydrophilic or hydrophobic substrates is, for example, useful for determining the part of surface polarization with respect to the flexo-electricity.

The external manifestations of the instabilities present in 5CB are cloud patterns, birefringent spots, rounded or striped domains. However, these instabilities, very similar in their pattern, have been interpreted by invoking fundamentally different mechanisms: isotropic electro-hydrodynamical instabilities (Felici mechanism) [6, 7], the flexo-electric effect and/or a surface polarization [8-11]. Ordo-electricity is also theoretically predicted [12]. In the next paragraph we will recall what these mechanisms consists of.

Let us consider the polar surface instabilities due to the coupling of the flexo-electric effect and possibly a surface polarization with the electric field at ambient temperature in 5CB and 7CB [8]. These instabilities can be clearly distinguished from instabilities resulting from a charge injection by the electrodes. Cloud patterns, appearing above a threshold voltage  $V_c$ , seem to decorate more or less surface irregularities. The threshold for the cloudy patterns appearance increases proportionally to the thickness. The frequency dependence of  $V_c$  is linear until a critical frequency  $f_N$ , and as  $(f/f_N)^{1/2}$  for frequencies above  $f_N$ ,  $f_N$  being quantitatively given by the nematic curvature elasticity relaxation:  $f_N = 1/2\pi\{K\pi^2/(2d)^2\eta\}$ , where  $K$  is the bend elastic constant,  $d$  the cell thickness and  $\eta$  the viscosity. Cloud patterns disappear at strong fields and high frequencies, which is explained by the decrease of the coherence length  $\xi$  [13] varying as  $E^{-1}$ , and  $E$  itself varies as  $f^{1/2}$ . Indeed, we have:

$$\xi_{AC} = \sqrt{(4\pi K/\Delta\epsilon)} E_{AC}^{-1},$$

where  $\Delta\epsilon$  is the dielectric anisotropy, and likewise for a DC field [11]. Consequently,  $\xi_{AC}$  is proportional to  $f^{-1/2}$ .

Besides, it is mentioned in the case of 5CB that, for larger thickness, instabilities superimposed to the cloud patterns appear, making their observation delicate; these instabilities are described like circular domains corresponding to a vortex flow

induced by a localized charge injection.

We will have recourse to these data in the discussion which follows.

## 5. Discussion

### 5.1. Possible mechanisms

There exist at least five different mechanisms responsible of electro-optic effects resulting of the liquid crystal structure deformation: dielectric, electro-hydrodynamic, flexo-electric, the surface polarization and ordo-electric. However, the following nuance must be brought up: the respective parts of each mechanism will be different according as the cholesteric structure is in a confined geometry or not. For example, if we consider a cell whose thickness is inferior to the pitch value, the frustration has partially achieved the part of the dielectric effect which would occur when the cell is submitted to an electric field. Indeed, the helicoidal structure is hindered to freely develop in the volume. For a critical thickness value, the twist is even completely suppressed: the structure is nematic and results from a geometric frustration of the cholesteric phase. Therefore, the cholesteric to nematic transition can be carried out by varying the cell thickness; another possibility is to apply an electric (or magnetic) field, since the coupling with the dielectric (or diamagnetic) anisotropy of molecules involves the untwisting of the helix.

In the same way, the interactions between deformations issue from the two electrodes will not be the same in a thick or a thin film.

Let us recall and comment the above quoted mechanisms.

(a) *The dielectric effect* is a direct consequence of the coupling of the electric field to the dielectric anisotropy of the liquid crystal  $\Delta\epsilon = \epsilon_{||} - \epsilon_{\perp}$ , where  $\epsilon_{||}$  and  $\epsilon_{\perp}$  represent the dielectric constants measured along a parallel and a normal direction to the nematic axis. The type of field-induced transition depends evidently on the sign and the amplitude of  $\Delta\epsilon$ . For the cholesteric phase that we consider, the condition  $\Delta\epsilon = +7$  and the cell geometry (field perpendicular to the helicoidal axis) do not promote the instabilities of dielectric-type. Large positive

values of  $\Delta\epsilon$ , due to the important amount of the cyano compound, avoid a dielectric bulk instability.

(b) *Electro-hydrodynamical instabilities* are a topic which has been covered in an important number of articles (see, for review, Refs. [1, 14]). Three types of electro-hydrodynamical instabilities resulting in domain patterns in liquid crystals can be distinguished. (i) A space charge formation can arise from the electrolytic separation of positive and negative charges by an external field itself. (ii) The charge injection from electrodes [15] can be another reason responsible for the space charge formation. (iii) An only ionic mechanism of space charge formation can occur. This Carr-Helfrich mechanism [16] is due to the conductivity anisotropy:  $\Delta\sigma = \sigma_{\parallel} - \sigma_{\perp} \neq 0$ .

We do not promote the hypothesis of isotropic electro-hydrodynamic instabilities for several reasons. Firstly, the observations are made for voltage values (less than 5 V) well below the value for which there is a clear convection motion of the fluid (from 8 V), detected by the displacement of foreign particles (silica spheres incorporated in the mixture). Moreover, the phenomenon occurs until frequencies (a few hundred hertz) and field amplitudes (typically 1 V/ $\mu\text{m}$ ) for which the electro-hydrodynamic instabilities are not favoured. Then, we should expect particular electro-optic phenomena when the sample is subjected to an electric field in its isotropic phase, which is not the case. Besides, the effect is not correlated with the charge carrier injection from electrodes, which can be blocked by SiO layers [5].

On the other hand, we can expect that the existence of domains due to flexo-electricity and (or) surface polarization are not accompanied by a fluid motion.

(c) *The flexo-electric effect.* It is well-known that nematic liquid crystals are non-ferro-electrics, i.e. present the inversion symmetry [2]. However, it is possible to break this property by imposing a splay or bend curvature distortion to the mean molecular orientation. From this mechanism results an electric polarization  $\mathbf{P}_f$  (per unit volume):  $\mathbf{P}_f = e_1 \mathbf{n} \operatorname{div} \mathbf{n} + e_3 \operatorname{rot} \mathbf{n} \wedge \mathbf{n}$ , where  $\mathbf{n}$  is the unitary director,  $e_1$  and  $e_3$  are the flexo-electric coefficients (splay and bend deformations).

Initially, Meyer had imagined that  $\mathbf{P}_f$  was related to the presence of molecular dipoles along (or

across) pear-like (or banana-shaped) nematic molecules [17]. Later on, Prost and Marcerou have shown that flexo-electricity is mostly of quadrupolar origin (due to the coupling of quadrupolar densities with director distortions) [18].

We will discuss in detail this effect in the next paragraph, since it appears to be relevant.

(d) *A surface polarization* can be superimposed to the flexo-electricity and be caused by asymmetric molecule orientations on the boundaries. It results from the breakdown of the degeneracy condition  $\mathbf{n} = -\mathbf{n}$  applicable to the director at an interface due to the polar nature of the interaction between the molecules and the ambient medium.

(e) *The ordo-electricity.* Even in the case of a uniform orientation, the surface order parameter can be different from the volumic one. Thus, there is creation of an order gradient on a distance of the order of the coherence length. The associated spatial variation of quadrupolar density induces an electric ordo-polarization [12, 19], very similar to the flexo-polarization. The most important difference between flexo- and ordo-electricity lies in the scale of the effects: the director  $\mathbf{n}$  varies on a macroscopic scale (determined by the typical dimension of the cell, of a droplet, ...) when the scalar order parameter  $S$  is approximately constant in volume but quickly varies near a surface or defects (disclination lines, for example). Generally,  $S$  varies on a correlation length with an amplitude at least two times smaller than the typical length associated to the director variations [20].

Let us discuss in more details the last three mechanisms.

## 5.2. The flexo-electric mechanism

The effects associated to the flexo-electricity in nematics have given rise to numerous studies, especially for the electro-optic effects present in *n*CB cyanobiphenyls nematic liquid crystals like 5CB. As mentioned in Section 4, the interest of such a molecule consists in the important response to an electric field and the experimental geometry of the cell.

Let us compare the characteristics of spiral nucleation and growth to the flexo-electric behaviour of nematic cyanobiphenyls.

(i) *Frequency and field amplitude dependence.* Two essential features for the spiral nucleation are: the threshold character in voltage and the frequency dependence. These characteristics, related to a system where the majority of molecules are aligned according to a mean direction (as a consequence of the dielectric effect), are found in the electric field effects of the pure nematic of the mixture. On this point, we again find the threshold character, typical of the spiral nucleation: between about 75% of  $V_{th}$  and  $V_{th}$ , this fringe exhibiting a linear variation with respect to the threshold voltage varied by changing the cell thickness [4]. Similarly, occurrence of the threshold for the cloudy patterns increases proportionally with the thickness.

Another similar behaviour concerns the frequency dependence of  $V_c$ , which is the critical voltage for the appearance of birefringent patterns in 5CB: linear until  $f_N$  and like  $(f/f_N)^{1/2}$  above (see Section 4). In the same way, spirals can nucleate in a zone of the state diagram  $\{V, f\}$  where the variation of the threshold field is constant with respect to the frequency [4]. Above a cut-off frequency  $f_c$ , the phenomenon is extinguished and  $V_{th}$  also varies like  $f^{1/2}$ ; in fact, the nucleation time of dynamic structures seriously starts to increase before the frequency is equal to the critical frequency corresponding to the  $f^{1/2}$ -like variation of  $V_{th}$ : cut-off frequency for the phenomenon and critical frequency for the threshold voltage do not coincide.

Then, the decrease of the coherence length  $\xi$  when frequency (or field amplitude) increases is in agreement with: (a) a nucleation time for spirals which drastically increases; (b) a growth speed which quickly decreases; (c) a stability (with respect to voltage or temperature parameter [4]) more important at high frequency (and, thus, weak values of  $\xi$ ) than at low frequency. These points are as consequences of the fact that the dynamic structures have to nucleate and, then, to grow in a volume (let us say a "parallelepiped") whose one dimension (the height, of the order of  $\xi$ ; the two other dimensions defining the electrode plane) is progressively reduced when the frequency is increased. This variation is schematically represented on Fig. 2. Simultaneously, this dimension being decreased, the perturbations due to surface deformations have a less important volumic extension.

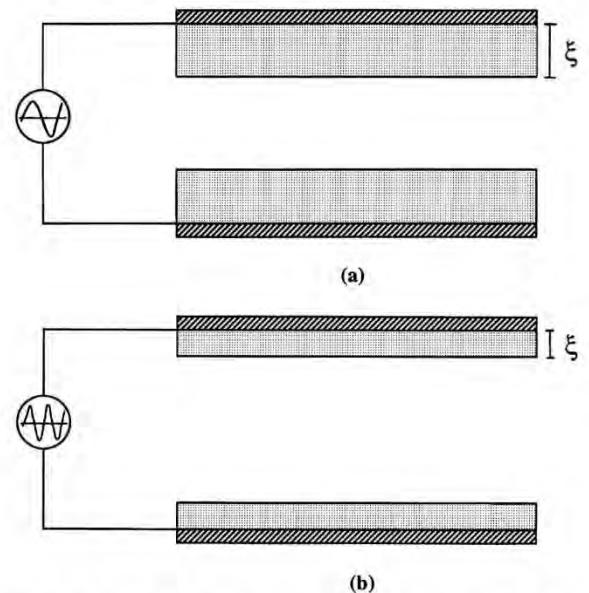


Fig. 2. A sketch of the coherence length  $\xi$  variation in a capacitor cell for: (a) a low- and (b) a high-frequency field. The shaded areas correspond to zones where surface distortions spread out in the material which is sandwiched between the two electrodes.

They can, consequently, permit to the structure to have a better stability with respect to the propagation of the disturbing homeotropic state due to an increase of the field amplitude.

On the threshold field value and the part of the frequency, it has been shown in the cholesteric case that the effective dielectric anisotropy value is reduced by the flexo-electric effect.

The expression for the critical field is

$$E_c = \pi t_0 \left[ \frac{\pi K_2}{\Delta \epsilon - \pi^3 e^2 K_1^{-1}} \right]^{1/2},$$

where  $t_0$  is the equilibrium twist,  $K_i$  the elastic constants,  $\Delta \epsilon$  the dielectric anisotropy and  $e$  the mean flexo-electric coefficient [21]. The flexo-electric coupling tends to keep the director in a helicoidal structure and increases the value of the critical field  $E_c$  for which the helix is totally untwisted, with respect to the value corresponding to a zero mean flexo-electric coefficient  $e$ . The following characteristics, proper to our observations, are in

agreement with these consequences: (a) when the voltage increase is stopped at a value for which the texture at the optical microscope consists in some filaments, linked to the cholesteric phase, in a homeotropic matrix, a dynamic process progressively gives birth to a “pavement” of the observation zone by spirals. That seems to express a struggle of the system against the helicity loss provoked by the dielectric effect. Besides, a weak increase of voltage destroys filaments disordered in their shape (non-still-spiralled) before spirals; there exists different limits in stability. (b) If the configuration of spirals in a homeotropic matrix has not been stated during the transition, the homeotropic state is reached at a definite value  $E_c$  of the field; if the field is increased from the spiral configuration, in order to get the homeotropic state, it will be reached at a superior value  $E_c + \delta E_c$  (where  $\delta E_c$  is a weak increase of field amplitude).

The frequency and field amplitude dependences are in agreement with the consequences of the flexo-electric coupling.

(ii) *About the confinement and the anchoring influence.* It is mentioned in the case of 5CB that, for larger thicknesses, instabilities superimposed to the cloud patterns appear [8]. Without dealing about the nature of these instabilities for each system, this characteristic is identical to what happens in our system out of the confined geometry. The confinement state remains important for two main reasons: (a) for keeping off the convective instabilities typical of thick samples and parasiting the spirals; (b) and – strictly with regard to the flexo-electric hypothesis – for promoting an inhomogeneous distribution in the orientation of molecules, which is a favourable factor to the flexo-electricity [22]. Likewise, the inhomogeneity of the electric field is also a favourable factor [23], which is the case with the cell, on account of the solution conductivity. On the other hand, the propagation speed, at fixed frequency, is larger in the case of a weak homeotropic anchoring than for a strong one [5]; this result agrees with the fact that flexo-electric deformations are more facilitated by a weak anchoring than by a strong anchoring. The growth phenomenon of dynamical structures is confined to a region in an important way where the coherence length influences the behaviour.

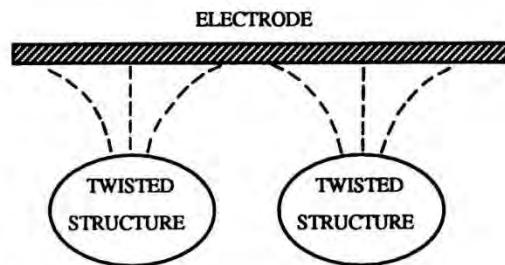


Fig. 3. A schematic cross-representation of the system when the spiral nucleation occurs: twisted structures (filaments in Fig.1) embedded in a pseudo-nematic matrix, exposed to flexo-electric disturbances, which intercept the structures. The splay-type deformations are just indicative of surface inhomogeneities and are not at scale.

In conclusion, we suggest a mechanism for spiral nucleation consequent on distortions in the director distribution near surfaces and propagating within twisted structures, twisted by a sufficiently “slack” twist – as the phenomenon is situated at the threshold of the untwisted cholesteric state – to allow this propagation (Fig. 3).

The studies connected with the flexo-electricity in cholesterics [21, 24, 25] recommend to choose materials exhibiting weak values for the dielectric anisotropy in order to provide against the inhibitory action of the dielectric effect on the flexo-electric effect. Thus, the phenomena which are involved exist before the dielectric effect is effective, i.e. the helix untwisting. In fact, during the process of a voltage increase, the flexo-electric and dielectric effects are in competition and it is always the last, by forcing an uniform orientation, which takes advantage. However, in addition to the situation described in the literature (weak values for  $\Delta\epsilon$  and weak part of the homeotropic orientation), another instance where the flexo-electricity is liable to appear is when the dielectric effect has almost achieved its function, i.e. when the voltage is fixed at a value near the threshold value corresponding to the untwisted cholesteric state. An important value of the dielectric anisotropy should not be a hindrance for obtaining a spectacular electro-optic response of the medium, a fortiori when this effect is obtained at the threshold of a mechanism – the cholesteric to homeotropic transition – whose motor is precisely the dielectric effect.

### 5.3. About the surface polarization

It is necessary to investigate the influence of a surface polarization by studying the part of a polarity induced by different surface treatments.

The direction of surface polarization vector  $\mathbf{P}_s$  can be reversed by deposition on electrodes of surface-active substances. In order to separate a surface polarization from the flexo-electric mechanism, it is possible to prepare two types of cells differing in the orienting covers and, consequently, in the type of surface polarization. (i) In the first type of cell, electrodes can be covered with layers of hydrophilic elastomers: polar cyano groups of 5CB molecules will be oriented towards the substrate. (ii) In the second type of cell, lecithin can be coated on the electrodes: an opposite direction of molecules is obtained. At this step of the study, we cannot conclude about a surface polarization contribution, which does not belong to the present investigation, but it is necessary to keep in mind a possible influence.

### 5.4. About the ordo-electric mechanism

We do not promote the ordo-electricity mechanism for two main reasons: (a) first, the formation of the structures is observed at temperatures far from the smectic A to cholesteric and the cholesteric to isotropic transition temperatures: important values for  $VS$  are improbable; (b) then, in ordo-electric point of view, there does not exist a significant variation of threshold voltage when the temperature is increased towards the isotropization temperature. The threshold value varies between 4.5 and 5.9 V when the temperature varies from 25 to 41°C (Fig. 4). This increase corresponds to a bigger effort to realign a medium which loses order, especially a certain degree of homeotropic order favourable to the dielectric effect and which preexisted in the cholesteric structure at the equilibrium.

### 5.5. Remarks

It is interesting to indicate some facts which presently limit a more quantitative description of this rich and complex phenomenon.

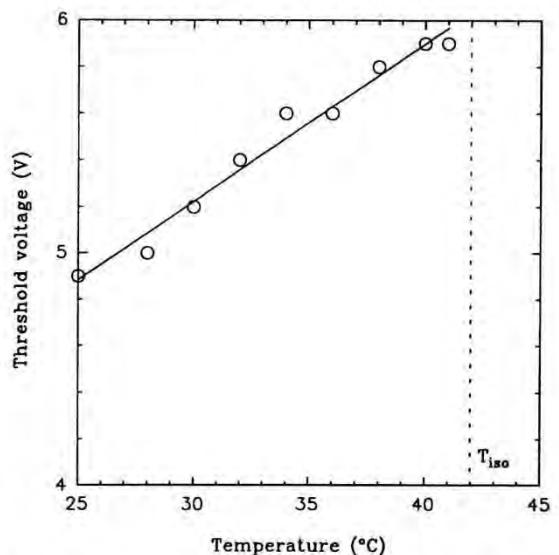


Fig. 4. Threshold voltage for the field-induced nematic state versus temperature. Cholesteric pitch = 3 μm, cell thickness = 3.5 μm.

(i) We have to determine the surface anchoring strength and the field homogeneity, which are not parameters easy to evaluate, and especially in cholesterics. (ii) We have mentioned that the importance of the surface polarization has to be estimated after a careful comparison of the results with hydrophilic and hydrophobic substrates. (iii) The theoretical studies related to large-pitch cholesterics submitted to an AC electric field and in the static case (high frequency) [26] assume a constant field, which does not seem to be reasonable in our case, due to the conductivity of 5CB and the dynamic properties of the patterns. (iv) The flexo-electric effect being related to the surface properties, often in a complicated manner and a fortiori in the cholesteric case, the evaluation of flexo-electric coefficients is not possible from a single type of experiment. (v) If we investigate a theoretical treatment by using the Frank energy expression, we can expect that the surface terms be important as the  $K_{24}$  term (called "saddle-splay") –  $K_{24} \operatorname{div}(\mathbf{n} \operatorname{div} \mathbf{n}) + \mathbf{n} \wedge \operatorname{rot} \mathbf{n}$  – which expresses the quantity of twist anisotropy in the system. Some authors have treated the problem of spiral waves, of single handedness, by a two-dimensional Landau-de

Gennes model [27]; however, this model is said not valid for the case that we describe (small values of order parameter for the unwinding transition are assumed). (vi) It would be useful to learn about the morphology of the structures. An experimental investigation could be the electron microscopy, notably by studying the contrast in cross-sections. It was, therefore, necessary to fix these field-induced structures in order to get a solid film which can be handled. Classical methods, like quenching or freeze-fractures, are hardly possible since the weak part of the polymeric material does not permit to keep the structures durably. It has been shown that this problem can be solved by creating, by photo-crosslinking, a polymeric network which traps the field-induced structures. The experimental process for enclosing the film in a resin without deteriorating it has been given [28].

## 6. Conclusion

We have described and studied the part of different mechanisms involved in the nucleation of field-induced spirals embedded in a homeotropic matrix near the untwisted state: dielectric effect, electrohydrodynamical instabilities, flexo-electricity, surface polarization and ordo-electricity. It appears that the behaviour of these dynamic structures suggests a mechanism based on the propagation from the electrodes of distortions in the distribution of molecules director. These disturbances can be related to flexo-electric-type distortions present in cyanobiphenyls nematics. Indeed, the homeotropic matrix is in majority constituted by 5CB nematic liquid crystal molecules. These distortions propagate within structures where a twist subsists. The topological nature of these structures and spiral patterns have to be defined.

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## References

- [1] L.M. Blinov, *Electro-Optical and Magneto-Optical Properties of Liquid Crystals* (Wiley, New York, 1983).
- [2] P.-G. de Gennes and J. Prost, *The Physics of Liquid Crystals* (Clarendon, Oxford, 1993; 2nd ed.)
- [3] M. Mitov and P. Sixou, *Mol. Cryst. Liq. Cryst.* **231** (1993) 11.
- [4] M. Mitov and P. Sixou, *J. Phys. II* **2** (1992) 1659.
- [5] M. Mitov and P. Sixou, *Mod. Phys. Lett. B*, **9**(15) (1995) 929.
- [6] M. Nakagawa and T. Akahane, *J. Phys. Soc. Japan* **52** (1983) 3773.
- [7] D.K. Rout and R.N.P. Choudhary, *J. Phys. D* **22** (1989) 289.
- [8] M. Monkade, P. Martinot-Lagarde and G. Durand, *Europhys. Lett.* **2**(4) (1986) 299.
- [9] O.D. Lavrentovich and V.M. Pergamenshchik, *Mol. Cryst. Liq. Cryst.* **179** (1990) 12; **192** (1990) 239.
- [10] O.D. Lavrentovich, V.G. Nazarenko, V.M. Pergamenshchik, V.V. Sergan and V.M. Sorokin, *Sov. Phys. JETP* **72**(3) (1991) 431.
- [11] O.D. Lavrentovich, V.G. Nazarenko, V.V. Sergan and G. Durand, *Phys. Rev. A* **45**(10) (1992) 6969.
- [12] G. Barbero, I. Dozov, J.-F. Palierne and G. Durand, *Phys. Rev. Lett.* **56** (1986) 2056.
- [13]  $\xi$  is the coherence length associated with electric field. It must be distinguished from  $L = K/W$ , where  $K$  is the elastic constant in the usual one-constant approximation  $K_1 = K_2 = K_3 = K$  (1, 2 and 3 indices are associated with the three basic types of deformations in liquid crystals: splay, twist and bend [2]) and  $W$  is the surface anchoring energy.  $L$  is the de Gennes-Kléman extrapolation length characterizing the strength of the surface anchoring. Generally, we have  $L \leq \xi$ .
- [14] L.M. Blinov, *J. Phys.* **4**(40) (1979) C3–247.
- [15] N. Felici, *Rev. Gen. Electr.* **78** (1969) 17.
- [16] W. Helfrich, *J. Chem. Phys.* **51** (1969) 4092.
- [17] R.B. Meyer, *Phys. Rev. Lett.* **22** (1969) 918.
- [18] J. Prost and J.-P. Marcerou, *J. Phys.* **38** (1977) 315.
- [19] H.R. Brand, *Mol. Cryst. Liq. Cryst. Lett.* **3** (1986) 147.
- [20] M.A. Osipov and T.J. Sluckin, *J. de Phys. II* **3** (1993) 793.
- [21] J.S. Patel and R.B. Meyer, *Phys. Rev. Lett.* **58** (1987) 1538.
- [22] N.V. Madhusudana and G. Durand, *J. Phys. Lett.* **46** (1985) L195.
- [23] J. Prost and P.S. Pershan, *J. Appl. Phys.* **47**(6) (1976) 2298.
- [24] J.S. Patel and S.D. Lee, *J. Appl. Phys.* **66**(4) (1989) 1879.
- [25] S.D. Lee, J.S. Patel and R.B. Meyer, *J. Appl. Phys.* **67**(3) (1990) 1293.
- [26] P. Ribiere and P. Oswald, *J. de Phys. II* (1990) 1730.
- [27] T. Frisch, L. Gil and J.-M. Gilli, *Phys. Rev. E* **48**(6) (1993) R4199.
- [28] M. Mitov and P. Sixou, *J. Mat. Sci. Lett.* **14**(21) (1995) 1518.