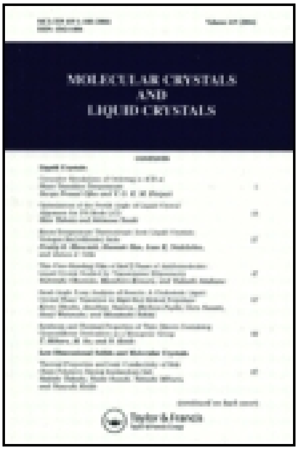

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Study of a Cholesteric Liquid Crystal Polymer in Solution in a Low Molar Weight Liquid Crystal: Phase Diagram and Electric Field Effect

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Study of a Cholesteric Liquid Crystal Polymer in Solution in a Low Molar Weight Liquid Crystal: Phase Diagram and Electric Field Effect

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We have studied the effect of an electric field on a binary mixture between a cholesteric liquid crystal polymer and a low molar weight liquid crystal from results a cholesteric polymer film in a confined geometry. The phase diagram shows, moreover an excellent compatibility between the compounds, a very large variation of the cholesteric pitch. According to the magnitude and the frequency of the applied electric field, various behaviours are observed: stable homeotropic nematic, stable birefringent filaments, "fingerprint" texture, . . . We have focused our study on the effect of an electric field on the filament texture corresponding to a large cholesteric pitch. The transition of a texture with filaments to a homeotropic structure is described by the existence of two optical thresholds (the appearance of the homeotropic and the disappearance of the filaments) whom the variation is studied according to the frequency. We are describing particular phenomena added to this general behaviour, like the nucleation and the growth under field of quasi perfect spirals issue of the bending of the filaments just before the obtaining of the homeotropic state. Besides, we give an example where the quenching of the spiral texture is possible thanks to the polymeric compound of the binary mixture. From the configuration of the filament proposed by Lequeux or Oswald *et al.*, it is possible to imagine a mechanism where, in given conditions, a propagation of the authors' structure is produced.

INTRODUCTION

The present work relates to the electric field effect on a film of a cholesteric solution whose thickness is nearly equal to the pitch of the cholesteric structure.

Contrary to the previous studies about this subject, concerning low molar weight liquid crystals, the film is formed from a mixture containing a cholesteric polymer. Precisely, it is a side-chains copolymer with mesogen and chiral groups for which the pitch of the cholesteric structure can vary by adjusting the relative proportions of the two groups. Besides, and this is one of the principal interests of polymers with regard to small molecules liquid crystals, it is possible to fix, by a rapid quench, the observed textures and to make an analysis of the obtained film. On the other hand, when we apply an electric field, there can exist a slower kinetics, associated to the nature of the polymeric compound of the mixture, and, thus, an easier observation of the modification of the texture with regard to the field variation. The characteristic times of this kinetics can often be adjusted by modifying the concentration in polymer.

In most cases, these advantages have no reality in the sense where the mixtures of a polymer in a second compound (low molar weight liquid crystal or polymer) are heterogeneous; indeed, the compatibility condition between the constituents of a binary mixture is generally very difficult to obtain from mixtures containing polymers since the entropic contribution to the free energy of mixture strongly depends on molecular weight. The work presented here is out of this difficulty because a total miscibility is obtained in a large scale of temperature and for all the studied concentrations. This situation, probably linked to strong interactions between mesogens, is rather exceptional for mixtures with a polymer.

In this article, we will quote at first (part one) the different studies concerning the cholesteric low molar weight liquid crystals in a confined geometry, that is to say, in the present case, a planar film whose thickness is of the order of the cholesteric pitch. We will recall what are the conditions for obtaining filament textures and the action of an electric field on these.

For the case of liquid crystals polymers and their mixtures, comparatively to the one of small molecules liquid crystals, there will be recalled that these filament textures have never been observed, to our knowledge.

Our experimental conditions are specified in part two. In order to examine the possible existence of filament textures in a solution containing a cholesteric polymer, it is necessary to know the phase diagram and the correlative variation of the cholesteric pitch in a such mixture (part three). In the case of a study under electric field (part four), we show that the birefringent filaments of whom characteristics look like those seen with low molar weight liquid crystals can be observed. In addition, for given specific conditions, the nucleation and the growth of the filaments in a quasi perfect spiral form can be seen. The characteristics of these spirals are studied and a qualitative discussion of their origin is presented (part five), the elaboration of a quantitative model requiring, indeed, an exhaustive study of parameters acting on these characteristics. Although we have observed the described phenomena below (in particular the spirals) in several types of mixtures—cholesteric polymer with a nematic or cholesteric (or chiral nematic) or also smectic small molecules liquid crystal—we have chosen, in this first approach, to focus our study on the simplest case in an interpretative point of view: a cholesteric polymer/nematic small molecules liquid crystal mixture. Indeed, with pure cholesteric polymers, the characteristics concerning the pitch and rheologic behaviours often limit the achievement of such experiences and the other mentioned mixtures are more complicated.

I. SITUATION OF THIS STUDY

a. Low Molar Weight Liquid Crystals

The studies on the cholesteric or chiral nematic liquid crystals are numerous [15 and references cited by the author] and the observed results (electrohydrodynamic instabilities, unwiding of the cholesteric structure) can be explained with regard to the principal parameters as follows:

- —the thickness of the sample, or more exactly the thickness/pitch ratio
- —the existence (or no) of an electrical conductivity
- —the sign of the dielectric anisotropy
- —the anchoring: planar or homeotropic.

Among all these studies, we will mostly examine the case where the sample thickness is of the order of the cholesteric pitch. When a homeotropic anchoring is favored, particular textures formed from birefringent filaments embedded in homeotropic liquid crystal and showing a pronounced degree of organization have been observed (with or without an applied electric field) under a polarizing microscope.[1-6] These textures with filaments have also been investigated in the nematics doped by chiral solvents. The pitch of the induced cholesteric structures is chosen relatively large (for example of a few microns) and the sample relatively thin (thickness is of the order of the pitch or slightly greater). The appearance conditions and the characteristics of these filaments, particularly as a function of the amplitude of an applied electric field and of the thickness/pitch ratio have been studied[1-6] and a pattern for the director arrangement within filaments was given.[2]

b. Mesomorphic Polymers and Their Mixtures

The works on the cholesteric liquid crystal polymers with an applied electric field concern the side-chains polymers or copolymers.[7] The mesomorphic polymers and the low molar weight liquid crystals submitted to an electric field show similar results (modification of textures, unwiding of the cholesteric structure, . . .) but the observation of the birefringent polymer filaments has not yet been made.

II. EXPERIMENTAL CONDITIONS

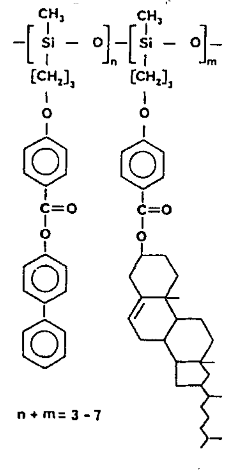
The chemical formula and characteristics of the nematic liquid crystal K15 (furnished by BDH) and the side-chains polysiloxane copolymers are respectively given in Table I-a and I-b.

Polysiloxanes with cholesterol mesogenic unit in the side-chain were synthesized by Dr. F.H. Kreuzer for Consortium fur Elektrochemische Industrie GMBH. The backbone consisted of two different monomer units which were attached to three alkyl flexible spacers. The copolymer was synthesized according to the general scheme outlined previously by Finkelmann for polysiloxanes.[8,9]

Polymeric materials offer a unique advantage over low molar weight substances in that the required cholesteric mesophase may be attained by annealing at an elevated temperature and locking the phase into the glassy matrix by a thermal quenching.[10] The selective reflection wavelength, and thus the cholesteric pitch, of a particular sample depends on the mole fraction x of the chiral component. The number of siloxane units is in the range from 4 to 7 with 5 being the predominant component; in the Table I-a, liquid crystals polysiloxanes (L.C.P.) are named by the selective reflection that they exhibit in the cholesteric phase (from red to blue,

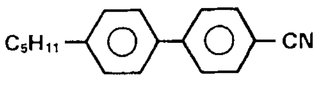
TABLE I

General formulas of used liquid crystals. a) Cholesteric liquid crystal polymer. b) Low molar weight liquid crystal



(a)

L.C. Silicone	Red	Gold	Green	Blue
Reflection wave lenght (nm)	675	595	540	450
Glass transition temperature (°C)	40/50	43/53	45/55	40/50
Clearing temperature (°C)	180/210	id	id	id
Viscosity at 150°C (Pa.s)	0,5/1,0	0,5/1,0	0,5/1,5	0,5/1,5
Processing temperature (°C)	140/170	id	id	id



K15

(b)

- Crystal / nematic transition temperature : 24 °C.
- Nematic - isotropic transition temperature : 35,3 °C.

which corresponds to an increase of the pitch). These compounds have glass transition temperatures between 40 and 55°C, depending on the m/n ratio: cholesteryl sidegroups number/biphenyl sidegroups number; above these temperatures they are cholesteric up to 180–200°C; at these temperatures they become isotropic. The reflection wavelength, depending on m/n, can be adjusted from 450 to 700 nm (for normal incidence) and by mixing the four basic materials.

Possible applications of these easily orientable liquid crystal polysiloxanes are optical data storage,[11] non absorbing optical filters[12] or the decorative sector.[13]

In our experiments, an excellent compatibility has been obtained by mixing this polymer with the low molar weight liquid crystal (L.M.W.L.C.) cyanobiphenyl K15. For systems such as sidechains L.C.P. mixed with L.M.W.L.C., the enthalpy in both isotropic and anisotropic phases will contain an unfavorable component due to the mixing of the apolar mainchain with the L.M.W.L.C. and a favorable (or, sometimes, unfavorable) component due to the mixing of sidechains with the L.M.W.L.C. Mixing or demixing will be controlled by the balance of these components.[14] It is, thus, interesting, in the presented example, to have obtained the compatibility for all ranges of concentration. This interaction between the two components leads sometimes to observe the presence of the smectic A phase, reflecting a microsegregation of sidechains and mainchains, and to the observation of the homeotropic structures where the mesogens of liquid crystal and of the polymer are aligned perpendicularly to the capacitor plates. Lastly, it can explain the important effect of an applied electric field (the dipolar moment of C-N group is worth 3.4 Debyes). The dipoles of the two mesogens may be bound but their accurate arrangement is unknown.

The textures were observed by standard means associating an optical microscope with a Mettler FP52 hot stage (equipped with optical windows) allowing the direct observation of the preparation during its heating or cooling (between crossed polarizers). The samples are consisting in conventional sandwich cells, whose thickness is adjusted by mylar sheets; furthermore, using ITO conducting transparent electrodes, a continuous or alternating voltage up to 200 volts and of a few MHz can be applied. All the study is made at room temperature (24°C).

III. PHASE DIAGRAM

Several polymer/mesogenic solvent couples were used for the L.C.P.–nematic L.M.W.L.C. mixture.

Only one of the four copolymers is used for the presented mixture, the one corresponding to a selective reflection in the blue of the visible spectrum (called "Silicone Blue"); using the other copolymers furnishes similar results. The phase diagram was obtained using differential scanning calorimetry and optical microscopy.

The Figure 1 shows the phase diagram obtained with the SB/K15 mixture. The two compounds have very different isotropic-anisotropic transition temperatures: for the polymer around 200°C and for the low molar weight liquid crystal of 40°C. The isotropic-anisotropic transition temperature of the mixture follows a quasi linear law between these two temperatures. In addition, for the large concentrations in polymer and for the relatively low temperatures (less than 130°C), a smectic A phase occurs. Most textures observed with this phase are homeotropic. The cholesteric phase between the isotropic and smectic phases presents a pitch which varies within an important gap, as a consequence it leads to a selective reflection which changes red into blue for the temperatures between 70° and 180°C and for the concentrations containing between 90% and 50% of polymer. For the lower temperatures and the smaller concentrations in polymer, the cholesteric pitch strongly increases and even up to the infinite for the smallest concentrations. Working at 24°C and selecting a phase diagram region corresponding to the concentrations in polymer around 10%, a relatively large pitch can be obtained and the textures with

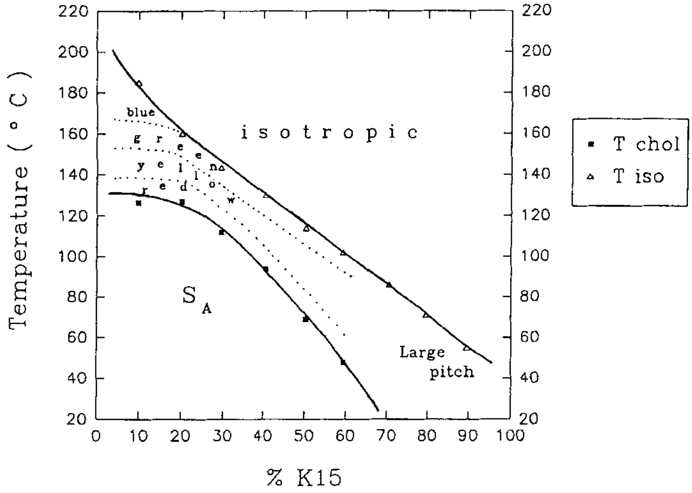


FIGURE 1 SB/K15 phase diagram. We have mentioned the qualitative variation of the pitch in the cholesteric region: selective reflections colours and large pitch zone.

filaments can be easily observed. These last will be studied in fuller details, where we will see that they are producing original manifestations during the unwiding of the helix in the cholesteric-nematic field-induced transition. Let us precise that using a mixture law for calculating the value of the dielectric permittivity leads to obtain a positive dielectric anisotropy, since the cyanobiphenyl compound (whose dielectric permittivity $\Delta\varepsilon$ is equal to 11 at 26°C) is the dominate compound (91%). On the other hand, if the electrical conductivity is not important, thus if we don't get a dynamic scattering storage mode,[15] and with $\Delta\varepsilon > 0$, we must obtain the untwisting of the structure and a homeotropic nematic at relatively strong fields.

IV. ELECTRIC FIELD EFFECTS

For the smaller concentrations in polymer, when the pitch of the cholesteric phase is important (a few microns), very original phenomena are obtained such as the filaments and their evolution under an electric field. We have examined this state in the case of SB/K15 mixture as it is particularly easy to obtain since it is performed at room temperature. The 9:91% mixture has been chosen because it is representative of the phenomena described below which are occurring when we submit a fingerprint-type cholesteric to an alternative electric field.

In our study, the filament structure is well visible when a low voltage is applied (less than five volts). Yet, the observed structure and its temporal evolution depend on the value of voltage and on its frequency. The evolution of the filaments is investigated as a function of the applied voltage at fixed frequency.

The Figure 2 shows the evolution of the threshold voltages of SB/K15 mixture (respectively 9:91%); the 3.5 μm thick sample is observed at room temperature (24°C) by optical microscopy between crossed polarizer and analyser.

The voltage from which the first optical occurrences of the "fingerprint"-

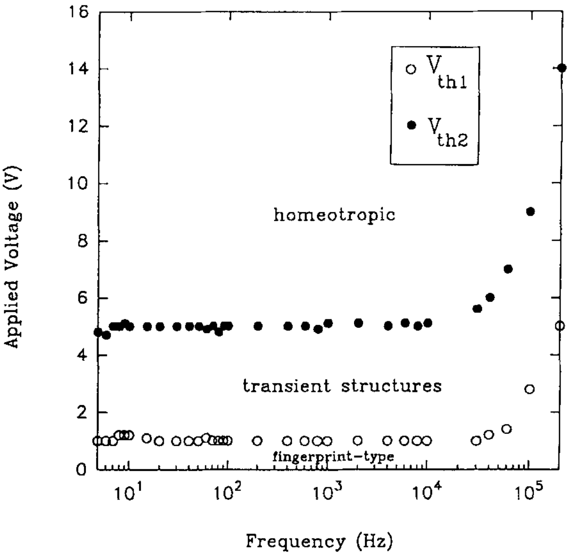


FIGURE 2 V_{th1} threshold voltage (beginning of the cholesteric-homeotropic nematic transition) and V_{th2} threshold voltage (complete homeotropic) *versus* the frequency.

meotropic transition are visible is represented by V_{th1}. This threshold voltage varies slightly around 1 V. The second threshold voltage (V_{th2}) for which the total homeotropic is obtained, varies between 5 and 14 V for the explored scale (between 5 and 0.2 MHz). The experimental precision on V_{th} is not better than 20% because the threshold value generally varies following the observed zone in the sample on account of the influence of imperfect strong anchoring at the surfaces, the presence of impurities or also the uncertainty of the thin thickness.

Beyond V_{th2} and for weak frequencies (less than 5 Hz) a turbulent regime may appear, where the fluid is put in motion, the effect detected by the observation of particles in suspension in the sample.

Between V_{th1} and V_{th2} we observe a sequence of structures as typically represented in photographies a to f of Figure 3. When the electric field strength increases, the homeotropic part of the sample increases and shows birefringent filaments embedded in a dark liquid crystal matrix. Observations being made between crossed analyser and polarizer, the transmitted light in the homeotropic nematic state is perfectly polarized in the same direction as the incoming light, from which the dark is obtained in optical microscopy. Above the threshold voltage of the cholesteric-nematic transition the sample becomes perfectly clear. A theory for this continuous phase transition was first presented by de Gennes[16] for the case of a magnetic field, and all the experimental evidence has supported this theory. Qualitatively, the theory can be understood as follows: starting with a uniform helical structure of positive diamagnetic anisotropy with a magnetic field applied perpendicular to the helical axis at low fields, the structure is in a low energy configuration. However, at higher fields values, a lower energy structure can be achieved by unwiding the helix so that at every point the molecular axis is parallel to the field. Since this requires overcoming elastic forces between the molecules, and therefore costs

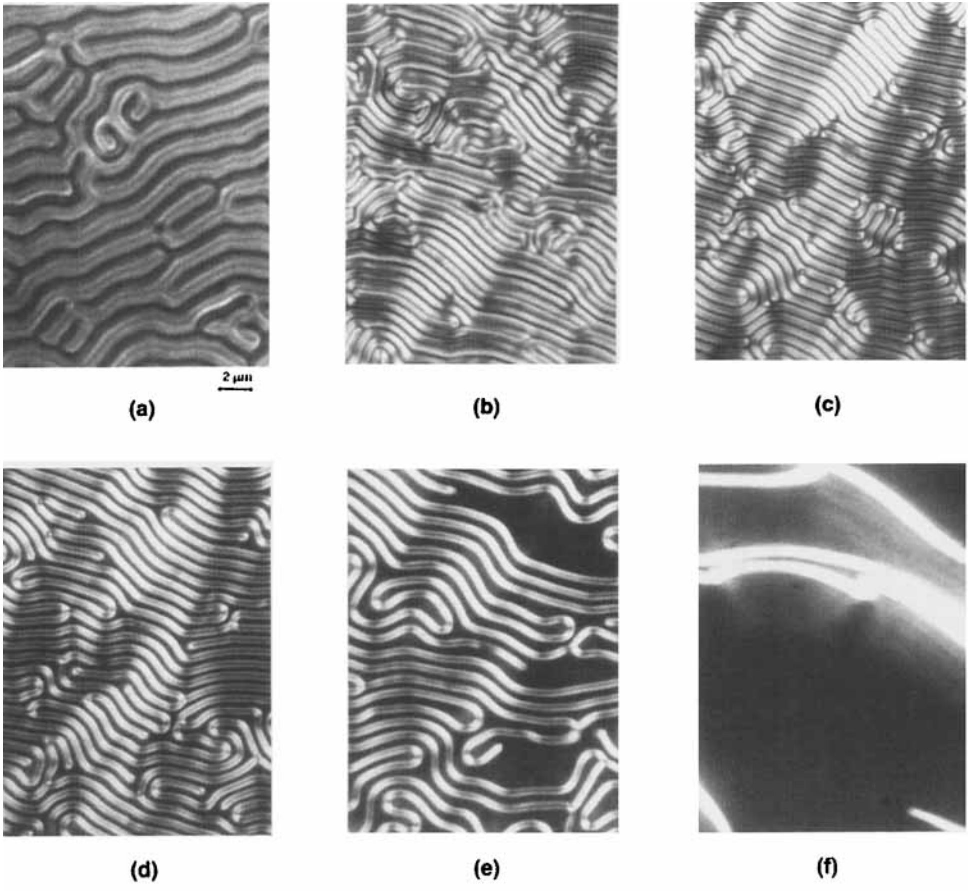


FIGURE 3 Steps of the cholesteric-homeotropic nematic field-induced transition. $f = 20$ Hz. a) Fingerprint-type texture. No field. b) $V = 2.6$ V. c) $V = 3.3$ V. d) $V = 4.7$ V. e) $V = 4.9$ V. f) $V = 5.4$ V.

energy, it does not occur until a critical high field is reached. But at intermediate fields, a distortion of the helix minimizing the sum of elastic and field energies should occur.[4]

Figure 3c illustrates a texture usually encountered for electric fields between 600 and 1500 V/mm and similar to the one observed by Asai *et al.*[3] for lower fields (about 420 V/mm) and for low molar weight liquid crystals.

When the electric field increases the birefringent filaments collapse. Various modes of the filament retractation are seen whom the sequences are described below.

—At weak frequencies (less than 5 Hz), the observation is very perturbed by the phenomena of instabilities. When the period of the applied voltage becomes of the order of the building up time of the structure, it is not possible to obtain stationary structures; for low frequencies and elevated voltages the system goes successively (during one half a period) through all the structures of the sequence, then describes again the sequence in inverse sense, and so on. The filaments are subjected to beats and alternatively present a blue birefringence (also existing in the absence of electric field) and a yellow one (characteristic of a filament which is going to retract itself).

—Between 5 and 40 Hz and for voltages near V_{th2} (in a band whom the inferior boundary is about 75% of V_{th2}) it is important to remark that the filaments bend and spiral around themselves (Figures 4 and 5) and this in less than one minute. A detailed description is given at the end of this part.

—Between 10 and 500 Hz the filaments are submitted to a very varied alteration of their birefringence (from blue to violet through yellow). Figure 7 shows a structure where several birefringences coexist.

—Between approximately 100 and 500 Hz and always for voltages nearly equal to V_{th2}, we note the presence of the delimited and particular domains which coexist with the standard filaments. In these domains the filaments move around and regenerate periodically (Figure 8). Within this frequency range it is the last structure to resist before the homeotropic takes place.

—Between 500 Hz and 100 kHz the most characteristic effect is the birefringence change of the filaments. Figures 9a and b show the spectacular effect of the birefringence change for a very weak increase of voltage.

The Spiral Texture

There exists a zone in the phase diagram {frequency, threshold voltages} for which a spiralled configuration of the filaments† is observed.

The characteristics and typical obtaining conditions of these spirals are as follows:

—It is necessary to apply a voltage for which the homeotropic is the majority in the sample, that is to say between 0.75 V_{th2} and V_{th2} (by inferior value). If V_{th2} is raised by a sweeping rate too quickly, the phenomenon may not be perceived.

—The band of frequencies mentioned for the nucleation of spirals (between 5

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†Similar spirals structures have also been observed in the SB+CE1 mixture by M. Kamayé and J. M. Gilli in our laboratory.

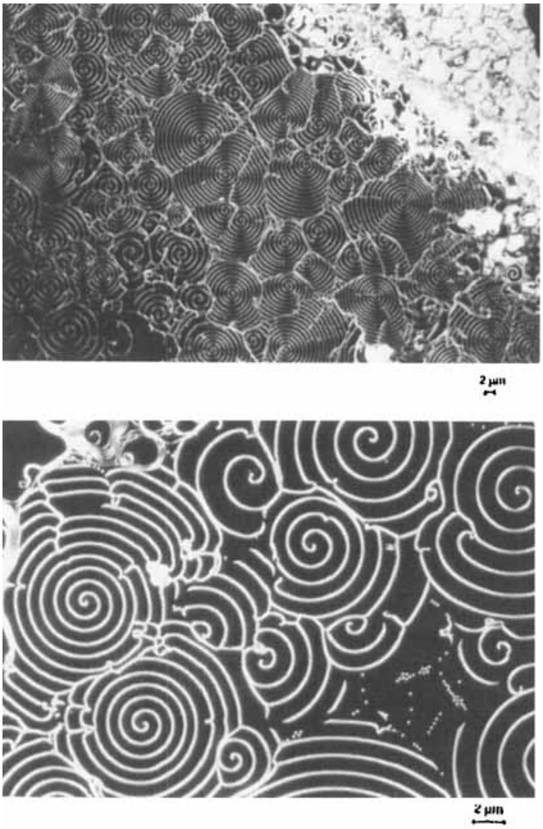

FIGURE 4 Spiralled filaments.

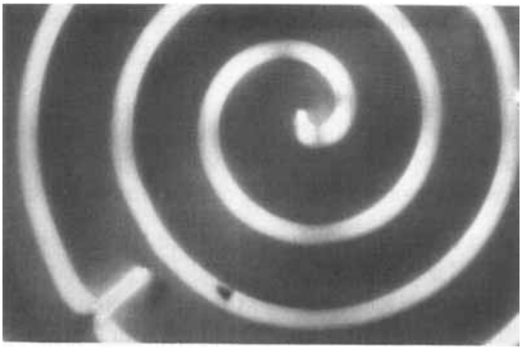

FIGURE 5 Zoom of a spiral.

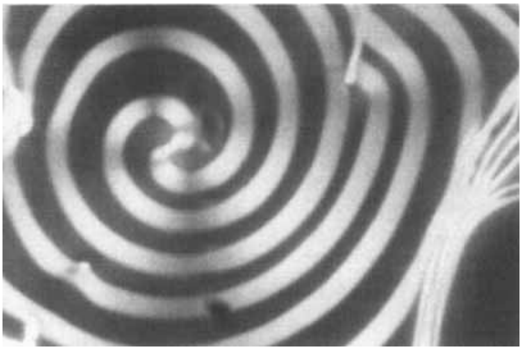

FIGURE 6 Double-spiral.

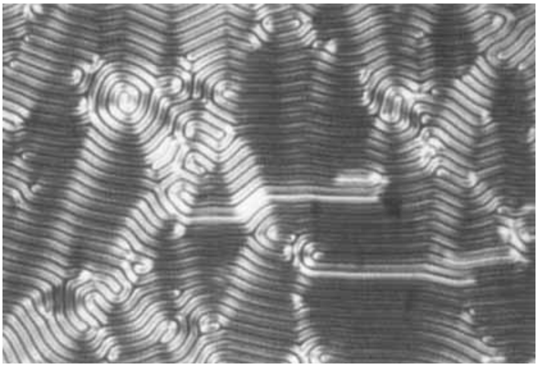

FIGURE 7 Several birefringences coexisting at $f = 300$ Hz and $V = 3.5$ V.

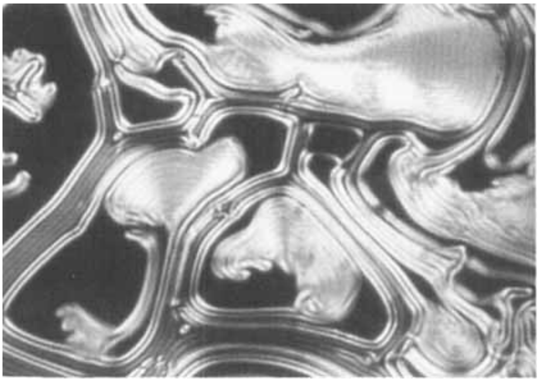

FIGURE 8 Particular domains where filaments regenerate periodically. $f = 8$ Hz. $V = 4.5$ V.

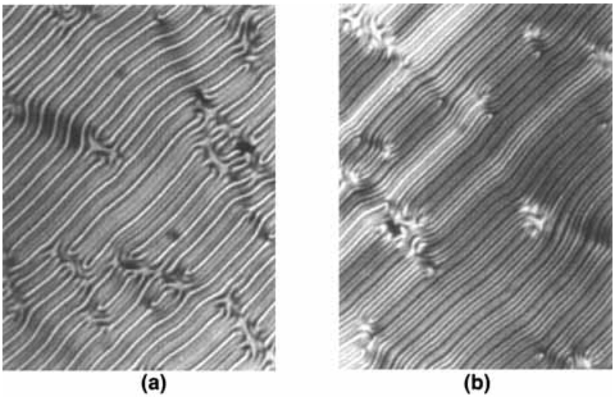

FIGURE 9 Strong effect on the birefringence for a very weak increase of voltage. f = 20 kHz. a) V = 3.30 V. b) V = 3.44 V.

and 40 Hz) must be taken account for an observation time of less than one minute. It is possible to observe spirals at higher frequencies but for longer times; we don't consider, here, longer observation times.

—Spirals can grow until paving all the sample (Figure 4) if the observation, at fixed frequency and voltage, is made during a sufficiently long time.

—These spirals are the last structures which are resisting to the field-induced homeotropic propagation.

—We observe left-handedness like right-handedness spirals.

—There can exist several spiral "pitches" for a same couple (frequency, voltage).

—We can also observe "double spirals" (Figure 6) which seem resulting of the connection, by their extremities, of a right-handedness spiral with a left-handedness spiral (see discussion part for the possible condition of connection).

—Figure 10a shows the variation of V_{th2} (for the low frequency regime) versus the thickness of the sample (always for the 9% SB/91% K15 ratio, so for a fixed value of the pitch). The band where spirals can be observed is also mentioned; it follows up linearly the V_{th2} variation. For each sample, spirals have similar geometrical features. Their nucleation seems not to depend on the thickness, thus the thickness/pitch ratio; nevertheless, the thicker the thickness is, fewer the number of spirals.

Quenching of the Spiral Texture

We are giving an example exhibiting the possibility to fix the spiral texture. We have chosen a L.M.W.L.C., M24 (cyanobiphenyl furnished by BDH), which presents, when it is mixed in SB (we keep the same ratio 9% of polymer and 91% of L.M.W.L.C.), the cholesteric phase at higher temperature than room temperature. The transition temperatures for the mixture are successively:

T (K-S_A): 53°C
T (S_A–fingerprint type cholesteric): 66°C
T (cholesteric-isotropic): 87°C.

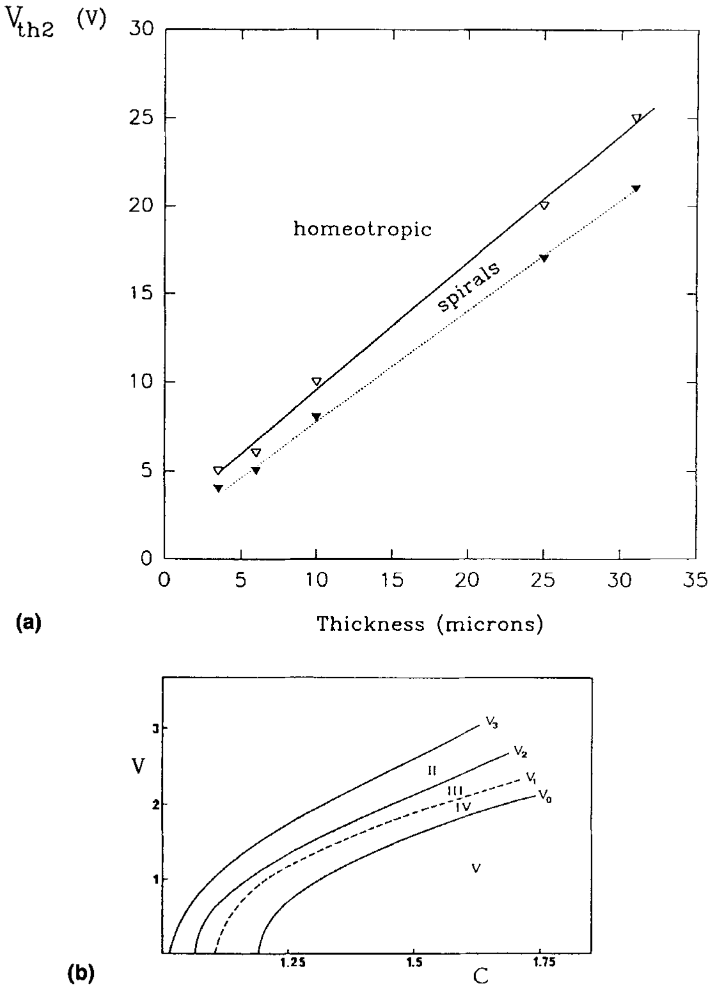

FIGURE 10 a) V_{th2} threshold voltage (complete homeotropic) evolution *versus* the sample thickness. We have also mentioned the band where spirals are appearing. b) Theoretical phase diagram of "cholesteric fingers" under electric field from Reference [5]. V is the applied voltage to the cell. C is the dimensionless confinement ratio: *d*/*p*, where *d* is the thickness sample and *p* the natural cholesteric pitch. The different zones correspond successively to: I - nematic phase, II - metastable isolated fingers: they disappear from their ends but their width is well defined, III - stable isolated fingers: they lengthen from their ends, IV - periodic pattern, V - transient translationnaly invariant configuration.

We have succeeded in doing a cooling of a sample (the experimental conditions are the same as before) between 80 and 24°C though the cooling speed was slow (0.2°C by minute). The spiralled form of the filaments, in a homeotropic matrix, has been kept. When we have switched off the field (at room temperature), what was homeotropic (black between crossed polarizers) has become a fingerprint-type texture whereas spiralled filaments were subsisted.

Figure 11 shows the result: field-induced spirals in a fingerprint matrix. This supercooled texture has been preserved about ten minutes; then it is become

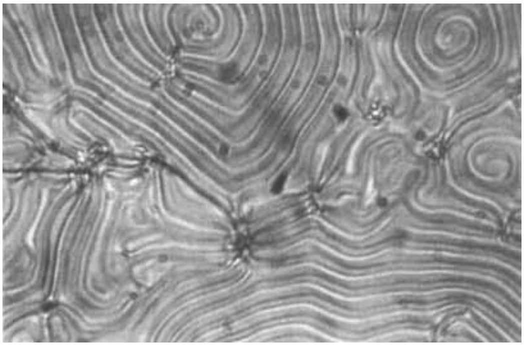

FIGURE 11 "Fixed spirals": field-induced spirals in a fingerprint matrix. The sample has been cooled from 80°C (from a spiral texture) to 24°C (room temperature) on account of 0.2 °C by minute. The spirals have been kept. When the field has been switched off, the homeotropic is become fingerprint-type. .

crystalline again on account of the metastability of the mixture. By taking account several parameters like the nature of the L.M.W.L.C., mixtures between two polymers or transition temperatures, the possibility to obtain a more stable fixed texture can exist. The influence of the polymeric compound of the mixture has been clearly underlined.

DISCUSSION

In low molar weight liquid crystal scope, the optical phenomena connected to these filaments and studied by various authors consist either of a parallel band alignment of the filaments—a little as the Williams' striped domains patterns—or of a filament juxtaposition in domains recalling those of the texture named polygonal fields (8), or still of a domain aggregation inside which the filaments are wound into joined figures. The observation and the modelisation of filaments have been studied by several authors but no spiral formation born of these filaments has been reported. The Press and Arrott's studies by digital simulation allow to specify the structure of given filaments,[2] and particularly the arrangement of the director within the filaments. From the assumptions of a translationnal invariance in at least one dimension (direction parallel to the capacitor plates) and of an equality between the three elastic constants of "bend," "twist," "splay," the authors' computation consists in treating the equations describing the energy of such a liquid crystal texture. The obtained solutions depend on a critical thickness z_0 (equal to $p_0/2$ where p_0 is the equilibrium pitch) below which only the homeotropic solution is stable. A first configuration called "translationnally invariant cholesteric," can be visualized as having the director rotating with height on a cone whose axis lies along homeotropic (z) axis. The half angle of this cone varies from $\Theta = 0$ at the surfaces to some Θ_{max} halfway between the surfaces. The second configuration called "periodic cholesteric," stabler than the previous one, can be visualized by a periodic structure inside which the helix axis is tilted with regard to the z axis.

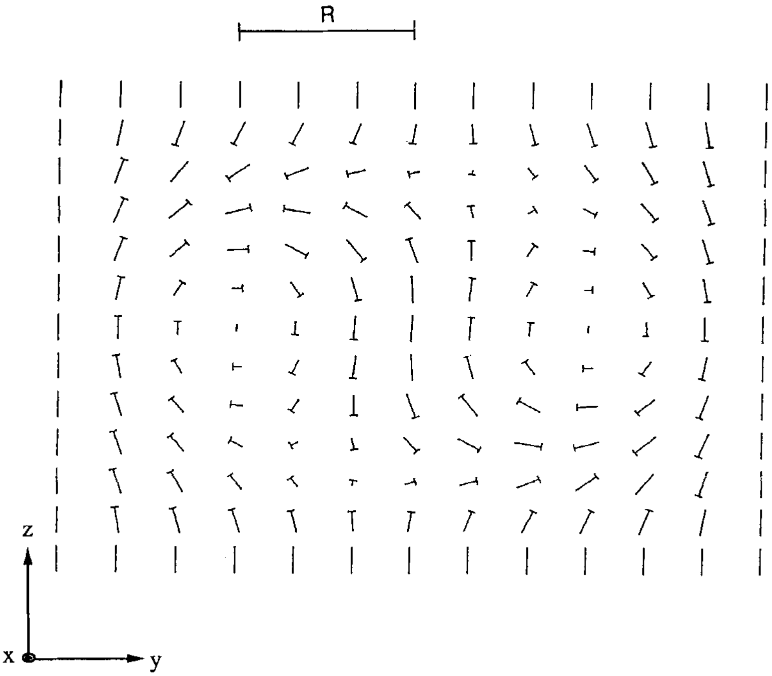

FIGURE 12 Director-field representation of a "cholesteric finger" by a cross-section (from Reference [17]). The head of the nail is behind the plane of the figure and the tip is pointed on the observer.

Nevertheless, Press and Arrot haven't examined the part of the elastic anisotropy that has prevent them from giving an entire description of the cholesteric filaments. Lequeux, Oswald *et al.*[17,18,5] have studied the geometry of cholesteric liquid crystals taking into account the elastic aspect of these structures. They have given a representation of "cholesteric fingers" similar to Press and Arrott's (see on Figure 12 the representation given by Lequeux) which may permit us to understand the steps of the cholesteric-homeotropic nematic transition and the spiral texture.

Cholesteric-Homeotropic Nematic Field-Induced Transition

Experimentally, there exists one structure more stable than the homeotropic one (where the director of the molecule remains constant), the filament structure. Lequeux, Oswald *et al.* have created a model for this structure.[17,18] Ribière and Oswald[5] have studied the changing of textures (stable nematic phase, stable filaments, periodic patterns, . . .) according to the magnitude of the electric field and the ratio between the cholesteric pitch p and the sample thickness d (Figure 10b). We examine the different zones of the phase diagram {Voltage, d/p ratio} described by these authors and, in order to compare our results with their expectations, we have considered, in Figure 10a, the variation of the threshold voltage *versus* the thickness. If we convert the thickness scale into a d/p ratio scale (the pitch of our mixture is about 3 μm), we obtain, like expected in their model, a linear variation of the threshold voltage V_{th2} *versus* the d/p ratio. Even the qualitative results depend, evidently, on the values of the elastic constants, we can remark that we obtain a slope of about 2.2 volts when the slope (of the line delimiting I and II domains in Figure 10a, where I represents the nematic phase and II the metastable isolated fingers region) obtained by the authors is, for a ratio between 1 and 1.75 about 5 volts for the experimental phase diagram and 3.5 volts for the theoretical one (obtained from the calculations of reference [18] including electric field effects). Thus, our slope value is in good agreement with the theoretical value. We can't make more investigation on account of the difference between the two explored scales for the d/p ratio (between 1 and 1.75 for the authors and between 1 and 10 for us) but we can also remark that the ordinate at the origin in our case (2.2 volts) is nearer of the experimental value (2.1 volts) than the theoretical one (0.9 volts). Which concerns the optical response, when we will apply a field, the first region where the molecules will align themselves along the field will be the region near the walls, since it is the region where the dielectric torque has a strength of the order of the elastic forces (there exists one length along the z axis, at $z = 0$ and $z = d$, where the molecules are already orientated according the field axis). Then, when the field increases, we observe the growth of homeotropic zones between the filaments. An examination of the structure of two adjacent filaments (Figure 12) shows that the frontier zone between them contains molecules which already possess the homeotropic orientation before applying the field. So, the field has for effect to increase the correlation of the molecules belonging to this frontier zone with the neighbouring molecules (by forcing the homeotropic direction) according to the z-axis, on the one hand, and according to the x and y-axis on the other hand. A consequence of this last effect is a decreasing of the filament thickness when the field increases (like it can be seen in Figure 3). The experimental study shows that the V_{th2} evolution as a function of the frequency exhibits, in fact, two types of regimes, distinguished by a frequency f_c (for which V_{th} diverges), around 20 kHz, appearing as a limiting frequency between two regimes (rather a real cut-off frequency). This behaviour is like nematics one where two regimes are also observed[19]: for the first regime, the low-frequency regime, V_{th2} is linear compared to f, whereas in the second regime above the critical value f_c, the high-frequency regime, V_{th2} is varying like $f^{1/2}$. However, the analogy must be made with precaution since, in the existing models for the threshold values prediction, the sample conductivity is not taken into account.

Spiralling of the Filaments

Using a chiral compound in a L.M.W.L.C., Ribière and Oswald[5] have observed the nucleation and the growth of the cholesteric filaments under electric field. Although they have not observed spirals, two aspects of their study can permit qualitatively to understand the existence of spirals: the asymmetry between the two extremities of one type of filament and the helicoidal structure of the director within the filament. Which concerns the internal structure of a filament, we see in Figure 12 that, from an orientation parallel to the x-axis for a molecule at the center of the filament, it is possible to be on the way to obtain a homeotropic orientation (according the z-axis) at a distance near of the radius filament R, by a director rotation of the central molecule either according to the y-axis, or according the z-axis. Nevertheless, this transition can be carried out by a "left" or a "right" rotation, which confers a different structure to the cylinder constituting the filament. Thus, when a field is applied, we can imagine that for a certain threshold value, the molecules will have a tendency to adopt a spiralled configuration due to the intrinsic filament structure. Besides, Ribière and Oswald have examined the topology of a "finger" tip and they have shown that for a segment of finite length, two configurations are possible experimentally: either the two ends of the filament are identical and there is always a defect inside the filament or there is no defect and the two ends of the finger are not the same, one end being sharper than the other. The asymmetry introduced by the existence of these two sorts of tips for a cholesteric filament, added to an internal helicoidal structure having a right or left sense, should explain that, under the action of an electric field, the cholesteric filament in situation of spiralling presents two types of handedness. Besides, the authors are specifying that two ends of opposite signs collapse together while two ends of the same sign repel; this fact should explain the formation of double-spirals issue of the connection of two spirals by the tips which can result of spirals having different handednesses (see Figure 6).

CONCLUSION

We have presented a binary mixture of a polymer in a low molar weight liquid crystal which, in addition to the very wide variation of the cholesteric pitch that it exhibits, is providing an example of a cholesteric-nematic field-induced transition where original manifestations of the unwiding of the helix are occurring: a spiralled formation of cholesteric filaments just before the homeotropic nematic state. Some qualitative elements have been given, in this first approach, to try to understand the "spiral texture" and its characteristics.

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