Liquid Crystals
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713926090

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To cite this Article Gilli, J. M. and Kamayé, M.(1992) 'First observations of a spiral instability at the smectic A—cholesteric transition under an electric field', Liquid Crystals, 11: 5, 791 — 796
To link to this Article: DOI: 10.1080/02678299208029031
URL: http://dx.doi.org/10.1080/02678299208029031

LIQUID CRYSTALS, 1992, VOL. 11, NO. 5, 791–796

First observations of a spiral instability at the smectic A–cholesteric transition under an electric field

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(*Received 14 October 1991; accepted 14 January 1992*)

Archimedian spiral wave instabilities have been discovered in chemical [1] and biological systems [2]. We present here the first example of such an instability pattern, encountered in a physical medium [3]. This instability is directly observed by polarized optical microscopy, on a positive dielectric anisotropy smectic A sample with homeotropic organization between parallel glass slides. The arm of the spiral is probably constituted of a 180° Bloch wall, separating indistinguishable smectic domains, and incorporating progressively the helicity of the cholesteric phase, excluded from the smectic.

Our recent work on a co-oligomer side chain liquid crystal [4, 5] demonstrates the interest of this low kinetic material for the investigation of the transitional aspects of blue phases. Moreover blends of this compound with low molar mass liquid crystals allow the observation of a smectic A to cholesteric transition of adjustable cholesteric pitch and temperature conditions [6].

The particular case of a 50 per cent (w/w) concentration of this high pitch co-oligosiloxane with a commercial cholestric, CE1, of opposite chirality, gives the opportunity to study this phase transition at T_{S_ACh} of about 160°C on samples enclosed between untreated glass slides, with variable thickness, starting from a low temperature homeotropic configuration.

When the temperature is increased above T_{S_ACh}, starting from this homeotropic texture, a spectacular transition is observed consisting of the nucleation and growth of strongly birefringent fingers. By changing the experimental parameters such as the blend concentration, C per cent, or the rate of heating, two very different fingers are observed. The first one, strongly coloured by Newton phenomenon, appearing with fast heating rates, are identifiable to those described by Oswald [7] and also observed during the unwinding of a cholesteric texture, induced by a decrease of the sample thickness [8]: they correspond to the direct growth of a periodic TIC cholesteric distorted organization. The second ones, with a white tint, occur for a low heating rate and give the optical appearance of cylindrical shaped objects which deviate the incident polarized light strongly (see figure 1). It is of fundamental importance to emphasize here, that, within a temperature range of a few degrees, it seems possible to observe the thermodynamic coexistence of these second type fingers and homeotropic smectic domains: this fact allows us to make the assumption of an existing intermediate phase between the lower S_A phase and the upper Ch, analogous, in this particular glass slide geometry, to the Shubnikov phase, for superconducting materials under magnetic fields [9, 10]. The nucleation and growth of these second type fingers with increasing

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0267–8292/92 $3·00

Figure 1.

Figure 1. The second type fingers observed between crossed polars without an applied field: the large fingers on the lower part of the figure correspond to the association of four single fingers, as that observed in the upper part of this figure.

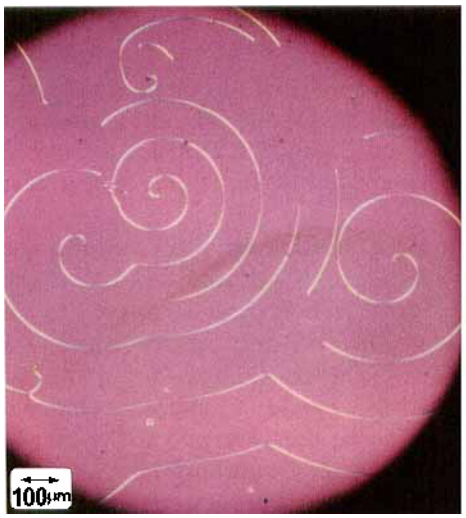

Figure 2.

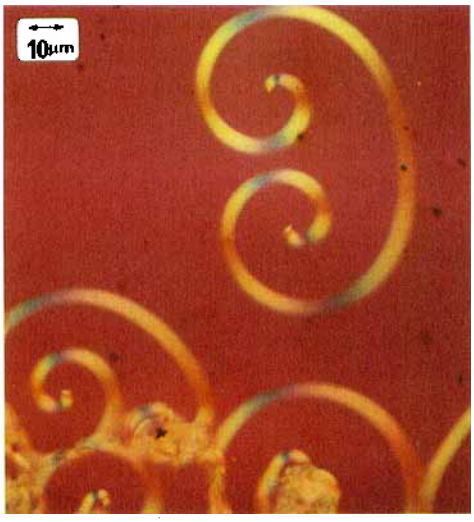

Figure 3.

Figure 2. Different pitch spirals are observable here in the presence of the AC electric field: they rotate around their axis constituted by the impurities. The observation is made with crossed polars and a 45° inserted quartz first order retardation plate.

Figure 3. Observation as in figure 2. Associated double spirals rotating in a reverse sense as a Frank–Read source. This situation is obtained when the electric field is abruptly applied to a second kind straight finger. This phenomenon is only observable for a short time and a higher order turbulent instability visible on the lower part of the figure rapidly invades the whole domain.

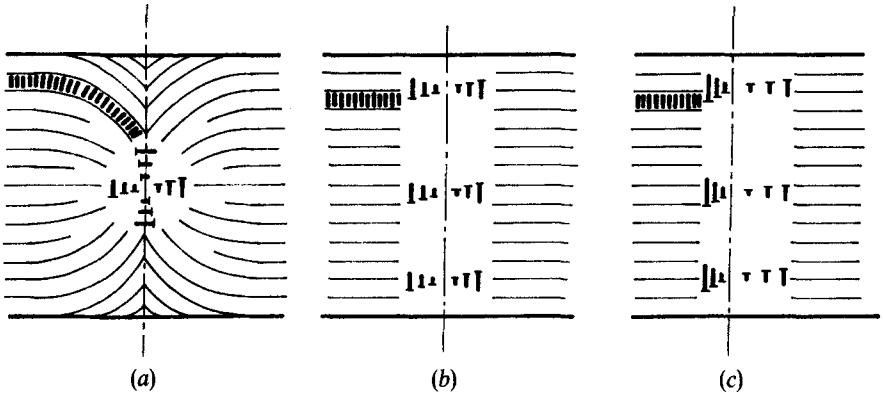

Figure 4. (*a*) A possible model for the transverse organization of the second type fingers nucleating from the unperturbed homeotropic S_A phase when the temperature is slowly increased: such a double twist structure is probably compatible with imperfect smectic order in association with helical screw dislocations of the layered organization (analogue of the Abrikosov vortex). The relation existing between this director field and the classical smectic oily streak models should be noted. (*b*) The complex director organization is in fact very close to the 180° twist wall (generally called a Bloch wall in the case of magnetic domains. (*c*) The destabilizing torque exerted on the central part of a Bloch wall (in the case $\Delta\varepsilon > 0$) can lead to such an initial deformation of the twist structure. This deformation does not lead to a static minima of the free energy but a dynamic drift of the wall can be imagined on this basis. This kind of deformation can be transposed to the more realistic model of figure 4 (*a*).

temperature, obey a very particular behaviour, excluding branching or coalescence: they nevertheless fill progressively the observed slide area by a straight progression or by bending and multiple associations as seen in figure 1. The observation of the extremity of these fingers reveals the probable presence of a point singularity in the director field. These arguments and others (more extensively developed in a paper in preparation) allow us to hypothesize on the double twist cylinder nature of these objects as shown in figure 4 (*a*): in fact this double twist organization probably gives the possibility of an imperfect structural coexistence between twist and smectic layering with favourable energy, relative to both the cholesteric or smectic perfect ones. It is important here to note the relation existing between this model and the naive twist walls expected in this particular geometry in the absence of strong anchoring conditions (see figure 4 (*b*)).

The same sample sandwiched between glass slides was investigated in the presence of an electric AC field, by the use of conducting transparent electrodes. The observation of the transition, starting from the homeotropic S_A texture, reveals a strong influence of this field on the temperature at which, the structure appears, and the geometrical ordering of the birefringent fingers.

The temperature of appearance of these fingers, precursors of the Ch phase, is increased by the presence of the field (see figure 5). This behaviour originates from the stabilizing effect of the electric field associated with the positive dielectric anisotropy of the mixture (CE1 is a strongly polar nematogen because of the C–N bond parallel to the long molecular axes).

For particular values of the frequency and amplitude of the AC field, a slow heating rate for a 20 μm thick sample, reveals heterogeneous nucleation around the impurities present in the bulk of the sample, of a constant speed rotating birefringent spiral, shown

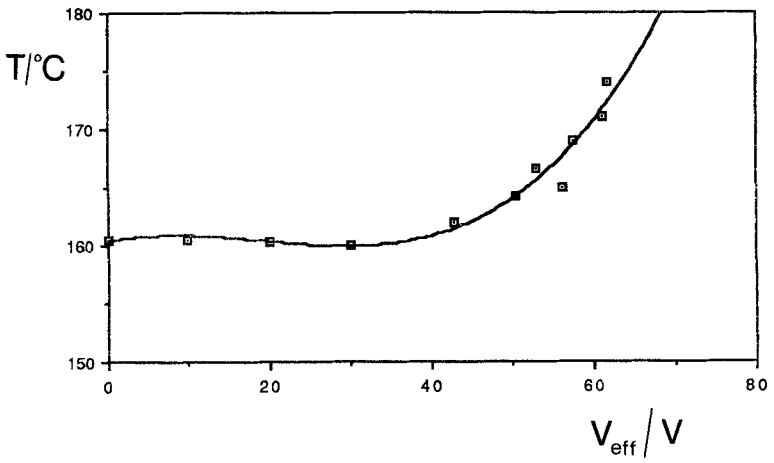

Figure 5. Temperature of appearance of the birefringent fingers versus the applied AC electric field strength: the moving spiral shaped fingers are observable, in the 160–180°C temperature domain, for an applied voltage above 35 V and a frequency $10^3\,\text{Hz} < f < 10^4\,\text{Hz}$, when the temperature is increased or when the electric field is decreased.

in figure 2, which progressively extends its length. An interruption of the heating allows the observation of this stabilized phenomenon: the quasi-perfect spirals give the optical impression of a rotation at constant speed, with both right or left possible senses. The spirals are limited, in their radial extension, by the collision of their external arms with other spirals nucleated in the vicinity. These collisions lead to a spectacular breaking or association of the arms continuing their transverse progression with the same speed. Spirals of different pitches can be observed simultaneously in the microscope field, revealing the constancy of this speed: indeed, the simple relation $S = \theta a/2\pi$ relates, far from their axis, the arm transverse speed S to the rotational speed θ and to the pitch a of these Archimede's spirals. Two different coexisting spirals 1 and 2 with $\theta_1 = 2\theta_2$, $a_2 = 2a_1$, (see figure 2) demonstrate the constancy of this electric field induced transverse speed of displacement at a given temperature and field. Keeping other parameters constant, there exists an important temperature domain (≈ 15°C) allowing the observation of these stable spirals for the range of applied voltage V_{eff}, accessible with our apparatus (see figure 5). An important reduction in S is observed when the temperature is decreased. The different coexisting spiral modes result in different ratios between the observed surfaces occupied by the unperturbed S_A and birefringent nucleating thick walls. The typical values of S observed are increased approximately from 10 to 100 $\mu\text{m s}^{-1}$ in the range of V_{eff} applied, with a rotation speed of 1 to $10^{\circ}\,\text{s}^{-1}$.

The aspect of these spiral shaped fingers is modified as compared with the previously described second type finger, seen without the electric field in the same conditions of low heating rates. Far from the spiral axis, their transverse structure observed between crossed polars shows only poor variations of the transmitted intensity. A rather sharp interface separates them from the unperturbed homeotropic domains. In the neighbourhood of the impurity carried in rotation by the spirals and constituting their origin, helicity is observed along the arm. Their free extremity, rather different from that observed without the field gives the impression of a terminating rolled sheet.

A rather different process of nucleation of these spirals has been observed with samples free of impurity. Starting without an electric field from a nearly linear second type finger of finite length, the application of an increasing voltage leads to a progressive bending of the finger and, as seen in figure 3, to the association of two spirals of reversed rotating sense. Their geometrical transformation is reminiscent of the Frank–Read dislocation multiplication process, well known by metallurgists.

All of these field-induced nucleation effects present a high frequency cut-off at about 10^4Hz: above this frequency the nucleation is rather similar to the without AC field case. At low frequencies, other isotropic or anisotropic typical electrohydrodynamic instabilities, involving translational flows, appear and all of the spirals previously described become unobservable. At the actual level of our study, no clear effect of the frequency on the rate of rotation or on other parameters of the spirals has been detected in the possible domain of study (10^3 Hz $< f < 10^4$ Hz).

Recent work [11, 12] in the field of excitable medium theories using a perturbative approach, predict the transverse drift of magnetic chiral Bloch walls under the action of a magnetic field rotating around the optic axis of the twist. This phenomenon can be simply understood by a screw-nut translation–rotation coupling: the tendency of the wall molecules to align with the magnetic field drives them in the rotation. This cooperative rotation is impossible in the normal magnetic regions, but it brings the equiprobable left- or right-handed Bloch wall present in the sample to move in both possible transverse translations. As suggested in [11] this mechanism can lead to the observation of spiraling fronts rotating around Bloch lines provided that one extremity of the drift wall remains attached on a fixed point of the sample.

In spite of the striking analogies existing between this theoretical approach and our experiment, we must underline now the main differences existing between our experimental case and this theoretical model.

We are dealing, in our previously described experiment, with electric fields and the nature of the interaction there is rather more complicated (the material is not ferroelectric) and the characteristic times of the permanent dipole reorientations is translated in this kind of material by strong variations of $\Delta\varepsilon$ with the applied field frequency.

The applied field is here alternative and not rotative: a simple analogy with the well-known monophased electric motors reminds us that an equiprobable rotation torque can be obtained on the rotor in both rotation directions. The alternative field must be decomposed in rotating components with two opposed senses. The rotor rotating in an arbitrary sense undergoes the main influence of the same direction component: the other component is seen with an apparent higher frequency and effects of the reverse torque can be neglected to a first approximation.

The handedness of the arms is not arbitrary, but is fixed here by the chemical composition of the blend and all of the walls probably possess the same type of chirality.

It is important here to note that the association of these two last differences are annihilating, bringing about, as in the theoretical case, the possibility of an arbitrary direction of the displacement for the walls.

If we try now to make the transposition of the model of [12] closer it is important to remark that the moving walls are obtained in the case of a second order perturbation. This means that this displacement is necessarily associated with a smooth deformation of the twist structure of the wall of the kind of the naive representation shown in figure 4(*c*). This can be a simple way to understand the displacement and curvature of the wall

in terms of superficial tension differences between both wall interfaces. Moreover this deformation seems necessary to find a resulting torque on the twist structure at low field frequencies: in the case of an undistorted $+\pi/2$ $-\pi/2$ twist structure, the integrated torque appearing in the analogue of a Zwetkoff experiment goes to zero.

Two other main remarks can be made to invalidate the attempt of analogy of our experiment with the model previously described.

(i) No frequency variation has been found experimentally up to now: while the theory predicts a linear dependence of the displacement speed for a low rotating field frequency and near threshold amplitude of the field. Consequently, we have to suppose our experiment to occur in a strongly saturated regime and (or) in the case of very high excitation frequencies compared to the analogue of the critical frequency for a Zwetkoff experiment [13].

(ii) The field amplitude effect on the same displacement speed is in our experiment particularly difficult to extract; the field amplitude having here a secondary effect in particular on the temperature of observation of the phenomenon due to the complex influence on the unperturbed smectic domains. The increase of the speed of displacement of the walls when the voltage is increased is also understandable through the probable decrease with temperature of the rotational viscosity constant γ_1 involved in the 180° director reversal (with a characteristic time $0{\cdot}05 < \tau < 0{\cdot}5$ s) at the crossing of the walls. As a concluding remark concerning this complex experiment we want to recall that in such a multicomponent system, the orientational order variations probably couple with the concentration gradients and that the translational diffusion constant has a possible importance comparable to γ_1.

The authors would like to thank P. Coullet and L. Gil for fruitful discussions.

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