Experimental and Theoretical Study of Electroconvection in Homeotropic Nematic Liquid Crystal

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Electroconvection patterns have been studied in a homeotropic nematic liquid crystal, experimental and theoretical, with unusual combination of material parameters, negative conductivity and positive dielectric permittivity anisotropies. Exploring the complete phase diagram in the (U, q) space three distinct morphologies, rolls and squares, soft squares and hard squares have been identified. We perform a linear and nonlinear analysis of the full nematohydrodynamic equations and carry out simulations of a suitably constructed Lorenz model. We obtain in agreement with the experiments modulated rolls and squares at threshold.

Keywords: nematic liquid crystal, electrohydrodynamic instabilities

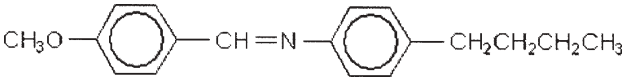
As it is well known, the nematic liquid crystals are anisotropic systems, the application of external fields (thermal, electric, magnetic) produce specific instabilities [1] – [2] with periodical roll type structures, straight or oblique, square and hexagons.

Commonly, when referring to electroconvective phenomena (EC) of nematic liquid crystals, we actually understand the standard model, meaning the model where the liquid crystal molecules are planar aligned, the dielectric anisotropy $\Delta\varepsilon$ is negative and the electrical conductivity anisotropy $\Delta\sigma$ is positive. Like A. Buka, N. Eber and W. Desch have showed in the paper "Convective Patterns in Liquid Crystals Driven by Electric Field", in reality there exist eight distinct cases depending on the molecules mode of alignment and on the positive or negative values of the dielectric anisotropy $\Delta\varepsilon$ and of the electrical conductivity anisotropy $\Delta\sigma$,(table 1).

Table 1
DIRECTOR ALIGNMENTS AND THE SIGN OF DIELECTRIC ANISOTROPY $\Delta\varepsilon$ AND ANISOTROPY OF THE ELECTRICAL CONDUCTIVITY $\Delta\sigma$

Case	Molecular alignment	$\Delta\varepsilon$	$\Delta\sigma$
A	planar	< 0	> 0
B	homeotropic	> 0	< 0
C	homeotropic	< 0	> 0
D	planar	> 0	< 0
E	planar	> 0	> 0
F	homeotropic	> 0	> 0
G	planar	< 0	< 0
H	homeotropic	< 0	< 0

Obviously, the standard case is case A, the most representative compound of this class being the MBBA nematogen,



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well studied theoretically and experimentally [3 – 5], or I 52 type nematics or Merck Phase 4 and 5.

In this paper we shall study the case B, theoretical and experimental, when the liquid crystal molecules are homeotropically oriented, the dielectric anisotropy is positive and the electrical conductivity anisotropy is negative and we limit ourselves solely to the conduction regime. In fact pairs of systems connected by this reversal transformation (cases A $\leftrightarrow$ B, C $\leftrightarrow$ D, E $\leftrightarrow$ F and F $\leftrightarrow$ G in table I) show close analogies.

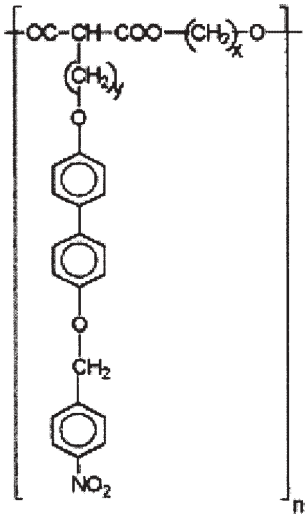
The difference between the standard case A and case B, studied by us, is the symmetry of the system, in case A the planar geometry is anisotropic the wave vector direction is selected by the boundary conditions, compared to case B that satisfies the isotropy condition and the direction of the wave vector of the striped is chosen accidentally at threshold, which corresponds to a spontaneous breaking of the rotational symmetry. Also, the phase diagram $U_c = U_c(\nu)$ is similar to the one of the standard model where a conductive regime appears with oblique and normal rolls at low frequencies and a dielectric regime, more difficult to emphasize experimentally for homogenous textures.

Presumably the critical fluctuation associated with the continuous transition from the smectic to the nematic phase are responsable for the negative conductivity anisotropy in the whole nematic range.

The EC instabilities in a homeotropically aligned (no external anisotropy is imposed) nematic with the unusual combination ($\Delta\varepsilon \rangle 0$, $\Delta\sigma \langle 0$) has been anticipated in the early literature [6], the experimental study has started only recently [7].

Experimental part

We have worked with the p–(nitrobenzoloxy)-biphenyl nematic, stable in the temperature range (110 - 94)°C, inside a homeotrop structure with ($\Delta\varepsilon \rangle 0, \Delta\sigma \langle 0$). Below the temperature of 94°C until 75°C this mezomorphous substance is a C type smectic, and furthermore until the temperature of 66.5°C is an F type smectic. This compound has been discovered in 1970 [8] and has the following chemical formula.



yPBNx

where y is the number of methylene groups in the side group spacer, PBN polyester with biphenyl and nitrobenzyl moieties and x number of methylene groups in the main chain spacer.

Different kinds of mezophase are obtained dependent of the temperature [9 - 10].

The working cells are ITO sandwich type with a thickness d=20μm , over which we apply a sinusoidal voltage. The molecular structure of the liquid crystal inside the sandwich plates is visualized with the aid of a polarizing microscope and recorded by a CCD camera connected to a frame grabber card. Images have been digitized with a spatial resolution of 512 x 512 pixels and 256 grayscales. The study is made around the transition temperature (96 – 98)°C, and the temperature can be stabilized with a precision within 0.1°C.

For the case of the homeotropic alignment that we have studied, the molecular director **n** is perpendicular on the support plates and parallel to the electric field **E** (or the Oz axis).

The threshold voltage U_c, a function of the dimensionless frequency $\omega\tau_q$ where $\tau_q = \frac{\varepsilon_\perp}{\sigma_\perp}$ is the charge relaxation time, is represented in figure 1. Below the dimensionless cut frequency $\omega_{cut}\tau_q = 0.56$ only rolls and squares (R+S) type of structures appear, and above this value soft square (SS) and hard squares (HS) structures appear.

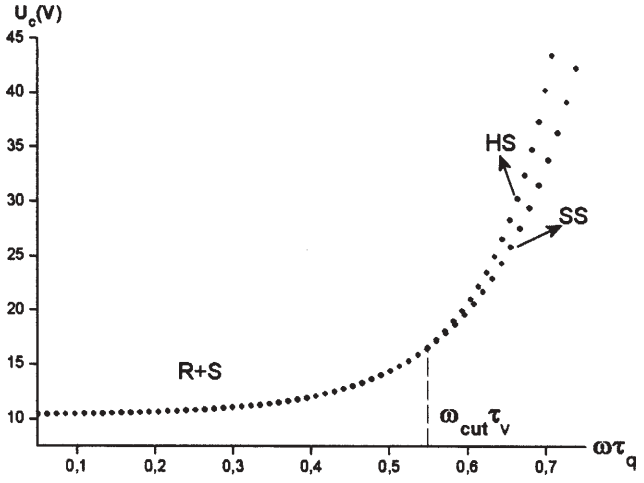


Fig.1. The threshold voltage U_c function on dimensionless frequency $\omega\tau_q$ for a liquid crystal with homeotropic orientation in which the specific structures R + S, SS, and HS appear

Increasing the operating frequency, there appears a new structure formed by areas to regular rolls and overlapping regions with two almost orthogonal roll directions, so called squares structure, (fig. 2a). This structure is disordered overall due to the spatial variation of the molecular director, existing no preferential direction in the plane x-y of the cell due to the homeotropic director anchoring. The size of the overlapping regions increases with the frequency up to a point ω_{cut} with $\omega_{cut}\tau_q = 0.56$. Above this value, only square type structures appear, (fig. 2b). For these structures the undulated character of the rolls is maintained, and the slow spatial variation in the x – y plane, therefore are called soft squares (SS).

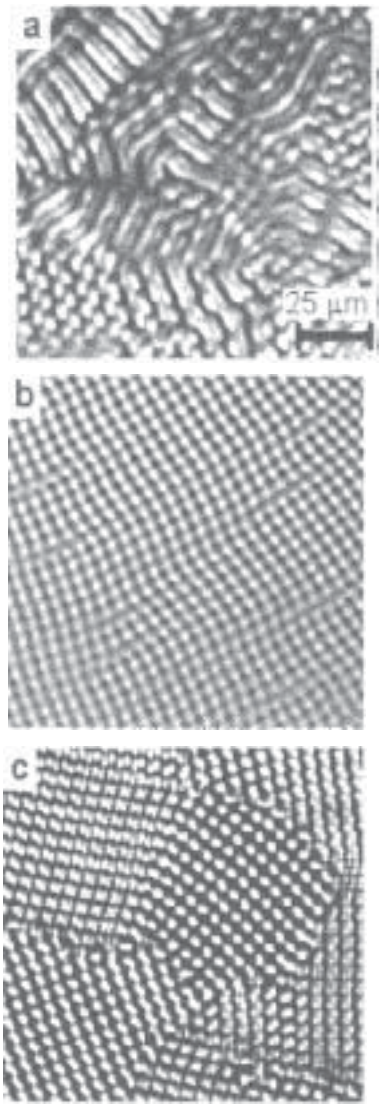


Fig. 2. Spatial structures of the type: a) roll and square (R+S), b) soft squares (SS), c) hard squares (HS)

Further increasing the frequency it can be obtained a new structure which contains domains of well ordered square grids separated by sharp domain boundaries, fig. 2c, called hard squares (HS). This structure is also maintained when grater tensions are applied on the working cell, until the appearance of the spatial-temporal chaos.

Results and discussions

The electrohydrodynamic instabilities of nematic liquid crystals with homeotropic structure can be initially treated inside a linear theory, of Carr – Helfrich type. Within this theory, the Galerkin theory is the most appropriate. Considering a symbolic vector of the type U = U(V, *n*, *v*)

which depends on the electric potential V, on the molecular director *n* and on the speed *v*: then the vector $V(q, z, t)$ is periodic in time t with period and can be expanded in series over the position variable z within a complete set of functions which satisfy the correct rigid boundary conditions $n=z, v=0, V=0$. The periodic time dependence is captured by a Fourier expansion in time and we obtain:

$$U_c^2 = \frac{K_{33}\pi^2 2q'^2}{\varepsilon_0 \varepsilon_\perp S_1} \tag{1}$$

$$S_1 = \frac{1}{1+K_1 q'^2}\left[I_h Q'(q') \frac{\alpha_2 - \alpha_3 q'^2}{\eta_h(q'^2)} - \Delta\varepsilon^{ef}(q') \right]$$

when

$$\Delta\varepsilon^{ef}(q') = q'^2 \Delta\varepsilon' \frac{[\sigma(q')(1+\Delta\sigma') + \omega'^2 \varepsilon(q')(1+\Delta\varepsilon')](1+q'^2)}{\sigma(q'^2) + \omega'^2 \varepsilon(q'^2)}$$

$$\eta_h(q') = \eta_2 + (\eta_1 + \eta_2 + \alpha_1) I_1 q'^{-2} + \eta_1 \lambda_1^4 q'^{-4},\ K_1 = K_{11}/K_{33},\ I_h = 0.97267,$$

$I_h = 0.026036$, $I_1 = 1.24652$, $\lambda_1 = 1.50562$, For details the paper [6] can be consulted.

The material parameters of the liquid crystal that we used p-(nitrobenzyloxi) - biphenyl have the values:

$\varepsilon_\perp = 7.5\varepsilon_0$, $\Delta\varepsilon = 3.9\varepsilon_0$, $\Delta\sigma/\sigma_\perp = -0.65$, $K_{11} = 9.5 \cdot 10^{-12}$ N, $K_{33}/K_{11} = 2.5$ at which we add the viscosity coefficients and the Leslie viscosity coefficients

$\eta_1/|\alpha_2| = 1.06$, $\eta_1/|\alpha_2| = 0.21$, $\alpha_1/|\alpha_2| = -3.5$, $\alpha_3/|\alpha_2| = 0.15$. The coefficient α_1 is possitive and of high value and α_3 is also positive and of a higher value than standard.

We have graphically represented the theoretical relation (1) as function of the dimensionless frequency $\omega\tau_q$ and compared it with the experimental data, (fig. 3). It can be clearly seen that there is a good concordance between the experimental data and the Carr-Helfrich theory.

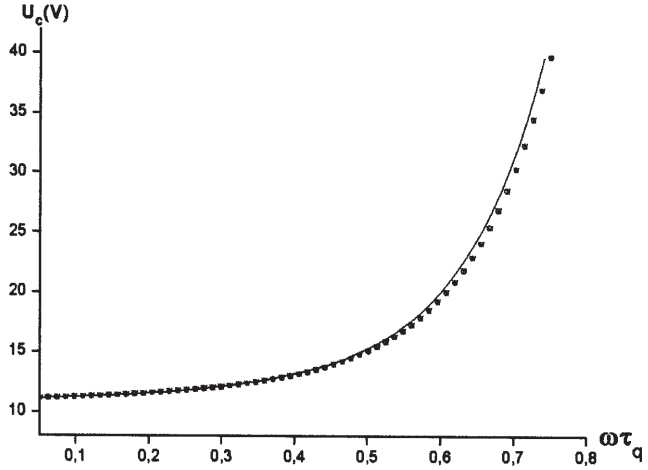


Fig. 3. The critical tension U_c function of the dimensionless frequency $\omega\tau_q$; the points describe the experimental data and the curve the theoretical data

Concerning spatial structures experimentally obtained of roll and square type, soft squares and hard squares, see figure 3, they can no longer be studied theoretically within linear models of Carr-Helfrich type because they are the result of a nonlinear behavior of electrohydrodynamic instabilities.

In the regions of nonlinearity we shall start either from a nonlinear equation for a complex amplitude A or from the following equation set

$$\begin{aligned} \dot{X} &= \chi(Y - X) \\ \dot{Y} &= -ZX + rX - Y \\ \dot{Z} &= XY - bZ \end{aligned} \tag{2}$$

which descried the Lorenz model [11], when χ the Prandtl number and r is the control parameter. Solving these nonlinear equations is easier through numerical simulation [12 - 13].

We have solved the set of equations (2) using a standard pseudo-spectral code on a two-dimensional periodic domain which cohered up to N=30 rolls with wavelength $\lambda_c = 2\pi/q_c$. The resolution was at last 6 grid points per λ_c. Thus, the relevant time scale is set by the horizontal diffusion's time $t_D = N^2 \cdot t_o$. We started with random initial conditions and let the system evolve for a least $5t_D$. It can be noticed that the value of control parameter $\varepsilon(\varepsilon = V^2/V_c^2 - 1)$ is smaller in the simulation than in the experiment (this is bound to the fact that it yields the best contrast of the structure without changing it its behaviour) [14 - 16].

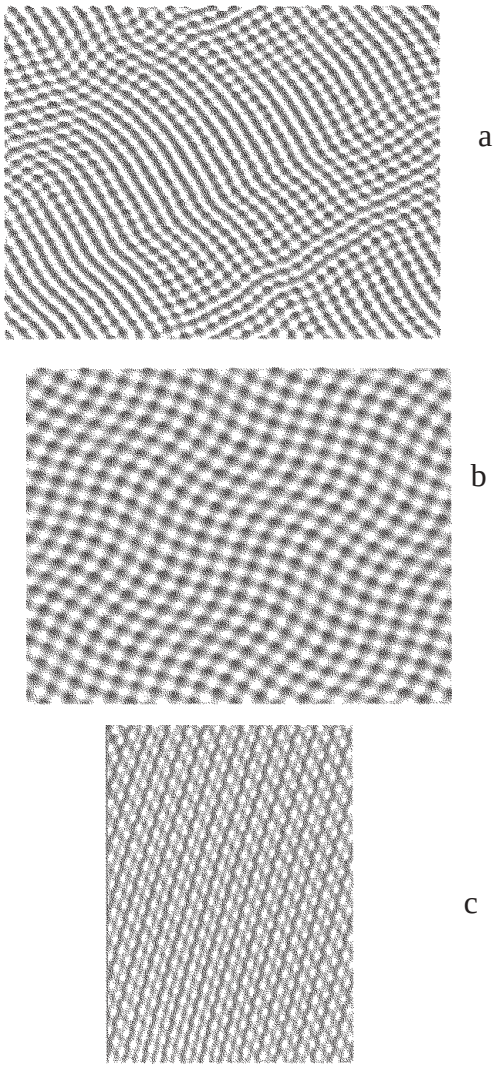


Fig. 4. Spatial structures of the type:
a) roll and squares, b) soft squares and c) hard squares obtained through numerical simulation of the set of equations (3)

With the increase of ω, close to ω_{cut}, for small values of the control parameter ($\varepsilon=0.01$), we obtain a rolls and squares type structure, figure 4a, a structure very similar to the one experimentally obtained, (fig. 2).

In a similar manner, for $\varepsilon=0.022$ and $\omega/\omega_{cut} = 1.07$ a soft squares type of structure is obtained, and for $\varepsilon = 0.81$ a structure of hard squares type. These structures are similar with the experimental ones, (fig. 2b and 2c).

We mention that similar results have also been obtained by A. Buka et. al. [17], through simulation of a Swift - Hohenberg type nonlinear equation.

Interestingly, in simulation the undulations sometimes become completely regular, i.e. the soft squares settle into a static, spatially quasi periodic attractor. It seems that a hard squares structure presents a super lattice structure, which often represents quasi periodic structures, where several groups of wave vectors interact.

Conclusions

Electroconvecting patterns have been observed for nematic liquid crystals with homeotropic alignment and with strongly positive dielectric anisotropy and negative anisotropy of the conductivity.

In the (U, q) space three distinct morphologies, rolls and squares, soft squares and hard squares has been observed. We perform a linear and nonlinear analysis of the full nematohydrodynamic equations and carry out simulations of a suitably constructed Lorenz model. We obtain in agreement with the experiments modulated rolls and squares near the threshold.

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