CONVECTIVE INSTABILITIES IN NEMATIC LIQUID CRYSTALS

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Synopsis

We give a schematic review of some crucial anisotropic properties of nematic materials and we apply it to a comparative study of various convective phenomena near the threshold: instabilities induced by a thermal gradient (Benard–Rayleigh type), electrohydrodynamic effects in the presence of fluctuating electric charges and a new type of hydrodynamic shear-flow instability. Many experimental similarities are found between these phenomena. The application of electric and magnetic fields is shown to control the development of convection. The analogies are understood in terms of a general phenomenological model where the interplay between the time constants for the relaxation of two coupled variables plays the dominant role.

1. Introduction. General properties. Liquid crystals offer the physicist of condensed matter as well as the one of fluid phases a wide open field of research. The existence of strongly anisotropic properties gives rise to many original phenomena or, at least, to dramatic changes, when compared with similar effects in an isotropic fluid¹). Here, we restrict our attention to the simplest mesophase, the nematic one. The physics of cholesterics (or twisted nematics) presents many analogies while the existence of helical structure gives rise to new effects in particular optical ones. The more nearly ordered layered smectics can display solid as well as liquid-like properties which are possibly the most original, but their physics is not yet as developed partly because of the difficulty of obtaining samples of uniform orientation.

A nematic liquid crystal is an anisotropic fluid made of long, rod-like, molecules aligned along one direction, without ordering of their centres of gravity. The directional order is characterized by a unit vector \mathbf{n} , the director field. Above a first-order transition (at a critical temperature $T_c \approx 47$ °C for methoxy-p-n benzilidene butyl anilin, MBBA, used in the experiments described here) a disordered isotropic liquid phase is recovered.

The optical properties of a well-aligned material are those of a positive uniaxial crystal but the birefringence is extremely large $(n_{\rm extra} - n_{\rm ord})/n_{\rm ord} \approx 0.25$. Optical tests of alignment can be made with a very high accuracy if thin enough layers are

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Fig. 1. An elastic energy is involved when a nematic is splayed (b) bent (c) or twisted (d). The undistorted ordered state is schematized in (a).

used (the transparency of liquid crystals, especially in unaligned samples, is limited by the strong light scattering due to the fluctuations of orientation).

In fact, many other properties are anisotropic: the electrical conductivity ($\sigma_a = \sigma_{\parallel} - \sigma_{\perp} > 0$), the thermal conductivity ($k_a = k_{\parallel} - k_{\perp} > 0$), the magnetic susceptibility ($\chi_a = \chi_{\parallel} - \chi_{\perp} > 0$) and the dielectric constant (in MBBA $\epsilon_a = \epsilon_{\parallel} - \epsilon_{\perp} < 0$); \parallel and \perp refer to directions with respect to n). The last two properties tell us that, in a static regime and in the absence of other constraints, molecular ordering can be obtained by the application of fields, the direction n being perpendicular to the applied electric field E, or parallel to the magnetic one E. It is also possible to align a nematic film contained between two parallel plates by inducing the ordering from the boundary conditions by suitable surface treatment. We call "planar" the case where the director axis is in the plane of the limiting plates of a liquid-crystal film and "homeotropic" when it is perpendicular to these plates.

Unlike isotropic fluids, nematics have elastic properties: they will resist torques²). An elastic energy $E_{\rm el}$ is involved if a single domain material is splayed, bent or twisted (the corresponding distortions are shown in fig. 1). $E_{\rm el} \approx K ({\rm d}\theta/{\rm d}x)^2$ where K is a Frank elastic constant; θ the distortion angle of the director along x.

The viscosity is typically that of an isotropic liquid (\approx 0.1 P) but is also anisotropic. Fig. 2 gives the three geometries used in the pioneer Miesowicz³) shearflow experiments, the liquid crystal being aligned by a magnetic field. One obtains $\eta_{\rm B} < \eta_{\rm A} < \eta_{\rm C}$. The hydrodynamic behaviour is well described in terms of two coupled variables, the director n and velocity v^4). For example, in the geometry of fig. 2c, a torque $\Gamma_{\rm c} \propto \partial v/\partial z$ is exerted on the director, due to the shear velocity. In the case of fig. 1b, a torque $\Gamma_{\rm b} \propto \partial v/\partial z \ll \Gamma_{\rm c}$ is also present. If the orientation

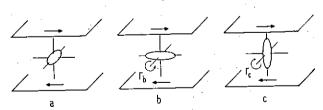


Fig. 2. The three shear-flow geometries. In cases b and c, a torque is exerted on the director. In the case a studied in section (2.3) no torque exists.

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niaxial Optical rers are of the molecules is induced by the effect of the boundaries, the molecular orientation will be modified in the bulk of the film. In section 2.3, we discuss the situation of fig. 2a where no coupling exists between n and v as long as the shear is small enough. The five experimental parameters η_{Δ} , η_{B} , η_{C} , Γ_{b} , Γ_{c} can be used to get the values of the five viscous constants which characterize completely the hydrodynamic behaviour of the nematic.

- 2. Convective effects. We follow the schematic review given above to study some convective phenomena in nematics dealt with in the last few years.
- 2.1. Benard-Rayleigh thermal convection in isotropic fluids. As a well-known leading example of this class of phenomena⁵), we recall briefly the convective effects observed in an infinite horizontal layer of ordinary fluid contained between two solid conducting boundaries heated from below (see fig. 3).

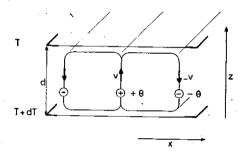


Fig. 3. Thermal convective instabilities in isotropic liquids heated from below. A local temperature fluctuation θ induces a vertical fluid motion at a velocity v. A roll instability occurs above a temperature-difference threshold function of the viscosity, heat conductivity and thickness.

The distance between the plates is typically $d \approx 1$ cm. When the vertical temperature gradient $\beta = \mathrm{d}T/\mathrm{d}z$ is large enough, formation of convective loops is observed. We concern ourselves with the behaviour close to the linear threshold $\beta \geq \beta_{\rm e}$. At the critical point, the production of internal energy due to the upward flow of locally warmer liquid under the influence of the buoyancy force, is large enough to compensate for the viscous losses.

We consider a simplified one-dimensional model where only the effect of variations along the axis 0x perpendicular to the rolls is considered. The effect of the thickness d is expressed by saying that the horizontal wave vector of the periodic disturbance k_x is of order $\approx \pi/d$ as observed experimentally (an exact two-dimensional calculation leads to $k_x = 3.117/d$). The two equations (A.1) and (A.2) of table I can be used to describe the linear-instability problem. The former gives the thermal relaxation of a temperature fluctuation $\theta(x)$ with a thermal diffusivity time constant $T_{\rm th}[T_{\rm th}^{-1} \approx \kappa(2\pi/d)^2]$ where the thermal diffusivity $\kappa = k/\rho C$; k is the thermal conductivity, C the heat capacity]. The latter expresses the evolution of a vertical velocity fluctuation v(x). T_v is the time constant for the

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TABLE I

Retaining only the dominant contributions, one can express the various instability problems, discussed in this review, in terms of coupled equations for two fluctuating variables

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$\frac{\hat{\theta} + (\theta/T_{\rm th}) + \beta v = 0,}{\theta + (\theta/T_{\rm th}) + \beta v = 0}$	(A.1)
$\dot{v} + (v/T_v) + \alpha g\theta,$	(A.2)
$\dot{\theta} + (\theta/T_{\rm th}) + k_{\rm a}\beta\psi_z = 0,$	(B.1)
$\dot{\psi}_z + (\psi_z/T_z) + (\alpha g\theta/\eta) = 0,$	(B.2)
$\dot{n}_z + [n_z/T_z(E)] + Asn_v = 0,$	(C.1)
$\dot{n}_y + (n_y T_y) + Bsn_z = 0,$	(C.2)
$\dot{\psi}_z + [\psi_z/T_z(E)] + (qE/\eta_1) = 0,$	(D.1)
$\dot{q} + (q/\tau_0) + \sigma_{\text{eff}} \psi_z E = 0.$	(D.2)

diffusion of vorticity $[T_v \approx v(2\pi/d)^2]$ where the kinematic viscosity is $v = \eta/\rho$; η is the viscosity]. In the former equation, the effect of an upward velocity in the temperature gradient β is included in a coupling-destabilizing term βv . The buoyancy force also gives an additional coupling term in the velocity equation, $\alpha g\theta$ ($\alpha > 0$ is the thermal-expansion coefficient of the liquid). We use the principle of exchange of stability by stating that, at threshold v and θ are proportional to e^{st} with the real quantity $s = 0^+$. (Just above threshold this would give an exponential increase of the distortion with s strictly positive. In fact as soon as a finite distortion is obtained, the role of nonlinear terms which give rise to saturation effects should be considered.) By requiring that a nontrivial solution be obtained for the two linear homogeneous equations deduced from (A.1) and (A.2), we get the compatibility condition:

$$\beta_{\rm c} = 1/\alpha g T_{\rm v} T_{\rm th} = 16\pi^4 \kappa \nu / \alpha g d^4,$$
(1)
(the exact calculation gives $\beta_{\rm c} = 1704 \ \kappa \nu / \alpha g d^4$).

In the following examples, taking into account only the dominant acting mechanisms, we will be able to reproduce the structure of the coupled equations (A.1) and (A.2) and to obtain a sufficient condition for the development of instabilities in a similar way.

2.2. Thermal convection in a nematic. We consider a "planar" film [director n ($\parallel 0x$) in the plane of the film] (see fig. 4). The directional order is induced by a suitable treatment of the limiting parallel plates. The following experimental results are observed.

a) The threshold β_e is typically 10^2 to 10^3 smaller than the value we would obtain in a similar but isotropic liquid. In particular, the convective effects disappear if the warmer plate is heated strongly enough to obtain the isotropic phase (although

the quantities κ and ν are appreciably smaller in the isotropic state than in the nematic one).

b) The convective loops develop with their axis perpendicular to n and are unobservable if light polarized perpendicular to n is used.

The result (b) suggests that an orientation effect is involved in the convection: in the convective state, n is lifted in the xz plane off the x direction, $n = n_0 + n_z$ ($n_z \ll n_0 = 1$). We will consider here the coupled effects of a temperature fluctuation θ and of the distortion off the x plane characterized by the curvature $\psi(x) = \frac{\partial n_z}{\partial x}$. Eq. B1 again represents the heat equation. Eq. (B.2) describes the relaxation of a fluctuation of orientation $\psi(x)$, of time constant T_z , taking place under the restoring action of the elastic constant (bending is involved in the distortion) in the medium of effective viscosity γ ; $T_z \approx \gamma/K(2\pi/d)^2$. For a film 1 mm thick $T_z \approx 10$ s (this estimate is compared with values of T_v and $T_{\rm th}$ about 10^{-3} to 10^{-5} s for a film of similar thickness).

It is possible to modify considerably the value of T_z by the application of fields (we consider here only the static effect of fields). In a magnetic field H, we have:

$$T_{\pm} = \frac{\dot{T}_z(H=0)}{1 + (H/H_c)^2}.$$
 (2)

The positive sign refers to a "stabilizing" field H_x applied along 0x (which rigidifies the undistorted state) whereas the negative sign describes the destabilizing effect of a vertical field H_z . In fact, we must limit the vertical field value to values $H < H_c$. Above the critical field value $H_c = (\pi/d)(K/\chi_a)^{\frac{1}{2}}$ (called the Freedericksz critical field") the planar structure becomes spontaneously distorted in the absence of thermal gradients. The divergence of T_- as $H \to H_c$ is characteristic of the second-order phase transition and can be analyzed accurately in terms of the Landau mean-field model"). Similarly, a vertical electric field will decrease the time constant T_z .

Coming back to eqs. B, we analyze the coupling terms, The upward flow of a locally warmer portion of the fluid induces a destabilizing torque (similar to Γ_c in fig. 1c) which tends to increase the distortion. But the positive thermal-conductivity anisotropy in the presence of curvature, causes a "heat focusing" effect. Heat is accumulated over the regions where the curvature is concave downwards $(\mathrm{d}\psi/\mathrm{d}x) < 0$) and where an initial positive thermal fluctuation was present. The focusing is expressed by the term $\beta k_a \psi$ and offers the main destabilizing effect. The hydrodynamic torque due to the drag velocity under the effect of the buoyancy force is written as $\alpha g\theta/\eta$ in (B.2). The solution of eqs. (B.1) and (B.2) is carried out exactly as in the previous problem. The main difference with case (A) is the replacement of T_v in (A.2) by a much larger value T_z in (B.2). The threshold is consequently largely reduced. This result is well verified by the experiments⁸).

2.2.1. Remarks. As pointed by Dubois-Violette⁹), convection can be obtained by heating from *above* planar nematics of negative anisotropic heat conductivity $(k_a < 0)$. The materials we have studied up to now satisfy the inequality $k_a > 0$ and do not give rise to this effect.

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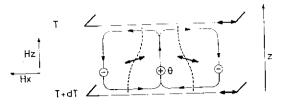


Fig. 4. In nematics, the gradient of the velocity fluctuation v induces a destabilizing torque on the director (situation of fig. 2c). Due to the thermal-conductivity anisotropy, an initial temperature fluctuation is reinforced. This is the dominant instability mechanism. Stabilizing $-H_x$ (destabilizing $-H_x$) magnetic fields increase (decrease) the threshold value. The double arrow lines indicate the direction of the director and the broken line the direction of heat flow.

In an homogeneous homeotropic nematic (*n* perpendicular to the limiting plates), convection also occurs when heating from *above*¹⁰). This experimental fact may be understood by using a reasoning similar to the planar case. However, the main destabilizing torques acting on the director are due to the horizontal component of the velocities and a two-dimensional analysis in needed.

2.3. Hydrodynamic instabilities. We consider the geometry of fig. 2a. In the presence of small enough shear rates¹¹), the orientation of n_0 , perpendicular to the flow lines and the velocity gradient, remains undistorted. The viscosity is that of a classical fluid. However, above a critical threshold $s = dv_x/dz > s_c$, this state becomes unstable and distorts. The mechanism is described schematically in fig. 5. We assume a small fluctuation of the director from n_0 to $n_1 = n_0 + n_z$ ($|n_z| \ll |n_0|$); the shear flow creates a torque Γ_z (cf. situation of fig. 2c) which tends to displace the extremity of n_1 along the flow lines and to bring n_1 to $n_2 = n_1 + n_y$ ($|n_y| \ll |n_0|$).

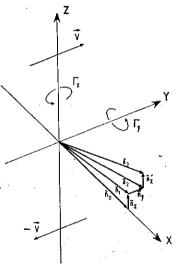


Fig. 5. Mechanism for the development of an instability in the case of fig. 2a. An initial fluctuation n_z is further amplified to n_z' via an n_y distortion.

Let us look at the projection of n in the yz plane: we are in the situation of fig. 1b. The torque Γ_y moves n_2 towards $n_3 = n_2 + n_2$, thus increasing n_z , and reinforcing the initial fluctuation. In a finite-thickness sample, n is anchored at both limiting plates; the distortion is not uniform and requires some elastic energy: the instability occurs when the destabilizing hydrodynamic torque exceeds the elastic one. Expressed in these terms, the problem is analogous to the distortion of a nematic under the destabilizing influence of a perpendicular field, mentioned above (Freedericsz problem). We have shown experimentally that, in the presence of a d.c. shear, and in the absence of applied fields, an homogeneous distortion can be obtained above a second-order threshold shear rate.

We concern ourselves with the distortion in the presence of an alternating shear characterized by a displacement $D \propto D_0 \sin{(2\pi t/T)}$. A convective roll instability develops if the amplitude D_0 is large enough and if the frequency T^{-1} is sufficiently small. The rolls are formed with their axis parallel to the velocity, and perpendicular to n_0 . Also, as in the thermal case, they are not observable in light polarized perpendicular to n_1 ; but here both distortions n_1 and n_2 play a role in the instability. In the shear, a fluctuation of orientation associated with a curvature $\partial n_1/\partial x \propto k_x n_y$ in the horizontal plane can be shown to give rise to a "hydrodynamic focusing" effect where a vertical component of the velocity appears. This component induces a destabilizing torque on n_2 similar to the effect of the buoyancy velocity $v_2(x)$ in (B). Both the effect of this term and of the homogeneous one described by fig. 4 are included phenomenologically in a coupling torque Asn_y in eq. (C.1) for the relaxation of a vertical distortion n_2 . Similarly the horizontal component n_y obeys a relaxation equation, where the presence of a vertical fluctuation n_z is included in a destabilizing torque Bsn_z . The time constant T_y is of the order of magnitude of T_z .

Fig. 6 gives an experimental instability-threshold curve. The average velocity given by D_0/T is kept constant and a vertical electric field E is applied. The part to the left of the solid curve shows the domain of existence of the roll instability (for a given electric field, increasing the frequency will be unfavourable for the development of the instability). The cusp-shaped curve is associated with two different types of instabilities: for low electric fields and close to the threshold (point E_1), one observes that n_z remains constant whereas n_y changes sign over each half period. For fields above the cusp value (point E_2) the instability involves only the oscillation of n_z around zero. As the electric field is increased, the ratio T_y/T_z decreases $(T_y$ is independent of E whereas T_z is a decreasing function of field). The lower branch of the instability curve corresponds to a ratio $T_y/T_z < 1$ and the upper one to $T_v/T_z > 1$ and only the fastest relaxing component oscillates appreciably. Keeping the frequency constant, the instability disappears above an upper value E_3 . By decreasing the time constant T_z , we have increased the threshold value [see formula (1)]. The behaviour is consistent with the analysis based on the solution of the phenomenological coupled equations (C.1) and (C.2). In particular, the cusp value corresponds to the condition $T_y = T_z$. In a linear-analysis treatment, no instability can take place when the two time constants become equal. In order to have a low instability threshold, we must have at the same time T_yT_z as well as T_y-T_z assuming large values.



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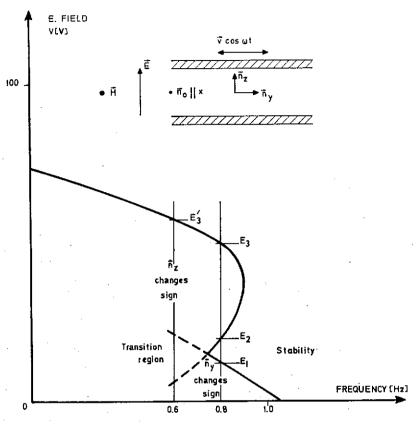


Fig. 6. Experimental threshold curve in the shear-flow instability problem. Sample thickness $d = 210 \,\mu$, average shear velocity $D/T = 0.148 \,\mathrm{cm/s}$.

- 2.3.1. Remark. Linear instability in laminar flow is not usually encountered in isotropic fluids in particular in the case of planar Couette flow considered here. The anisotropic behaviour of the nematics has given rise to original effects.
- 2.4. Electrohydrodynamic instability. If an oriented nematic¹²) is contained between the parallel plates of a capacitor (transparent conducting indium-oxide films sputtered on glass slides) a distorted state is obtained in the presence of large enough d.c. or a.c. electric fields. In particular, a strongly turbulent state (dynamic scattering) obtained in large fields is used in display applications, as the transparency is strongly reduced in this state. Some analogies can be drawn with the electrohydrodynamic effects in poorly conducting isotropic fluids¹³) in the presence of electric charges injected by the electrodes, but the class of phenomena observed is much richer in the l.c. case and has stimulated considerable interest in recent years. We consider schematically the mechanism for a planar MBBA film (fig. 7).

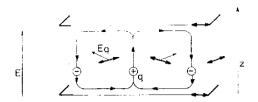


Fig. 7. Mechanism for the electrohydrodynamic instabilities. The motion of charges q induces hydrodynamic torques. The local electric field E_q also tends to distort the planar order. The effect of the anisotropic electrical conductivity reinforces the initial charge fluctuation.

A local positive fluctuating charge is moved upwards under the influence of the applied field. The velocity gradients give rise to torques on the director. The electric field created by the charge tends also to change the orientation of the director. When the dielectric-conductivity anisotropy is negative the torque acts in the same direction as the hydrodynamic one. When the electrical conductivity is of positive anisotropy, an electrical-focusing effect takes place which increases the initial fluctuating charge. The instability is again described by two coupled equations involving the fluctuation of the vertical curvature ψ and of the charge q. The viscosity η_1 takes into account both the hydrodynamic and local electric-field destabilizing effects. In the second equation, the dielectric relaxation time τ_q = $\varepsilon_{\parallel}/\sigma_{\parallel}$ and the effective conductivity $\sigma_{\rm eff}$ involved in the "electrical focusing" terms are functions of both conductivities and dielectric constants.

Fig. 8 gives the experimental instability-threshold curve obtained by Galerne on MBBA14) together with theoretical calculations for a sine- and square-wave electricfield excitation¹⁵). The lower left part of the curve corresponds to the "conducting" regime. In this regime, the distortion remains essentially fixed and the charge oscillates around zero with the electric field (as E changes sign the focusing effect changes sign). Above the cusp field, a "dielectric" regime occurs where the charges are essentially fixed but where the distortion oscillates. Here again, the two regimes

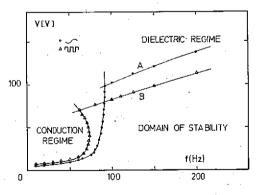


Fig. 8. Calculated (solid lines) and experimental dependence of the frequency threshold versus voltage in an a.c. electrohydrodynamic instability for MBBA. Some analogies can be drawn with the hydrodynamic threshold in fig. 6 (from ref. 14).

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threshold versus n be drawn with can be understood qualitatively from a comparison of the values of the time constants T_z and τ_q , as in the previous problem, and the results offer striking analogies. However, the electric field not only modifies the value of the ratio of the time constant, but also couples ψ_z and q. The reader should consult the detailed review articles for a more detailed analysis as well as for the description of the characteristic structures of the loops (electric-field dependence of the wave vector and orientation). In particular, decreasing the wavelength of the loops results in a strong reduction of time constant T_z .

3. Conclusions. We have presented schematically some basic properties of well ordered nematic fluids, omitting the complementary subject of defects (disinclinations) in these materials. We have applied this description to a comparative study of linear-instability problems in films, and have shown how mesophases vastly extend the range of effects met in isotropic fluids, in particular because the ordering properties of the films can be modified by external fields. We have also obtained a relatively universal description of the instabilities as due to the interplay, in each case, of two fluctuating coupled variables: the coupling terms are proportional to the value of the external field which creates the instability. The crucial parameters, which characterize the obtained modes, are the values of the relaxation time constants of the two variables. Clearly, the physics of mesophases based on relatively well established grounds (possibly apart from the more complex and ordered smectic phases) is wide open for developments and the crucial position of the subject matter halfway between the crystalline solid and the fluid state should attract the interest of physicists of statistical mechanics.

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* Delivered † The Russi Physico-Techn not hold any p University, Ist