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Hydrodynamic transitions to chaos in the convection of an anisotropic fluid

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Résumé. — Nous présentons la première suite ordonnée d'instabilités hydrodynamiques d'un écoulement convectif, observées depuis l'état de repos jusqu'au chaos dans un fluide anisotrope (un cristal liquide) soumis à un champ électrique. La symétrie des structures convectives décroît lorsque le paramètre de contrôle croît progressivement. L'étude des lignes de courant permet d'interpréter la séquence par la compréhension de l'évolution de l'écoulement. On montre que la première étape importante correspond à l'apparition d'un second mode de rotation dans une instabilité de pincement des rouleaux. Nous proposons une description simple des étapes essentielles vers le chaos, qui pourrait être étendue à d'autres systèmes convectifs.

Abstract. — We present the first spontaneous sequence of hydrodynamic instabilities due to the evolution of the convective flow from the rest state toward chaos in an anisotropic fluid : a nematic liquid crystal subjected to an AC electric field. The resulting structures are of decreasing symmetry as the control parameter is gradually increased. The study of the streamlines allows the interpretation of the entire sequence by the evolution of the convective flow up to the onset of chaos. We show that the first important step in the flow evolution is the build up of a second mode of rotation which is associated with a pinching instability of the vortex lines. A simple picture is then proposed in order to describe the essential steps of a possible route to chaos, and which could be extended to other convective systems.

A hydrodynamic system subjected to an increasing external constraint may exhibit a fixed number of bifurcations before reaching a chaotic state. A typical example is thermal convection in isotropic fluids where there exist sequences of time dependent states between the rest state and chaos [1, 2]. On the other hand spatial structures of decreasing symmetry are observed, depending on both the Rayleigh number (the constraint or control parameter related to the temperature gradient) and the Prandtl number [3-6]. However these experiments are usually performed in containers of relatively small lateral dimensions, and the structures are either final states or transient states obtained after some perturbation is applied with a fixed wave vector. For instance, R. Krishnamurti reported [3] the spontaneous evolution of convective flow when steps of thermal gradients were applied for long times. Krishnamurti observed that only two stable spatial structures would appear : firstly the rolls and secondly for a higher gradient, the rectangular cells. The latter structure was in fact obtained after a transient disordered state. Above the rectangles another disordered state was interpreted as a chaotic state. In such an experiment the evolution between the ordered states was left unexplained. In particular, one may pose the question of the mechanism which produces an ordered structure (the rectangular cells) from a transient disordered state. An important feature of Rayleigh-Bénard convection is that, due to the invariance under rotation around the vertical axis, the convective structure has no defined orientation in space within the plane of the layer. Therefore in extended containers the convective structure often appears disordered, i.e. it includes defects. Ordered structures become possible in relatively small containers, and this is due to a stabilizing effect induced by the lateral boundaries [7, 8].

In every case a better description of the evolution of a convective system towards chaos must include, in addition to the analysis of the different spatial structures, the description of the streamlines of the flows and of their evolution.

We present in this work an experimental study of the evolution of the convective flow from the rest state to the onset of the chaotic state, using an anisotropic fluid (a nematic liquid crystal). We show the first ordered sequence of spontaneous convective structures as the control parameter is increased, in an extended container and in the absence of defects. The streamlines are described in every structure and their evolution supports a simple picture of hydrodynamic route to a chaotic state.

A nematic liquid crystal is a fluid with an orientational ordering (denoted by a director **n**) and is characterized by a coupling between that ordering and the velocity gradients which are present [9]. The molecular orientation is controlled by elastic and viscous torques, and is sensitive to external fields. For instance in a nematic layer subjected to an electric field normal to it, a periodic modulation of the molecular axis can be produced which is then coupled to a convective flow [10]. The first convection occurs as rolls having an orientation perpendicular to the molecular axis direction **n**. Therefore in this case, there is no need for solid lateral boundaries, and convective structures can then be studied in extended containers with extremely large aspect ratios (up to 10^4). Provided the defects are carefully avoided, we will show that when the control parameter (here the voltage V) is slowly increased new instabilities appear. They lead to stable spatial structures [11, 13]. The main step in the evolution to disorder occurs under the form of a pinching instability of the convective rolls. Possible mechanisms will be briefly presented that would describe the structures. Chaos in our fluid corresponds to a disordering of the orientation, and we shall show that it is associated with topological singularities in the orientation field.

Up to the present time, many attempts have been reported that such an anisotropic fluid has been used under an electric field in order to look for and study the possible structures before chaos [12]. However the observed sequences were either incomplete or included irrelevant structures. No clear classification could be made, and the study of the evolution of the streamlines was omitted. The only reliable observation was that, as in Rayleigh-Bénard convection, two structures (rolls and rectangles) were often created between the rest state and the chaos. In fact, experimental data or accurate observation concerning the convection amplitude (velocity or molecular orientation) were difficult to obtain because of the presence of a large density of defects. The density could be very large even in small containers of aspect ratio Γ of order 10 (Γ is the ratio of a lateral dimension L_x or L_y over the thickness $L_z = d$). We have been able, by properly choosing the experimental conditions, to make samples with large aspect ratio up to 500 or 1 000 and with a low density of defects. This density is typically less than one per mm^2 for $d = 50$ μm, i.e. the mean distance between two defects is higher than 20 roll diameters [13]. In some experiments the density can be lowered by a factor of five.

Let us give, at first, a simple picture of a chaotic motion, and a possible way to reach it. In the chaotic state the flow field may be, at some time, considered as a superposition of elementary rotations randomly distributed in space and fluctuating in time. As the control parameter is increased, the time and space scales decrease. Let us consider here only the time-independent states, i.e. we restrict our picture of chaos to a spatially disordered superposition of elementary rotations. In a typical convection experiment the first structure is a set of parallel rolls with a preferred alignment, say along **y** (Fig. 1). The next step will add, to the first « mode », a second « mode » of rotation around **x**. Next, a third « mode » around **z** will be added. These modes can be sketched as slices of the former rolls, which afterwards have uniformly been re-oriented. The resulting structures will be ordered in space. Disordering may be caused by localized perturbations that produce defects in the structures. The chaotic state is obtained when these perturbations are, in addition, fluctuating in time. Such a structure would then appear as a randomly moving ensemble of still-ordered domains. As the control parameter increases, the local velocity increases, and the size of the ordered domain decreases. We shall see that this simple scenario can be illustrated at least for the first steps by our sequence of instabilities.

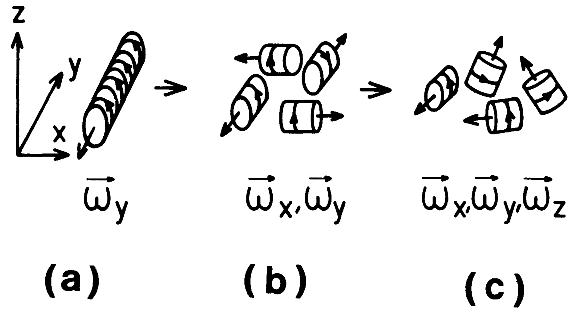


Fig. 1. — A simple model for the evolution from one convective mode to full chaos. a) Convection around **y** (rolls). b) After periodical sectionning of a roll, rotation of elements by $\pi/2$. The result is an ordered structure with two rotation modes (bimodal rectangles). c) Disorganization of the structure by defects. The time dependence may be periodic, the result is a chaos in space and in time.

Our experimental sample is a thin layer (50 μm) of a nematic compound either MBBA or Merck Phase V (an eutectic mixture of azoxy compounds of negative dielectric anisotropy). The layer is sandwiched between two glass plates (2 × 3 cm) coated with semi-transparent electrodes of indium oxides (Fig. 2). A proper coating with a polyimid (Kerimid from Thomson CSF-LCR, Orsay) rubbed in one direction ensures a well-fixed alignment of the molecular axis parallel to the plates, say along **x** (Fig. 2). An A.C. electric field **E** is applied normal to the layer along **z**. The observations and measurements are made under a polarizing microscope and on negative films. A microdensitometer is used for wave vector measu-

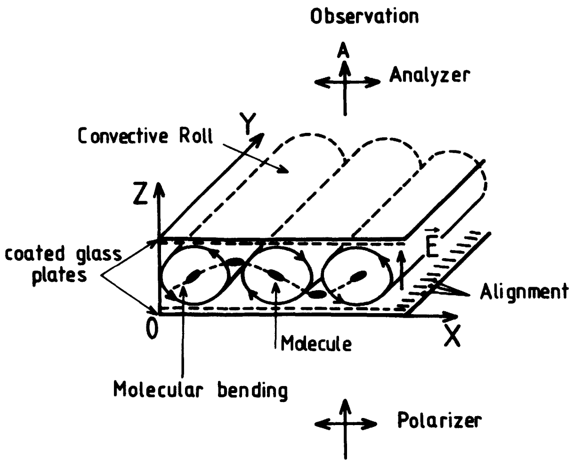


Fig. 2. — Experimental set-up. The nematic liquid crystal is sandwiched between glass plates. The molecular bending is maximum on the axis of a convective roll. The convection occurs as rolls normal to the molecular alignment.

rements, and video recording allows to study the dynamics of the structures and of the defects. Optically, such a nematic is a uniaxial positive crystal of axis parallel to the molecular direction **n** along **x**. A deformation of the alignment will modulate the refraction index for an extraordinary polarized incoming light and deviating the optical rays. Therefore one directly sees an image given by these rays (intersection of the caustics with the plane of observation). The flow is visualized directly by immersing a small glass sphere (3-5 μm diameter) inside the fluid. Although accurate values for the velocities cannot be given, the streamlines can be easily visualized. The A.C. electric field is varied by small steps of duration times long compared to the characteristic settling times for each structure. It is delivered by a generator with an amplitude mean-term stability better than 10^{-4} over 30 minutes. A structure is considered to be stable when it lasts for times long compared to the settling time, for instance several hours (more than 4 or 5 hours) compared to some 1 to 4 minutes.

1. The first convective structure (Normal Rolls or « Williams Domains »)

1.1 THE STRUCTURE. — Starting from the rest state in which the nematic layer is optically transparent, one gradually increases the voltage by steps of 0.025 volt every 2 minutes. The frequency of the field is set at 150 Hz. At some well-defined voltage threshold, $V_R = 7$ volts, bright lines appear on a darker background indicating a periodical distortion of the molecular alignment along **x**. These lines are parallel to **y**, i.e. normal to the molecular axis (Fig. 2). Observation of the glass spheres shows a rotation in a plane normal to **y**. In fact, two sets of lines are present, each one focused in a different focal plane. One set corresponds to the up and down planes of flow. The other one images the rotation axis of the rolls. The roll diameter is close to the sample thickness $d = L_z = 50$ μm. At threshold, the diffraction of a laser beam gives only the first order spots which indicate a phase grating, i.e. the index of refraction is sinusoidally modulated. This first structure is now very well known under the name of « Williams Domain » [14]. A theoretical one-dimensional model has been given [10] which allows to determine the voltage threshold assuming a given period for the convection. A two-dimensional model accounts for the wave vector at threshold [15]. Above threshold no model which would predict for instance the voltage dependence of the wave vector has been established. Let us briefly recall the so-called Carr-Helfrich-Orsay (C.H.O.) mechanism [10] of this instability. The nematic has a negative dielectric anisotropy $\varepsilon_a = \varepsilon_{\parallel} - \varepsilon_{\perp} < 0$, a positive conductivity anisotropy $\sigma_a = \sigma_{\parallel} - \sigma_{\perp} > 0$. At first, a voltage accross the sample (**E** field along **z**) is stabilizing for the structure. However, the ionic charges present in the bulk are dragged by the field and the resulting shear flow acts upon the molecular axis. Suppose a bend fluctuation ϕ of the molecular axis with a wave vector **q** along **x**. This distortion will focus the ionic charges in zones of high curvature $\psi = \partial\phi/\partial x$ since, due to $\sigma_a > 0$, the drag is increased. Along **x** a distribution of charges periodic in space is created that will add an extra field component $\mathbf{E}_t$ along **x** acting on the molecule. This action is destabilizing because it tends to reinforce the angular deviation ϕ in the bending. Such an increase in ϕ will in turn increase the drag of charges and therefore the flow (Fig. 3). There are two destabilizing torques : one due to the total local field $(\mathbf{E}+\mathbf{E}_t)$ and another due to the shear flow induced by the drag of charges. They are opposed to the elastic restoring torque and to the viscous dissipation. The threshold is given as a function of the frequency $f = \omega/2\pi$, or of ω, by [10] : $E_{\text{th}}^2 \simeq E_0^2 \left(\dfrac{1+\omega^2\tau^2}{\xi^2-1-\omega^2\tau^2}\right)$, where $E_0^2 \sim AK_{33}q^2$, K_{33} being the bend elastic constant, q the wave vector, $A = A(\varepsilon_{\parallel}, \varepsilon_{\perp})$ and ξ^2 depends on $\varepsilon_{\parallel}$, $\varepsilon_{\perp}$, $\sigma_{\parallel}$, $\sigma_{\perp}$ and on the viscosities. The threshold is obtained by equating these torques. At threshold a bend fluctuation is amplified if its wave vector q_0 is close to π/d. For a driving A.C. field with a frequency $f < 1/\tau \left(= \dfrac{4\sigma_{\parallel}}{\varepsilon_{\perp}}\right)$ (where τ is the charge relaxation time) the mechanism remains valid. The charges have enough time to alternate with the field; so the driving force is constant. This is the so-called « conduction regime ». It has been shown [9, 16] that in such a regime the curvature ψ of the molecular axis is practically constant in time, i.e. the convective flow is also almost constant in time. As the frequency of the field is varied, one obtains a transition line from D.C. to the cut-off frequency f_c given as a function

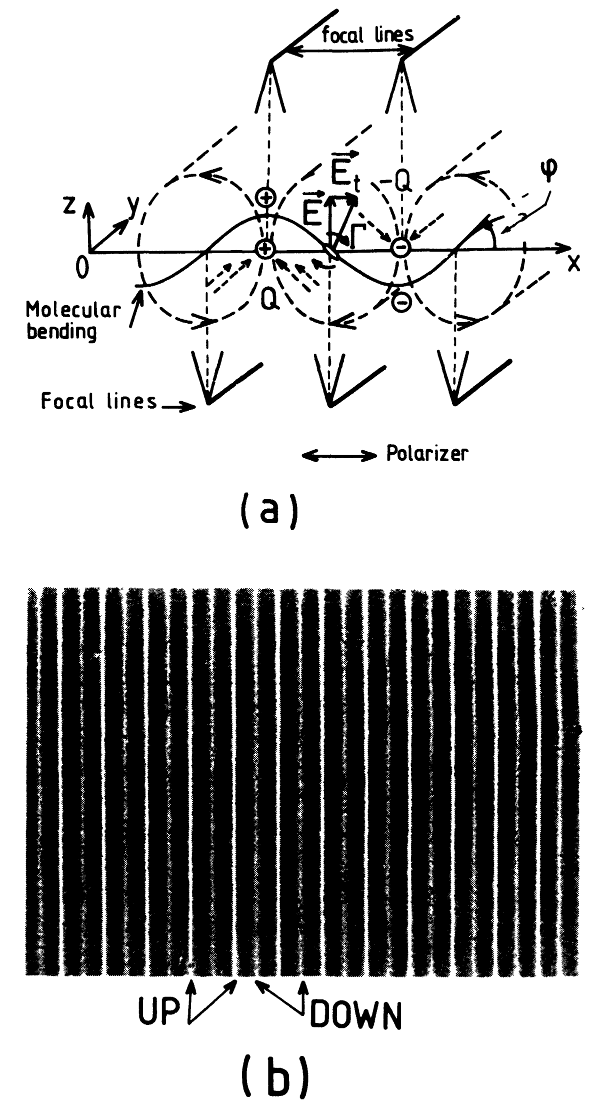


Fig. 3. — a) Mechanism for the instability. The drag of charges in the high curvature zones reinforces the bending. The molecule is tilted by the actions of the viscous torque and of the total local electric field. $\mathbf{E}_t$ is the periodical field produced by the focalization of charges in the bending zones. The transmitted light is focused by the periodical modulation of the index of refraction. The focal lines are in different planes according to the bending angle value.
b) In the picture of the Normal Rolls taken from above the sample and under microscope, only the focal lines for up and down motion are represented.

of the inverse of the relaxation time τ of the charges by $f_c = \dfrac{(\xi^2 - 1)^{1/2}}{2\pi\tau}$.

In fact, as we shall see later, this transition line cannot extend down to D.C. It is indeed replaced by a transition to an other instability at frequencies lower than some well defined value (typically $f \simeq 80$ Hz).

No theoretical model presently exists to account for the evolution of the system above threshold. On the other hand, experiments are rather difficult because of the large density of defects which are rapidly moving and disturbing the structure. For instance Kai and Hirakawa [12] have reported a state which they have named « Fluctuating Williams Domains ». We can explain that their « state » results from the interaction of the large number of defects and at the same time prevents one from observing the new instabilities we report here. A careful preparation and control of the anchoring of molecules allows us to obtain samples with a rather low density of defects. These defects can distort the structure over large distances ($\simeq 20\, d$) and this gives the mean density which can be tolerated. In fact we have been able to obtain samples with less than 1 defect per mm^2 for each of our experiments. (Typically the usual preparation technique would give a density of about 30 defects per mm^2.)

As we further increase the voltage V, the contrast of the focal lines tends to sharpen, and the focal planes for the up and down flows are now at different heights. The focal lines for the axes get closer to the upwards lines. This effect was theoretically analysed [17] and named the « squint » effect. The diffraction of a laser light shows the higher order spots indicating the presence of higher harmonics in the deformation of the alignment (index grating). These are typically non-linear effects, and are obtained close to threshold for $\varepsilon \simeq 0.1$ (where ε is the reduced voltage difference $\varepsilon = (V^2 - V_{th}^2)/V_{th}^2$ and V_{th} is the voltage at a threshold).

Above the threshold, the measurements of the velocity v of the small glass spheres as a function of ε give very close to V_{th} a linear variation, while one would expect a $\varepsilon^{1/2}$ dependence of v for such a direct bifurcation (Fig. 4a). This result would indicate that our critical regime is close to V_{th} within $\varepsilon < 0.1$.

The convection period λ measurements show a slight decrease as ε increases : $\Delta\lambda/\lambda \simeq 2\,\%$ for $\Delta V/V_{th} \sim 5\,\%$ (Fig. 4b). The increase of q with the control parameter, even measured on a very small range, is reproducible, and this result is to be compared to the Rayleigh-Bénard case where the opposite variation has been observed [18].

In our compound the typical cut-off frequency f_c is of order 500 Hz. Close to that limit, new instabilities are observed [13]. Close to D.C., different instabilities are also observed [13] and we shall limit our observations to frequencies higher than those for which the field amplitude varies with a period comparable to the settling time of an instability ($f = 5$ Hz). These new instabilities will be reported in a subsequent paper. For the present discussion, we shall fix the frequency at the intermediate value of 150 Hz.

1.2 THE DEFECTS OF STRUCTURE. — The elementary defect is an extra pair of rolls, and topologically it is a pair of disclinations. We shall name it « edge-dislocation ». Therefore around the core, the rolls are depressed on one side and compressed on the other side of a line normal to the roll axis and passing

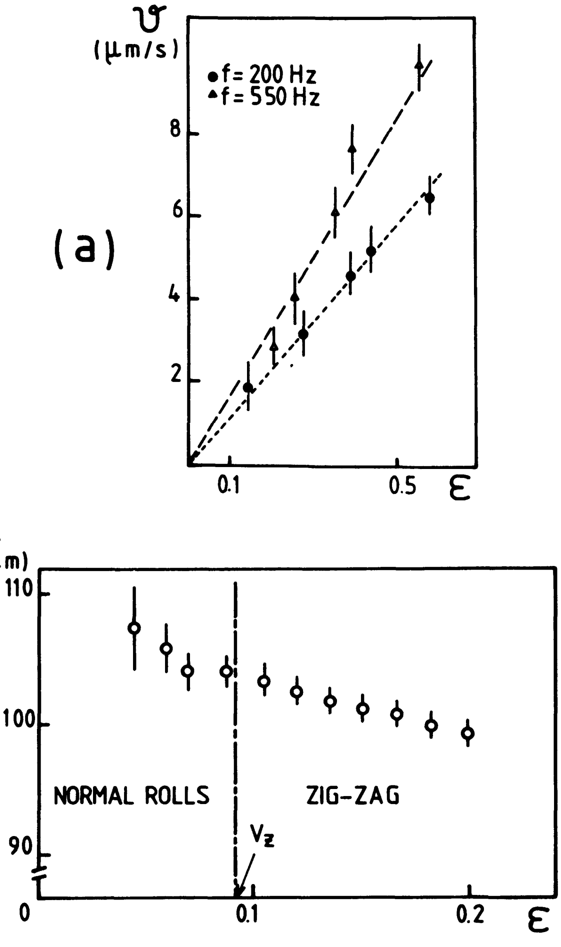


Fig. 4. — a) Velocity of particles immersed in the fluid, as a function of the reduced voltage difference above the threshold V_{th} : $\varepsilon = (V^2 - V_{th}^2)/V_{th}^2$.
b) Spatial period of the convection measured normal to the roll axis as a function of ε. V_z is the transition to the zig-zag structure.

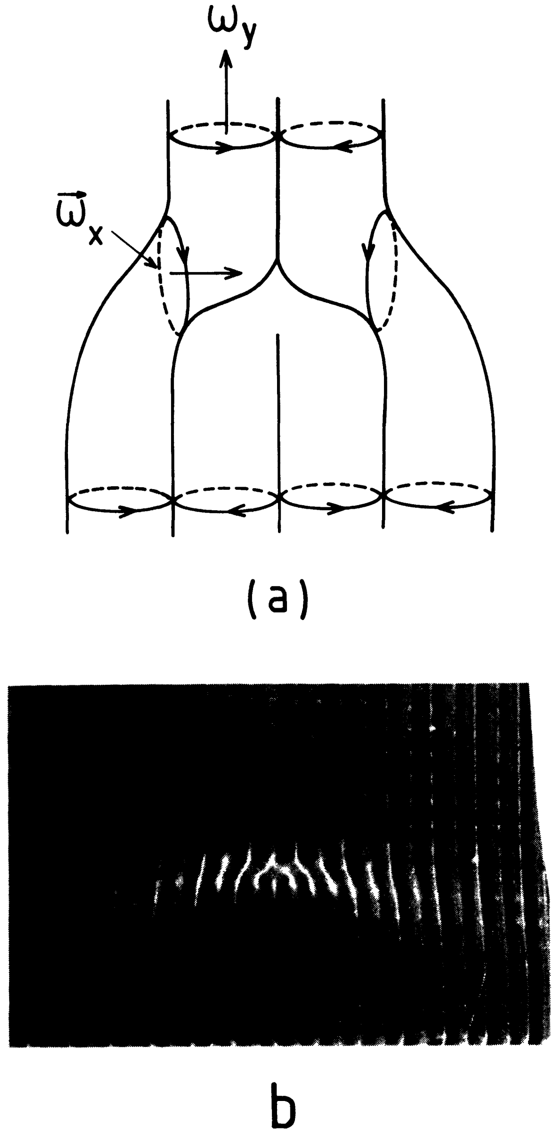


Fig. 5. — a) Schematic of an « edge-dislocation » and of the main streamlines as deduced from the observation. Around the core there exists a new rotation mode ω_x along x decoupled from ω_y.
b) Picture of an « edge dislocation » showing the pinching around the core and the extended deformation field.

through the core (Fig. 5). The deformation field of such a defect can be analysed in terms of a mixed-type elasticity (smectic-like) [13, 19]. At low-frequencies the streamlines show a small zone of rotation ω_x around x (Fig. 5), around the core. At these low frequencies (80 Hz $< f <$ 300 Hz) the dislocation motion is a glide, and the deformation field extends over a large distance. The rolls do not accept « compression » and relax it by a curvature. For higher frequencies ($f >$ 300 Hz) no ω_x component is obvious and correlatively the motion is almost a pure climb. The deformation field is of small lateral extent : the roll can accept compression and the curvature is therefore reduced.

2. The oblique roll structure (zig-zag) [13].

2.1 THE STRUCTURE. — The frequency is set at 150 Hz, and the voltage is further increased. Above a new threshold ($V_z \simeq$ 7.5 volts) the bright straight lines become slightly undulated along their axis y with a period Λ of order 5 to 10 roll diameters. This static deformation is almost sinusoidal and in phase from one roll to the other (Fig. 6a). The amplitude of the undulation increases continuously as the voltage increases. The period Λ also increases but more slowly. The motion of the immersed glass spheres shows streamlines locally normal to the roll axis. After some voltage (7.7 volts) the modulation becomes enriched in harmonics, i.e. the sine shape tends to be replaced by a more angulate deformation. Simultaneously, the period Λ increases more rapidly. Then after a long time (20-30 min) a final shape is reached which

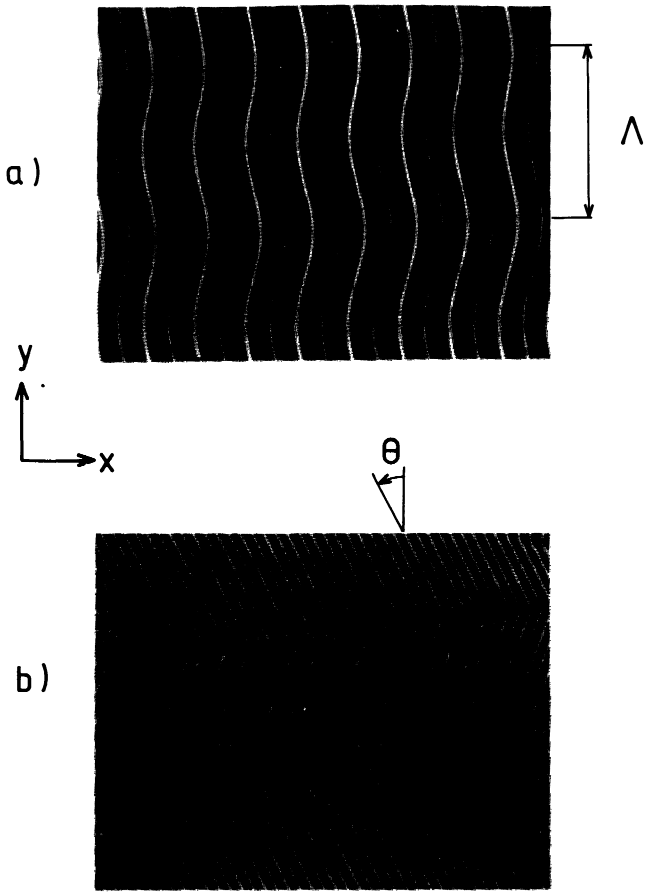


Fig. 6. — a) Undulated roll structure. The deformation is static and appears with a period Λ.
b) The zig-zag structure is obtained for a higher voltage and consists of domains of straight rolls symmetrically tilted over **y**.

is a zig-zag composed of rectilinear rolls tilted symmetrically by θ over **y** (Fig. 6b). The process of change in the shape is a continuous one and does not mean that a structure instability has occurred. In fact, the defects present in the sample tend to pile-up and form grain boundaries at the limits of domains tilted by $+\theta$ and $-\theta$.

These domains can increase their area at a constant voltage as the time evolves, in a process similar to annealing. The final structure is a set of large domains made of rectilinear rolls tilted by $+\theta$ or $-\theta$ over the **y** axis (zig-zag structure). This structure has in fact, already been observed [20] but was never recognized as resulting from a new instability. By building a Landau-like functional with two coupled order parameters, we have been able to describe correctly the evolution of the shape of the zig-zag structure [13, 21].

In the zig-zag structure the rotations occur mainly in plane normal to the roll axis, i.e. tilted by $\pm\theta$ over xOz. The velocity is about twice as larger than in the previous structure, and in addition there exists a very small axial component with opposite sign in neighbouring rolls (the ratio of the tangential *versus* axial velocities if of 10 to 100). The orientation of the molecules inside the bulk cannot be deduced directly from observation in polarized light. This is due to the adiabatic rotation around **z** of the polarization vector of the light along the molecular axis. Adiabatic rotation is effective when $p.\Delta n \gg \lambda_1$, where p is the pitch of the torsion of molecular axis, Δn is the birefringence, λ_1 is the wavelength of the light. In this case a twist of molecular axis around **z** will not be detected by the transmitted light. This condition always seems to be fulfilled in our systems, except maybe inside a boundary layer and for strong deformations due to high voltages. However, it is possible to find the average molecular orientation in the bulk by use of the alignment effect of a magnetic field and by comparing the bulk alignment to the surface alignment [13]. Then we can show that in the bulk the molecules are oriented in a plane close to xOz as in the Normal Rolls structure, and the flow makes an angle θ with the molecules so that the viscous dissipation is increased (page 158 of Ref. [9]) (Fig. 7). A model has been constructed from these observations [21]. It is based upon the fact that above some threshold in the shear, the flow and the molecular axis directions tend to separate. The effect is re-inforced by a lateral focalization of charges along **y**, and the torques due to transverse viscous forces. Therefore at its early stage, the deformation of a roll has a sine shape but must change into a rectilinear roll-structure tilted by a constant angle over the molecular orientation. The transition from the Normal Rolls to the undulated structure is continuous (direct bifurcation) with respect to the tilt angle θ, as a function of the reduced voltage difference ε.

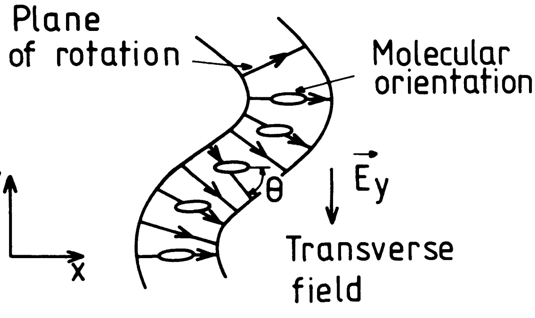


Fig. 7. — In the undulated roll the molecules remain aligned with **x** while the convection plane sinusoidally tilts over **x** along the roll. The curvature produces a new focusing effect for the charges which adds up a new horizontal component for the electric field acting on the molecules, thus reinforcing the angle between the flow and the molecular axis.

When the frequency is varied a transition line can be plotted which meets the transition line of the Normal Rolls structure at a frequency of about 80 Hz. Below 80 Hz one can observe directly a zig-zag structure when increasing the voltage from the rest state. However at threshold the structure does not appear with a sine shape but is rather a full zig-zag with a finite large value of the angle θ over **y** and remains almost constant as the voltage is further increased [28]. The flow in the zig-zag is characterized by a velocity of amplitude v_z (for instance) and of relative orientation θ. At every frequency the transition to the zig-zag is continuous with respect to the velocity amplitude (or equivalently the angle ϕ). However below 80 Hz, θ is of finite value at threshold, while it grows from zero for frequencies above 80 Hz. This suggests an analogy with the phase transitions of thermodynamic systems which are described by two coupled order parameters [22]. Here, the angles ϕ (or v_z) and θ can be the relevant coupled parameters. In this analogy the transition to the zig-zag at low frequencies would be first-order-like while it is second-order-like at high frequencies. Then the intersection at 80 Hz is a triple point with a multicritical character which can also be considered as a Lifshitz point [23], the zig-zag being the modulated phase. The existence of a direct transition from the rest state to a zig-zag is consistent with the earlier observations of Williams [14] (who never reported on Normal Rolls) and explains why it is impossible to obtain Normal Rolls for low frequencies.

2.2 THE DEFECTS IN THE ZIG-ZAG STRUCTURE. — In this case, the defect is also an extra pair of rolls. However its topology is quite new. Around the core it is unsymmetrical with respect to the roll axis but is rather symmetrical with respect to the **y** axis. This is consistent with a deformation in which the flow makes an angle $\pm\,\theta$ with the molecular axis kept along **x**. The deformation field of such a defect is strongly reduced, and these defects act in order to help extending a $\pm\,\theta$ domain at the expense of a $\mp\,\theta$ domain [13].

In conclusion the zig-zag structure is the final conformation of a convective flow resulting from the destabilization of Normal Rolls by a perturbation of the undulation of the wave vector $\mathbf{k}_y$. The flow is still normal to the roll axis i.e. it occurs in a plane tilted by $\pm\,\theta$ to the former one (the Normal Rolls Structure).

3. The periodical pinching (or « skewed varicose »).

3.1 THE STRUCTURE. — As the voltage is further increased, one observes, in one tilted domain of the zig-zag, a new instability arising above a well-defined threshold [11, 13]. At $V \simeq 8$ volts the bright focal lines appear slightly modulated by a smooth unsymmetrical sawtooh-like deformation. This modulation is static, and its amplitude increases continuously with increasing the voltage. The period λ_v of order 2.4 d remains almost constant. From one roll to the next, the deformation shows a phase slippage (the zero phase plane is normal to the roll axis). The result is a periodical pinching of the rolls along their axis (Fig. 8a). The phase plane of this periodic deformation

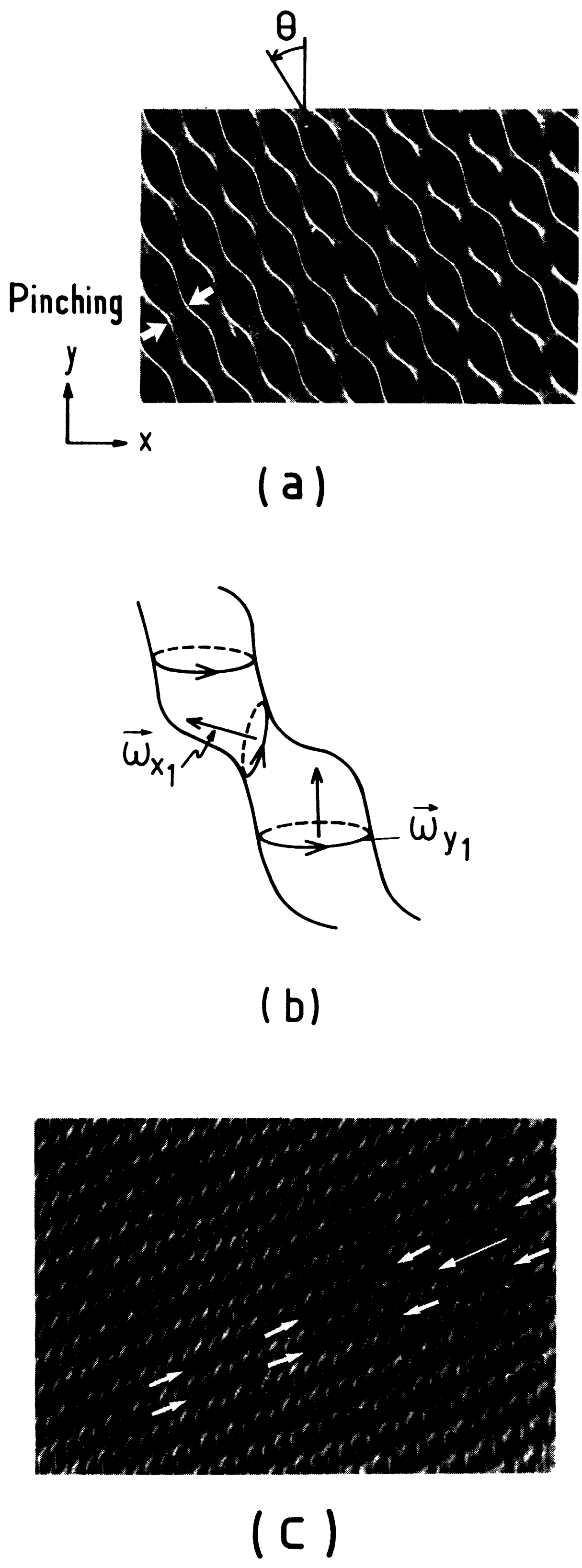


Fig. 8. — a) The periodical pinching produces in a tilted zig-zag domain a structure similar to the « skew-varicose ». b) The streamlines in the pinched zone show a new rotation mode around the oblique $\mathbf{x}_1$ axis, and decoupled from the initial $\boldsymbol{\omega}_{y_1}$ rotation mode (as in a dislocation core).
c) The typical defect in the varicose structure comes from a fault in the modulation period (one more period is added).

is oblique to the roll axis and in a direction $\mathbf{x}_1$ close to $\mathbf{x}$. This structure is quite similar to the so-called « skewed-varicose » observed in Rayleigh-Bénard convection [4, 6]. However, it is a stationary state in our system, contrary to Rayleigh-Bénard convection. The transition from the zig-zag state is continuous (direct bifurcation) [13].

As the voltage is further increased, the pinching increases such that the roll is « split » into cells oriented almost along the roll axis. Then the pinched part tilts, and its direction $\mathbf{x}_1$ gets closer to $\mathbf{x}$. The larger part (the cell) also tilts the same way in order to get closer to $\mathbf{y}$.

The streamlines are obtained from the motion of the glass spheres. We find that (Fig. 8b) :

— in the cells the convection $\boldsymbol{\omega}$ occurs mainly in a plane normal to the axis i.e. around a direction $\mathbf{y}_1$ close to $\mathbf{y}$;

— in the smaller pinched part the rotation is now distributed around a direction $\mathbf{x}_1$ rather close to $\mathbf{x}(\boldsymbol{\omega}_{x_1})$;

— in the curved zones the motion is strongly unstable, a particle would escape either towards the cell or towards the pinched part.

Hence it appears that the pinching acts to build up a new rotation element at $\pi/2$ to the former rotation axis. The first consequence is that in the pinched zone, the convection around $\mathbf{y}_1$ is suppressed in a plane parallel to $\mathbf{x}$ and passing through the inflexion points of the focal lines. Around these points the vertical velocity decreases to $v_z \simeq 0$. Taking $\mathbf{y}_1$ as the axis of the tilted roll in the zig-zag phase, the pinching perturbation may be represented as the sum of two elementary modes of perturbation : one of wave vector $\mathbf{k}_{y_1}$ is an undulation along $\mathbf{y}_1$, the second one $\mathbf{k}_{x_1}$ is a compression-dilation of the roll diameter [6].

In the tilted roll of the zig-zag structure the molecule makes an angle ϕ with respect to $\mathbf{x}$, and the elastic modes involved are a bend mode along $\mathbf{x}$ and a splay mode along $\mathbf{z}$. The velocity direction makes an angle θ with the molecular orientation $\mathbf{n}$. From optical observations we can deduce that in the pinched zones the angle ϕ is decreased. This would imply a mode of twist along $\mathbf{y}$ (thus explaining the oblique direction for the phase).

It is of some importance to notice that the topology around the pinched zone is strongly similar to that around the core of a low-frequency defect in the Normal Rolls structure where a pinching also occurs [24].

3.2 The defects. — Two types of defects can be observed in the periodically pinched structure. One is a trapped edge-dislocation which existed in the zig-zag. The second one is more specific and results from a fault of periodicity in the pinching modulation. It appears as the combination of an extra 1/2 period along $\mathbf{x}$ and an extra 1/2 period along $\mathbf{y}$ (Fig. 8b), when the pinching is total.

4. The bimodal structure (rectangular cells).

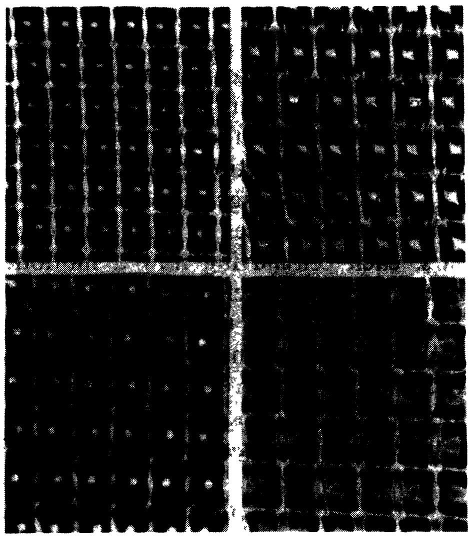
4.1 The structure. — A structure made of rectangular cells has often been observed and some authors have reported on it [12, 25]. However the flow lines and their formation have never been studied. Contrary to the two previous structures, it can be observed without special care in usual samples.

Our study of the defects and their interaction with the roll structure allows us to give an explanation for its easy observation [13, 24]. As we show it, it is indeed usually easier to produce it by rapid growing of large number of defects, in a way very similar to a martensitic transformation [27].

In fact, this structure is obtained here, in the absence of defects, by increasing the pinch of the rolls. At some voltage $V_B \simeq 12$ volts the cells are oriented along $\mathbf{y}$ and the pinched zone is along $\mathbf{x}$. Apparently the rolls are again along $\mathbf{y}$. By looking at the streamlines, one sees that in the middle of the side of the cells along $\mathbf{y}$ there are now nodal points (sources and sinks) where the flow is focused (the rotation mode is $\boldsymbol{\omega}_y$). In addition, the small individual rotations $\boldsymbol{\omega}_x$ obtained in the pinching are now connected together, and a new mode is obtained along $\mathbf{x}$. Therefore, each cell has a rotation $\boldsymbol{\omega}_y$ opposite to that in its near neighbour. Along $\mathbf{x}$ the rotations are also alternate. Then the elementary convective cell is a rectangular cell centred on a nodal point. At each corner is a stagnation point (Fig. 9) which separates each individual closed cell.

This structure is stable and even if it is obtained after the varicose it must not be confused with the quasi-« cellular » structure observed for a high pinching in the varicose where the elementary rotations, $\boldsymbol{\omega}_x$, are disconnected. The rectangles correspond to the sudden connection of these $\boldsymbol{\omega}_x$. Then a new rotation element $\boldsymbol{\omega}_x$ is created and is decoupled from the former one $\boldsymbol{\omega}_y$. The velocities of these two modes are almost equal and are higher than in the first convective structure (200 μm/s compared to 5 μm/s). We shall name that structure « bimodal », and the transition is similar to a first order one (inverse bifurcation). Optically the focal lines which are in fact caustics break up as the rectangles are formed. Careful observation in different focusing planes shows that the caustics give some characteristic singularities : hyperbolic umbilics, in the centres, swallow tails along $\mathbf{x}$, and lips along $\mathbf{y}$. From these caustics it is quite difficult to deduce the topology of the molecular orientation. However they clearly show that the up and down flow layers are no longer continuous but are replaced by localized up and down (sources and sinks) « tubes ».

4.2 Time dependent structure. — Before reaching the stable rectangular structure, a real sample can show oscillations of domains made of tilted rolls (typically of 5 × 5 unit cells) around the $\mathbf{y}$ direction between $-\theta$ and $+\theta$ (Fig. 10a). These oscillations are periodic in time, of period decreasing as V increases. At the



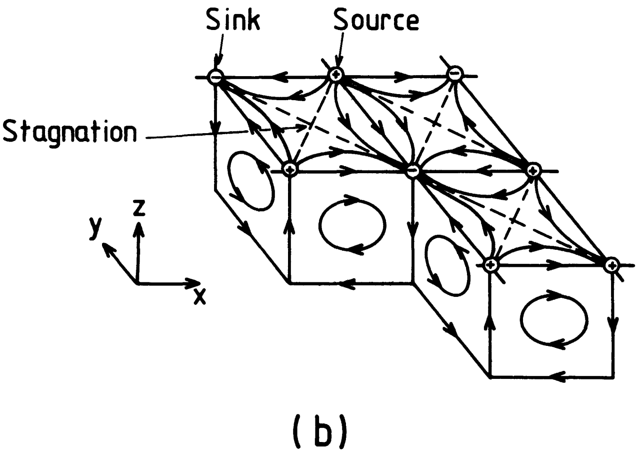
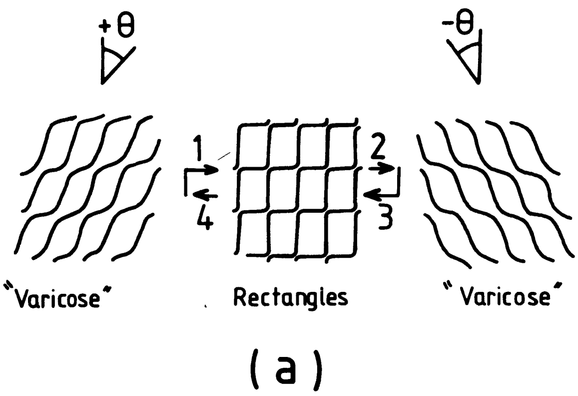


Fig. 9. — a) The bimodal rectangular structure. The observation at different planes shows the singular nature of the caustics produced by the molecular distortion.
b) The streamlines in the rectangles. The convection occurs between singular points (sinks, sources). Stagnation points indicate a zero vertical velocity. The up and down lines no more exist and are replaced by the singular points.



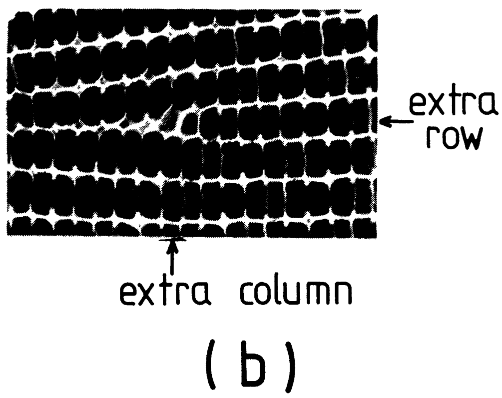


Fig. 10. — a) Oscillations around the bimodal structure. The periodic oscillations occur between the two directions of the zig-zag. During the variation of the angle, the varicose perturbation increases, rectangles are formed then open along the other diagonal and varicose decreases while the rolls are tilted the other way.
b) A defect in the rectangular structure. It is formed from the defect occurring in the modulation of varicose structure (Fig. 8c).

angle θ_{max} the rolls are rectilinear (zig-zag). When θ decreases, the rolls become more and more pinched. For $\theta = 0$ the rectangles set in and obviously the diagonal makes the angle θ with y. Then the oscillation goes on the other way, and the rectangle is unstable. As V is increased, the period decreases, and the size of the oscillating domain also decreases. Finally, at constant voltage, the whole structure stabilizes, after some time (10-30 min), on the bimodal rectangles. Usually the last oscillating domain is clamped around a defect and that duration is necessary for the elimination of the defect. This transient state is actually not understood, but in the presence of a large number of defects it produces a randomly fluctuating structure which can appear well below the rectangular structure. Therefore one may think of a Normal Rolls structure which fluctuates in space and in time [12]. Such a « state » does not allow the identification of the zig-zag and the pinched roll structures.

4.3 THE DEFECTS IN THE RECTANGULAR STRUCTURE. — The defects in the Rectangular Structure consist, as in the pinched structure from where they originate, of an extra half row connected with an extra half column (Fig. 10b). The topology of such defects shows a number of singularities (nodal and stagnation points) with a zero sum. Defects consisting of one extra period, either two rows or two columns, have not been encountered and this is probably due to the necessity of having a larger core area, in this case.

4.4 REMARKS ON THE NATURE OF THE TRANSITION TO THE BIMODAL. — When two distinct structures co-exist in a sample, i.e. rectangles and varicose, there is a clear sharp boundary separating them. On the other hand, as already mentioned, the optical pattern changes abruptly at the transition. At last, the oscillations are quite indicative of some relaxation oscillations which can occur around an inverse bifurcation. These qualitative facts are in favour of a transition varicose-rectangular structure of quasi-first order (or inverse bifurcation).

5. The dynamic scattering mode 1 (DSM 1).

In this section, following the terminology of reference [12], we will distinguish two different regimes, DSM 1 and DSM 2, and we will try to characterize them more accurately. At about 20 volts, the rectangular structure once again starts oscillating between $-\theta$ and $+\theta$, around y, in a way similar to the one previously described in § 4.2. However, here, the area of the oscillating domains is much smaller (3×3 unit cells), and the dephasing in time is almost total between the domains. These domains are made of elongated cells having some appearance of portions of rolls. In fact, a careful observation shows that the sharp caustics are now tending to smear out. After rotating the polarizer by $\pi/2$ one observes, inside these cells, loops of singular lines (Fig. 11a). These lines are thick ($\simeq$ 3-5 μm) and similar to disclination walls [9]. They have a rather distorted shape. Thus the flow no longer has simple symmetry elements, and at this point the symmetry with respect to the median plane xOy is lost. For a slightly increasing voltage these loops rotate in the cell around their mean direction. This rotation rapidly increases, and the average size decreases simultaneously. The appearance of coherent oscillations disappears, each cell having now a motion totally independent from its neighbour. The correlation length of the structure is now reduced to the order of one cell. Incoming light is strongly scattered i.e. the diffraction spots observed in the former structures are totally blurred and fluctuating in intensity in the Fourier space. The average frequency of the fluctuations is of order 10 Hz.

As the voltage increases, the diffracted light intensity concentrates on crescents centred around the spot of the directly transmitted beam. This means that the spatial disorder is stronger.

The singular lines indicate that the orientational order shows a large gradient due to a high shear flow. The structure is no longer ordered in the usual sense (over distances large compared to the characteristic length d), and there is now a rotation component around z. In fact it is the first step in which the ordering is lost simultaneously in space and in time. However the individual motions are still occurring on a scale comparable to d, the sample thickness.

a

b

Fig. 11. — a) The singular lines (disclination loops) of the molecular orientation are characteristic of the disordered structure (DSM 1) occurring above the rectangular one.
b) The full chaotic structure reveals a higher density of smaller disclination loops moving randomly with a large mean velocity (DSM 2).

6. The dynamic scattering mode 2 (DSM 2).

At about 35-40 volts where the elements of the previous structure decrease in average size, a new structure appears nucleated at some spots in the sample. It scatters the light more strongly so that it appears in transmitted light as more opaque than the background [26]. At the same time the average apparent frequency of the fluctuations is at least one order of magnitude higher ($\simeq$ 100 Hz). This structure extends in space with a very sharp front-edge which has a higher velocity in the y direction. The typical velocity is of 100-200 μm/s. The structure is in fact made of small disclination loops of diameter about 2-5 μm (Fig. 11b) and of small thickness. Each loop moves in an erratic way around a fixed position. Decreasing suddenly the voltage to zero shows that these loops are thin nematic wires indicating singularities on distances comparable to some 10 to 100 molecular lengths [9]. For high voltage (60 V) the loops are not longer visible under the microscope and the diffraction pattern of light appears as a diffuse ring from the diameter of which we deduce an average correlation length for the nematic orientation of 1 to 3 μm. The characteristic time associated to the fluctuations is measured by the mean of a photon correlation technique and is found of order 10 ms which is to be compared to the damping time of thermal fluctuations of the molecular axis (of order 1 ms).

We therefore refer to this regime as the chaotic state since there is no longer visible ordering of convection on relatively small space scales (the light wavelength) and the temporal fluctuations have times comparable to that of thermal fluctuations of the nematic ordering. Thus we may conclude that this is a regime similar to the fully turbulent regime of isotropic fluids. In fact there is also no longer orientational ordering on scales comparable to a light wavelength scale (1 μm) so that the nematic ordering could be considered as destroyed, on that scale.

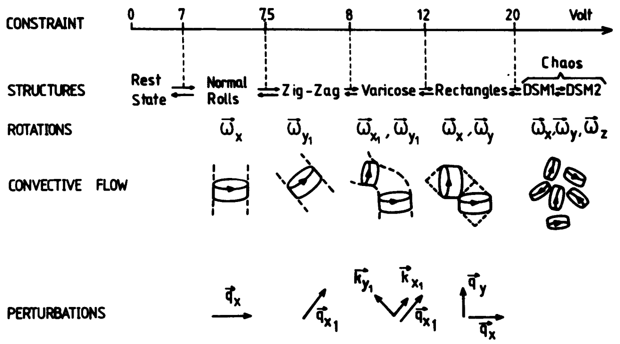


Fig. 12. — The full sequence from the rest-state to chaotic state. The **k** are the perturbation wave vectors while the **q** are the wave vectors of the structures.

The whole sequence is fully reversible, i.e. every ordered structure is reproduced when gradually decreasing the voltage, without noticeable hysteresis and provided that the voltage is smoothly decreased (steps of less than 20 mV per minute).

7. Conclusion.

Using an anisotropic fluid we have experimentally demonstrated the first coherent sequence of structures between the rest state and full chaos. The sequence is composed of ordered structures of decreasing symmetry and is fully reversible. The streamlines of the flows have been studied, and their evolution shows that the first main step in the disordering process is associated to a pinching instability of the convective roll. A new mode of rotation is added at $\pi/2$ to the first one. The resulting structure is bimodal, and it consists of an array of closed convective cells. The next step comes from singularities in the orientation that are due to exceedingly large values of the velocity gradients. The temporal disordering comes first from relaxation-like oscillations around what is believed to be an inverse bifurcation. The presence of defects destroys the spatial coherence of this relaxation-like oscillating structure.

The experimental observations seem to illustrate and substantiate our simple picture of transitions to a chaotic state. In fact, they show that an anisotropic fluid, even if driven to convection by a specific mechanism, might be a useful system for the study of the evolution of the flows. The nematic ordering fixes the orientation of the structures in the plane of the layer by the action of the elastic and the specific nematic viscous torques. Each of the structures involves different nematic deformation modes which enables us to consider that the orientational ordering acts as a selection mechanism for the structures. In addition, optical observations are made much easier than in isotropic fluids. It is believed that the main mechanism of a new rotation mode associated to a pinching of a vortex is not specific to anisotropic fluids and could occur in usual fluids where vortices are developed before the turbulence appears.

Acknowledgments.

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