Localized, Time-Dependent State in the Convection of a Nematic Liquid Crystal

A. Joets and R. Ribotta
Laboratoire de Physique des Solides, Université de Paris Sud, 91405 Orsay Cédex, France

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We report on the experimental study of a novel time-dependent localized state in the electroconvection of a nematic liquid crystal inside a container of very large aspect ratio. At threshold the convective rolls propagate inside isolated domains well localized in space. The phase propagation is attributed to a slow lateral drift of the ionic charges, while the localization, which is not due to finite-size effects, could result from the nonlinear focusing of a broad band of dispersive modes.

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The convection of a nematic liquid-crystal layer under either an electric field[1] or a thermal gradient offers simple examples of stationary states. It was recently discovered that, as a function of an increasing ac electric field, inside the so-called conduction regime,[1] a unique succession of ordered stationary structures precedes a chaotic state with a complicated time dependence.[2] It is therefore relevant to consider the possibility for the system to develop simple time-dependent states, even in a linear regime.

In this Letter we present a first study of a localized, time-dependent, convective state in a laterally extended layer of nematic liquid crystal under an ac transverse electric field inside the upper part of the conduction regime. In many respects its features are similar to those of the traveling waves found in binary mixtures.[3] We find that the localized domains are not induced by the lateral boundaries, and we suggest that they might result from a modulational instability.

The experimental setup is the now classical one: A nematic layer is sandwiched between two glass plates coated with semitransparent electrodes. The nematic material, either N-*p*-methoxybenzylidine-*p*-butylaniline (MBBA) or Merck Phase V, has a negative dielectric anisotropy ϵ_a and the molecular alignment is made parallel to the plates (along **x**). The lateral dimensions of the cell are $L_x = 2.5$ cm and $L_y = 1.5$ cm. The thickness of the layer is $L_z = 50$ μm, so that the aspect ratios are $r_x = 500$ and $r_y = 300$. The temperature is kept constant within 0.2 °C around 20 °C. An ac voltage V is applied across the layer and this constraint is measured by the dimensionless parameter $\epsilon = (V^2 - V_{\text{th}}^2)/V_{\text{th}}^2$, where V_{th} is the voltage at threshold. For each experiment, the frequency f is kept fixed at some value, while V is varied by increments $\Delta V = 0.025$ V. Because of the coupling between the velocity gradients and the molecular orientation, any convective flow periodic in space induces a periodic modulation of the optical axis of the crystal. Extraordinary transmitted light is periodically focalized.[4] This optical pattern is analyzed under a polarizing microscope by digital image processing. The fluid motion is directly determined by our tracing the trajectories of small glass spheres (3–5 μm in diameter) immersed in the fluid. We have recently shown[5] that the transition to a homogeneous stationary state of normal rolls (with axes perpendicular to **x**) is suppressed at low frequencies. In this Letter we show that it bifurcates also at higher frequencies (below the cutoff frequency f_c of the conduction regime) and is replaced by a new time-dependent state.

The frequency f is fixed at a value close to f_c: $f = 0.8 f_c = 600$ Hz (here $f_c \simeq 750$ Hz). The voltage V is smoothly increased up to a threshold $V_{\text{th}} = 15$ V where the instability begins to grow continuously (direct bifurcation), and a pattern appears which is inhomogeneous in space. It consists of isolated domains (Fig. 1) of small extensions l_x, l_y in the **x** and **y** directions, broadly elliptical ($l_x > l_y$), inside which a periodic structure (period $\lambda \simeq 60$ μm) of rolls aligned along **y** translates uniformly in the **x** direction with a velocity **v**. Typically $l_x \simeq 4d$ and $l_y \simeq 3d$, where d is the roll diameter. In fact, it is the phase, i.e., the relative position of the rolls along **x**, which propagates. These domains are, most often, randomly distributed in space. No deformation of the

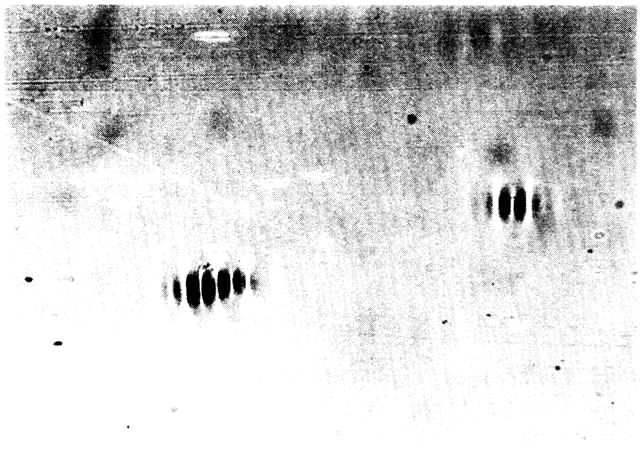

FIG. 1. Photograph of localized domains of traveling rolls, which do not appear here around the small solid particles.

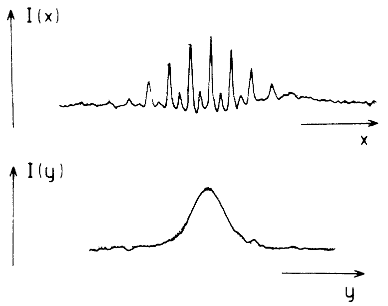

FIG. 2. Optical density profiles fitted to a soliton profile sech^2 (dotted lines): $I(x)$ is along the direction normal to the rolls ($\mathbf{x}$), $I(y)$ along a roll ($\mathbf{y}$).

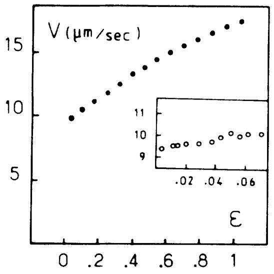

FIG. 4. Velocity of propagation vs reduced voltage. Inset: Velocity is finite close to threshold.

molecular alignment is detected in the space separating the domains; there, small-scale convection (rolls) does not occur. The velocity $\mathbf{v}$ has the same amplitude in the different domains, but its sign seems to change randomly from one domain to the next and from one experiment to the next. The lateral extension of the domains remains stable in time for fixed values of the external parameters V and f. In no case has a uniform translational motion of the domain itself (which would indicate a group velocity) been detected within our accuracy (better than 10^{-2} μm/sec). However, small fluctuations in position $\Delta x \simeq 2\lambda$ around an average location may sometimes be observed over a duration $\Delta\tau \simeq 10^3$ sec. When the experiment is repeated, some domains may appear again in the same place, while others appear at random positions in space. The permanent locations can be attributed to local imperfections: either in the molecular anchoring, in the electric potential (because of defects in the conductive coating), or because of the presence of dust particles. The intensity of light transmitted through the layer is directly related to the tilt angle ϕ of the molecules over $\mathbf{x}$ and hence to the local convective velocity gradient: $I(x) \sim (\partial\phi/\partial x)^2$, in the limit of low velocities and small ϕ. Typical profiles of I recorded along $\mathbf{x}$ and $\mathbf{y}$ inside a domain are shown in Fig. 2. The envelope of the profile along $\mathbf{x}$ can easily be fitted by a sech$^2(x/l_1)$ curve which is the typical shape for a soliton. Here $l_1 = 3.6d$ for $\epsilon = 0.2$. A similar fit works as well for the intensity profile along $\mathbf{y}$. We record the intensity profile taken along the $\mathbf{x}$ axis at a fixed height y, periodically in time with a period $dt = 0.12$ sec. We obtain a space-time diagram, from which the velocity v can be easily measured (Fig. 3). At threshold, v is found to be finite with a typical value around 10 μm/sec ($\simeq 0.16$ λ/sec), while the amplitude of the instability increases continuously from zero. The velocity v increases continuously with the applied constraint ϵ (Fig. 4). Simultaneously, the domains extend in both directions, maintaining a similar shape, and connect up completely above $\epsilon \simeq 1.0$. When a connection zone between two domains is perpendicular to the rolls, one obtains locally a "zipper state," if the velocities of the two domains are of opposite signs. There, dislocation pairs along $\mathbf{x}$ are created and annihilated periodically in time by the motion. When the connection zone is along the rolls axis, one obtains either a "source state," or a "sink state," if the velocities of the two domains are of opposite sign. The source (sink) is when the rolls move away from (towards) the connection border. The source and the sink, which are topological singular lines, may also exist inside a single domain. For $\epsilon > 1.0$, the velocity rapidly falls to zero and one recovers a quasistationary and homogeneous state. The velocity v measured at threshold decreases continuously with the frequency to reach zero at $f_1 \simeq 530$ Hz (Fig. 5). Simul-

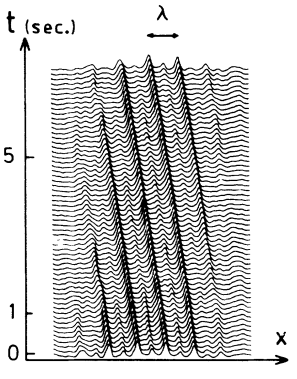

FIG. 3. Time dependence of an optical density profile along $\mathbf{x}$ showing motion to the left. The period between two successive profiles is 0.12 sec. The wavelength is $\lambda = 2d = 60$ μm and the domain contains five wavelengths. Here $\epsilon = 0.3$.

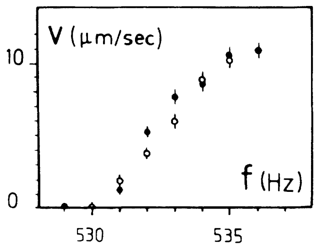

FIG. 5. Propagation velocity v at threshold as a function of the frequency f, showing the transition at $f_1 = 530$ Hz from the stationary to the time-dependent state.

taneously, the size of the domains diverges and the stationary normal rolls are recovered. The limit f_1 which separates the stationary and time-dependent states is a critical point.

Some essential elements for the description of the velocity field are obtained from the direct observation of the tracks of the glass particles (3 to 5 μm) immersed in the fluid. We clearly find two types of trajectories in any vertical (x,z) plane through a domain. In the first one, the particles rotate around an axis parallel to $\mathbf{y}$, which translates at the velocity $\mathbf{v}$ of the traveling rolls. The trajectory is then a cycloid. These particles remain confined inside the rolls. The second type of trajectories indicates an open motion, in which the particles move up and down between the rolls and simultaneously translate in the sense opposite to that of the phase of the rolls, with a slightly lower velocity. These trajectories are close to sinusoidal. This open stream usually extends well beyond the domain limits where it develops a large-scale flow of small velocity ($\simeq 1$ μm/sec$\simeq 10^{-2}\lambda$/sec).

The same experiment has also been performed with blocking electrodes (10-μm-thick Mylar sheets covering the electrodes) eliminating the contribution of isotropic instabilities by local charge injection,[6] with no significant change in the reported effect.

The one-dimensional theoretical model[1] is unable to account for any spatiotemporal dependence of the amplitude. However, this model shows that as the driving frequency approaches the cutoff, the relaxation time associated with the molecular orientation becomes comparable to the ionic relaxation time. The result is a loss of efficiency of the focalizing mechanism by the molecular alignment curvature $\partial\phi/\partial x$ and hence a significant increase of the threshold voltage. Another prediction may be made by consideration of all the different times relevant to this problem. These are the charge relaxation time τ, the curvature relaxation time τ_c, the transit time of ions τ_i,[7] and the time τ_f of the convective flow over a roll diameter d. The value of τ_f deduced from the experiment is of order 5 sec. The other numerical values are $\tau_i \simeq 0.16$ sec, $\tau \simeq 4\times 10^{-3}$ sec, $\tau_c \simeq 25\times 10^{-3}$ sec. Therefore, since τ_c and τ are comparable, the curvature is less efficient in maintaining the charge modulation stable in space against any possible lateral drift. Indeed, for a slight lateral shift (along the wave-vector direction $\mathbf{x}$) of the charge distribution and within each half-period of the electric field, the curvature will immediately adapt to the new distribution. This lateral drift of the charge distribution may occur in the presence of a gradient of conductivity $d\sigma/dx$, which contributes to a current density $\delta J = K\delta q E_x$ along $\mathbf{x}$. The E_x component being smaller than the applied field E, the ionic velocity v_i can be roughly estimated to be at least 1 order of magnitude smaller than the component of the velocity along $\mathbf{z}$, and the drift velocity can become of the order of the convective velocity v_c. Then the charge-density distribution takes a form such as $q = q_0 \cos k(x - vt)$, from which one defines a frequency $F = kv/2\pi = \Omega/2\pi$. The molecular orientation ϕ is then composed of a slow component $\phi_0 \cos(\Omega t - kx)$, on which is superimposed the well-known[8] small-amplitude fast oscillation with period $(2f)^{-1}$. The angle ϕ now oscillates around $\mathbf{x}$ with a typical frequency $F \simeq 0.15$ Hz (for $v = 10$ μm/sec and $k = 10^3$ cm^{-1}) which is to be compared with that of the excitation frequency $f = 600$ Hz. Thus the overall pattern moves uniformly as a progressive wave, in the direction of the molecules, along with a charge-density wave.

In the convection of binary mixtures, the traveling rolls often appear also inside domains of limited extent.[9] There, the strong modulation in space of the amplitude has recently been attributed either to wave reflection against the lateral solid boundaries,[10] or to a Benjamin-Feir instability.[11] In our case, the layer has such a large aspect ratio that the former mechanism of reflection is not relevant. It seems obvious that it is an intrinsic mechanism which causes the formation of the localized domains. An important remark is that this effect of strong localization occurs only inside a well-defined range of frequencies close to f_c. It is also found[12] at very low f ($\simeq f_c/20$) and especially in thin samples (5–20 μm) that homogeneous structures may travel with higher velocities [(1–5)λ/sec].

We believe that in the range of frequencies where localization takes place, the dispersive effects in the phase dynamics, as well as the nonlinearities, become important even very close to the onset. Then the fastest disturbance that grows out of the rest state is not defined by a unique wave vector, but rather by a broad band centered around a value close to the critical one. In real space this corresponds to an amplitude modulation (wave packet), as for the Benjamin-Feir instability of water waves.[13] This tendency to localization may be reinforced by the nonlinearities. This type of solution, although not yet fully demonstrated, could arise, as was first suggested by Newell,[14] from a complex Landau-Ginzburg equation, which is known to govern a large class of pattern-forming systems. This effect of localization has been evidenced numerically in the case of thermohaline convection, with a time dependence in the pattern evolution,[15] and has been considered for binary mixtures.[11] We believe that such a nonlinear mechanism of localization involving a modulational instability could account for our findings as well.

We have found in the convection of a nematic liquid crystal a new state which bifurcates continuously from the rest state and is at threshold simultaneously time dependent and well localized in space. As the frequency is increased, one passes through a critical point from a stationary to a time-dependent state. This result makes our extended system a particularly convenient and attractive candidate for further studies around codimension-2 points. In addition, it is now clear that the former theoretical model for the conduction regime, which is no longer valid at high frequencies, must be improved to include a transition to a time-dependent state.

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[7] The times are defined as $\tau_i = L_z/KE$, where L_z is the sample thickness, K the ionic mobility, and E the electric field; $\tau = \epsilon_\parallel/4\pi\sigma_\parallel$, where $\epsilon_\parallel$ and $\sigma_\parallel$ are the components of the dielectric constant and the ionic conductivity along the molecular axis; $\tau_c^{-1} = \eta^{-1}[(-\epsilon_a\epsilon_\perp/4\pi\epsilon_\parallel)E^2 + K_{33}\mathbf{k}^2]$, where K_{33} is the elastic constant for bend, $\mathbf{k}$ the wave vector, ϵ_a and $\epsilon_\perp$ are, respectively, the dielectric anisotropy and component normal to the molecule, and η is a bend viscosity. The numerical values are $L_z = 50\ \mu\text{m}$, $V = 15$ V, $K = 3.5\times10^{-5}\ \text{cm}^2\cdot\text{sec}^{-1}\cdot\text{V}^{-1}$ (Ref. 6), $\sigma_\parallel = 10^2$ esu (corresponds to a conductivity of $10^{-10}\ \Omega^{-1}\cdot\text{cm}^{-1}$), $\eta = 12\times10^{-2}$ P, $K_{33} = 8\times10^{-7}$ dyn, $k = \pi/d = 10^3\ \text{cm}^{-1}$, $\epsilon_\perp = 5.1$, $\epsilon_\parallel = 4.6$, and $E = 10$ esu ($\text{V}\cdot\text{esu}\cdot\text{cm}^{-1}$).

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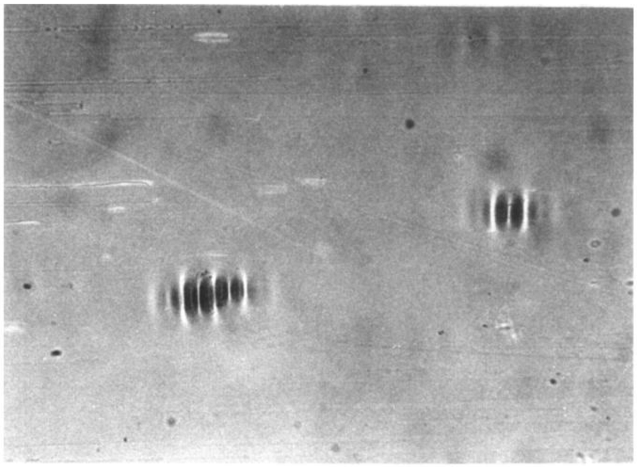

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