Oblique Roll Instability in an Electroconvective Anisotropic Fluid

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We have experimentally discovered that in a nematic liquid crystal subjected to an ac electric field, the first convective structure at low frequencies is in fact a set of oblique rolls, contrary to the accepted picture. We show that it is a new structure with a helical flow motion, and thus lower in symmetry than the usual normal rolls recovered at high frequencies. Besides indicating the limitations of the available theoretical models, these results clearly show that the highest-symmetry flow structure corresponds to the normal rolls.

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In order to study experimentally the disorganization of a convective flow inside an extended layer of fluid, it is preferable to start from an ordered flow with a well-defined wave vector. This means that the ordered structure must have an orientation fixed in space. In fully isotropic convection, i.e., when the fluid is isotropic and when there is no coupling to an aligning external field, the flow usually appears disordered at threshold (e.g., Rayleigh-Bénard[1]). In some cases the system may be rendered anisotropic and the flow oriented along a given direction by the coupling to a magnetic field[2] (magnetohydrodynamic convection). Another possibility is to use an anisotropic fluid, for instance, a nematic liquid crystal—hereafter referred to simply as a nematic. It has recently been found[3] that in a nematic subjected to an increasing ac electric field there exists a complete sequence of prechaotic stationary structures. These structures are ordered and spontaneously oriented with respect to the initial average molecular direction **n** (the anisotropy axis). However, up to now, the essential features of the different flows were not recognized, and more importantly, the identification of the flow of highest symmetry at the first threshold could not be determined.

It is presently well known that a layer of a nematic subjected to a transverse ac electric field undergoes a transition to a convective flow when the voltage reaches some threshold. After the first observation by Williams[4] of an ordered spatial structure, a one-dimensional (1D) electrohydrodynamical model was constructed.[5] In this model, the convective flow is made of parallel rolls oriented perpendicularly to the initial direction of the molecular axis. The frequency of the field is an additional parameter and it was established that the rolls would appear from dc to some cutoff frequency f_c, in the so-called "conduction regime." However it was often experimentally found that the ordering was less effective at low frequencies and indeed it is clear that Williams's observations are not accounted for by the model since his results show tilted domains of parallel rolls.[4] Such a discrepancy was either disregarded[6] or attributed to "inhomogeneities" in the alignment.

In this Letter, we present experimental results which show, in fact, that in a nematic under an ac electric field the convective structure is, at threshold and depending on the frequency, either a set of rolls perpendicular to the molecular axis, or domains of parallel rolls oriented obliquely to this axis, contrary to the widely accepted description. Our purpose is to identify the essential features of each flow in order to determine the highest-symmetry one.

The experimental procedure is the usual one and special care is taken to ensure a correct homogeneous molecular alignment along **x**. The nematic is sandwiched between two glass plates coated with semitransparent electrodes which are rubbed along **x** for planar alignment. The liquid crystal of negative dielectric anisotropy used here is a Merck Phase-V compound. Similar results were also obtained with N-(*p*-methoxybenzylidine)-*p*-butylaniline, but are not reported here. The experiments are started with the frequency set at 60 Hz, well below the cutoff frequency f_c ($\simeq 120$ Hz). At rest the sample is uniformly transparent. The voltage is increased by steps of 25 mV every minute. At $V_r = 14$ V, a static periodic bending of the molecular axis appears along **x** as a set of bright parallel lines on a dark background [Fig. 1(a)]. It corresponds to parallel rolls uniformly oriented perpendicular to **x**. This structure is consistent with the 1D model and was named the "Williams domains"; however, we shall refer to it as the normal-rolls (NR) structure. As the voltage is further increased, the NR structure becomes undulatory along the roll axis **y** at a

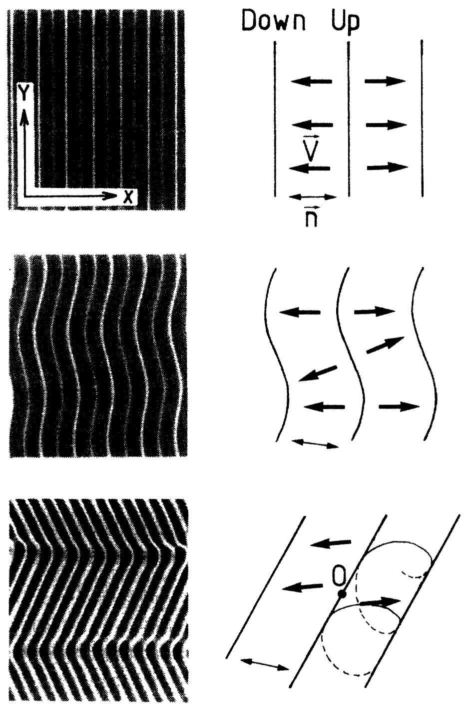

FIG. 1. Plan form of a sample observed under a microscope, and sketch of the velocity field. (a) Normal rolls. The bright lines are focal lines for the up and down flows. The up plane is a symmetry plane. (b) The undulatory rolls. (c) The oblique rolls, with symmetry about a point O.

well-defined voltage $V_{uz} = 15.5$ V. The deformation is static and has a sinusoidal shape with a spatial period $\Lambda = 2\pi/q_y$ of order 5 to 7 times the roll diameter d [Fig. 1(b)]. In order to describe the undulatory roll we shall measure the local tilt angle along **y**, $\theta = \theta_m \sin(q_y y)$. The maximum tilt angle θ_m over **y**, measured at the inflection points, increases with the voltage (Fig. 2), while Λ remains almost constant. Similar to the case of the NR, a control parameter ϵ_z is defined as $\epsilon_z = (V^2 - V_{uz}^2)/V_{uz}^2$ and we have found that $\theta_m \sim \epsilon_z^{0.43}$. Such a behavior is characteristic of a direct bifurcation. At higher voltage, θ_m remains constant while Λ increases sharply. The deformation is no longer sinusoidal but becomes angular while the rolls straighten [Fig. 1(c)]. The increase in Λ is limited by the defects which nucleate more easily and pile up along **y** in order to form grain boundaries. The final stage of evolution of the system is an ensemble of domains of parallel rolls, tilted symmetrically with respect to **y**. We shall refer to this last state as the oblique-roll or zigzag structure. To our knowledge, the undulation instability has never been previously identified. Only the final state (the zigzag) has already been reported[7] as a "modified Williams domain." In order to track the streamlines of the flow we immerse small glass spheres (3–5 μm in diameter) in the

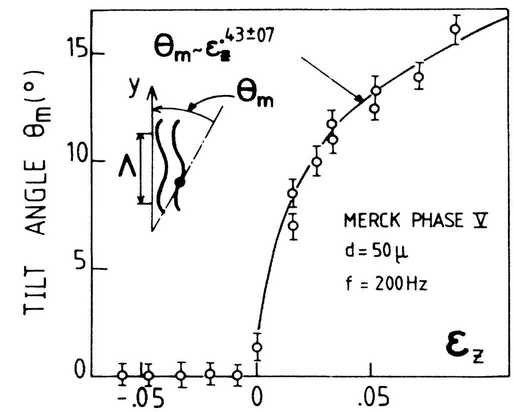

FIG. 2. Maximum tilt angle θ_m of the undulation as a function of $\epsilon_z = V^2 - V_{uz}^2/V_{uz}^2$, where V_{uz} is the voltage threshold value.

nematic. In the NR the convective motion is a pure rotation around the roll axis **y** and has a tangential velocity $v_t \simeq 5$ μm/s at 2% above the threshold. In the oblique roll, there exists, in addition, a small axial component v_a of the velocity, of order $0.1 v_t$. The v_a component changes sign from one roll to the next, and the continuity of the flow along the roll axis is ensured through the grain boundaries.[8] In order to describe completely the state of the nematic, it is necessary to determine accurately the molecular axis orientation inside the flow. While any inclination over the horizontal xOy plane is easily detected by the birefringence effects, the azimuthal tilt out of a vertical plane xOz cannot be measured by a change in the state of polarization of an outgoing light wave. This restriction is due to the Mauguin condition[9] which is always fulfilled in our experiments. Up to now, we have not been able to measure accurately this azimuthal deviation α. However, an estimation from the intensity ratios between the polarized and the depolarized intensities in a diffraction experiment would lead to $\alpha < \theta$. The experiments are repeated at a lower frequency value of 10 Hz. At a threshold $V_{zz} \simeq 7$ V a new instability occurs. The resulting structure consists of domains of parallel straight rolls tilted symmetrically over **y**, by a fixed angle $\theta \simeq 30°$. The motion of the glass spheres indicates an axial component for the velocity, as in the oblique rolls which were obtained at higher frequencies, and for higher voltages. We plot the different thresholds as a function of the frequency f and obtain a structure diagram with a triple point M (Fig. 3). Typically the point M occurs at a frequency $f_M = 30$–40 Hz which increases with the conductivity σ. It decreases by 40% when a stabilizing magnetic field $\mathbf{H} \simeq 5$ kG is applied along **x**. It is clear that the oblique rolls, which are consistent with Williams's observations[4] rather than with the 1D model, correspond to a flow field with helicoidal streamlines, while in the normal rolls the

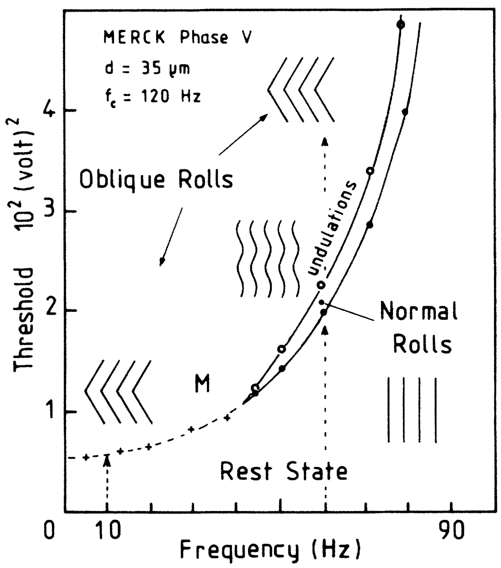

FIG. 3. Structure diagram. Below the triple point M the transition is direct to the oblique roll with a finite angle θ_m ($f=10$ Hz). Beyond M, the normal rolls are met first, followed by the undulatory rolls which then change continuously into the oblique rolls ($f=60$ Hz).

motion is a pure rotation around the roll axis. It is also clear that the symmetry in a vertical mirror of the normal rolls is replaced in the oblique rolls by a symmetry about a point [Figs. 1(a) and 1(c)].

In the following, we would like to suggest some elements and outline some conditions for an anisotropic mechanism of the undulation instability. We start from the stable convective flow of the NR where the molecule is subjected to two simple shears: $s_x = dv_x/dz$ along x and $s_z = dv_z/dx$ along z. In the initial 1D model only s_z was considered. However, we notice that s_x can be destabilizing, i.e., if s_x were alone, a small fluctuation $\delta \mathbf{n}_y$ of $\mathbf{n}$ out of the xOz plane by an azimuthal angle θ could be amplified. Such a condition is similar to that of Pikin's instability,[10] although the initial conditions are different here (the Leslie coefficient α_3 is negative in our experiment). Once the molecular axis deviates from the xOz plane the ionic charges also deviated. If we suppose a fluctuation of splay of $\mathbf{n}$ such that $\delta n_y \sim \delta n_{y0} \sin k_y y$, then the charges are also periodically focused along $\mathbf{y}$. This focusing induces a transverse component E_y for the total electric field $\mathbf{E}_T$ responsible for the dielectric torque which acts on the molecule. This torque adds up to the viscous torque exerted by the drag of charges in the destabilizing process. The result is a static undulation of the rotation axis of the flow, i.e., an undulatory roll. Such a mechanism would be a mere extension of the 1D mechanism.[5] However, here no characteristic length appears along $\mathbf{y}$ in order to impose a spatial period Λ at threshold. A homogeneous solution corresponds to a uniform value for the deviation angle θ along $\mathbf{y}$, in the absence of boundary conditions in this direction. The final state is then a domain of parallel straight rolls at an angle θ with $\mathbf{y}$: the oblique-rolls structure. The misalignment of the molecular axis with respect to the rotation plane implies transverse forces $f_j = \partial_i t_{ij}$, where t_{ij} is the complete viscous-stress tensor. These forces induce transverse components of the flow which create an axial component of the velocity of reversed sign from one roll to the next one. A three-dimensional linear stability analysis has recently been given by Zimmermann and Kramer.[11] This analysis confirms our previous observations regarding the stability domain for the normal rolls, but it does not account either for the undulation instability, or for the essential difference in the flow symmetry between the NR and the oblique rolls.

In conclusion, we find that in laterally extended containers, two intrinsically different structures may occur at the first convective threshold, depending on the frequency. At high frequencies, beyond a triple point it is the usual normal-roll structure consistent with the 1D theoretical model. At low frequencies the normal rolls are not a stable solution as shown by Zimmermann and Kramer and the 1D model is no longer valid here. Then the normal rolls are replaced by oblique rolls rather consistent with Williams's first observations. In the oblique rolls the velocity field is helicoidal. The normal rolls are of highest symmetry and, therefore, a hydrodynamic description of the evolution to the chaotic state must start from this structure. A purely anisotropic mechanism that could be based upon our suggestions remains to be built for the undulation which is the basic instability to the oblique rolls.

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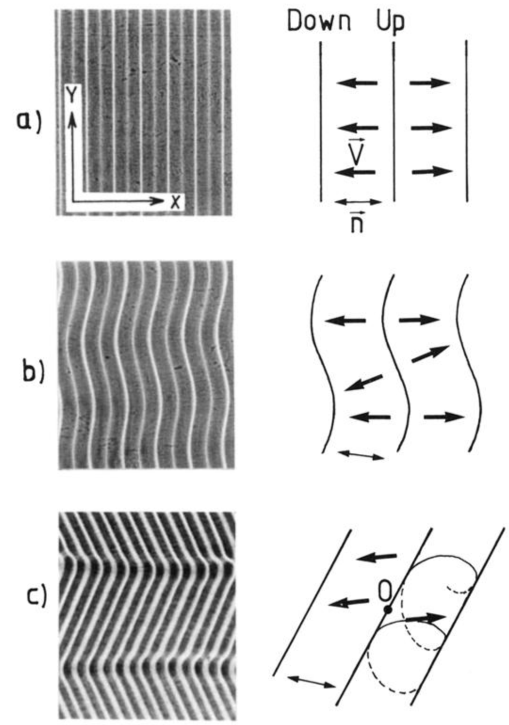

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