Excitability in liquid crystal

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(Received 25 July 1994; accepted for publication 29 July 1994)

The spiral waves observed in a liquid crystal submitted to a vertical electric field and a horizontal rotating magnetic field are explained in the framework of a purely mechanical description of the liquid crystal. The originality of the experiment described in this paper is the presence of the vertical electric field which allows us to analyze the spiral waves in the framework of a weakly nonlinear theory.

The study of excitable spiral waves has been motivated by the problem of cardiac fibrillation. Excitations in the cardiac muscle propagate as wave fronts, and the break of such fronts is at the origin of spirals waves which in turn leads to arrhythmia. Chemical systems exhibit similar phenomena. In the Belousov–Zhabotinsky reaction, for example, chemical oscillations propagate with basically the same properties as the cardiac excitation. Chemical systems have been intensively used to study the properties of excitable waves. One of the main advantages of these systems is their ability to be controlled and their simplicity in comparison with biological systems.

Spiral waves have also been observed in nematic liquid crystals. In this paper, we intend to discuss excitable phenomena in the framework a mechanical theory of liquid crystals.

The experiment (Fig. 1) consists of a nematic liquid crystal sandwiched between two glass plates in a homeotropic geometry (molecules perpendicular to the glass plates) subjected to a magnetic field $H$, parallel to the plates and an electric field perpendicular to them. The magnetic field can either be fixed, or rotating in the $(x,y)$ plane at a frequency $\omega$ around an axis perpendicular to the plate while the electric field $E$ parallel to the $z$ axis oscillates at high frequency in order to avoid convection effects. A motor rotates a plate at a frequency $\omega$ supporting two small permanent Nd–Fe–B magnets, which produce a magnetic field of 0.5 T between the poles. The homeotropic anchoring is achieved by treating our glass plates with lecithin. The thickness, $d$, of the sample was in general about 75 $\mu$m. The liquid crystal used is MBBA, nematic at room temperature with negative dielectric anisotropy. More experimental details can be found in Ref. 8.

The originality of this experiment is in the use of molecules with a negative dielectric anisotropy so that the homeotropic state (vertical alignment of the molecules) can be destabilized by the application of the electrical field. As a consequence, we can observe a great variety of phenomena in the vicinity of the Fredericksz transition. When a stationary horizontal magnetic field is applied to the sample the nematic director tends to align along the field direction. Due to the homeotropic boundary conditions which impose the initial vertical alignment, an elastic restoring torque is present. The balance between these two effects defines the onset of the Fredericksz transition. The order parameter for these experiments is the projection of the director on the $(x,y)$ plane. Above the threshold of the Fredericksz transition, walls which separate domains with opposite orientations in which the director points along the $z$ axis appear. In analogy with ferromagnetic systems, we shall refer to these walls as Ising walls, since the order parameter, the projection of the director on the horizontal plane in our case, vanishes at the center of the wall. To be consistent with this previous definition, walls in which the director never points along the $z$ axis will be referred to as Bloch walls. The effect of the electrical field used in Ref. 8 is to induce an Ising–Bloch (IB) transition of the static domain wall. Such a transition is produced by the electrical field which forces the molecules to reorient toward a plane perpendicular to the $z$ axis. In Fig. 2 we show an experimental measurement of the IB transition for a static domain wall. As yet understood in terms of general argument in Ref. 13 and qualitative argument in Ref. 6, the effect of the rotation is to induce a movement of the Bloch wall. Bloch walls of different chirality may be connected by a vertical line (umbilic line) in which the director points toward the $z$ axis. This configuration naturally evolves into a spiral wave when subjected to a rotating magnetic field. We show in Fig. 3 a photograph of the formation of a spiral wave obtained using the experimental setup described in Fig. 1.

Nematic liquid crystal is a liquid of long molecules which possesses quadrupolar properties (de Gennes, Chandrasekar). In a continuous description, each point of the nematic possesses a privileged direction $n(r)$, which corresponds to the mean orientation of the molecules (n is called the director and $n^2=1$ by definition). In order to describe such a liquid, the usual hydrodynamical equations have to be supplemented by an equation which describes the local orientation of the molecules. A great deal of physics have to do with the dynamics of the director field. Although a generic deformation of the orientation of the molecules always induces a flow, there exist many cases in which this flow follows adiabatically the dynamics of the director and thus can be neglected in the first approximation. This is the case of the experiment described here. Although hydrodynamical effects are present, many of the phenomena observed, as for example the motion of walls, the formation of the spiral waves, can be explained in the framework of a purely mechanical description of the liquid crystal. This approach starts with the Frank free energy of the liquid crystal which takes into account the elastic deformation of the di-
rector field. In the presence of the electric \( \mathbf{E} \) and magnetic field \( \mathbf{H} \), this free energy is given by

\[
F = \frac{1}{2} \int dV \left[ K_1 (\nabla \cdot \mathbf{n})^2 + K_2 \mathbf{n} \cdot \nabla \times \mathbf{n} \right] + K_3 (\mathbf{a} \times \nabla \times \mathbf{n})^2 - \chi_a (\mathbf{H} \cdot \mathbf{n})^2 - \epsilon_a (\mathbf{E} \cdot \mathbf{n})^2,
\]

where \( \mathbf{H} = H_x \mathbf{e}_x + H_y \mathbf{e}_y + H_z \mathbf{e}_z \), \( H_x, H_y, H_z \) are the magnetic field components, \( K_1, K_2, K_3 \) are the elastic constants, \( \chi_a \) is the magnetic susceptibility, and \( \epsilon_a \) is the electric susceptibility. The equation which describes the dynamic of the director \( \mathbf{n} \) is obtained as the balance of the various torques acting on it,

\[
m \times \frac{\partial \mathbf{n}}{\partial t} + \mathbf{v} \times \frac{\partial \mathbf{n}}{\partial t} + \mathbf{n} \times \frac{\partial \mathbf{F}}{\partial \mathbf{n}} = 0,
\]

where \( m \) represents an inertia and \( v \) the viscosity damping. In the case where \( v \) is large enough, the inertial term in Eq. (1) can be neglected. Equation (1) then reads

\[
\mathbf{n} \times \frac{\partial \mathbf{n}}{\partial t} = -\mathbf{n} \times \frac{\partial \Phi}{\partial \mathbf{n}},
\]

where we have introduced a new time variable normalized to the viscous time scale \( \tau = t/v \). One can solve Eq. (2) in the following way:

\[
\frac{\partial \mathbf{n}}{\partial \tau} = -\frac{\partial \Phi}{\partial \mathbf{n}} + \lambda \mathbf{n},
\]

where the arbitrary constant \( \lambda \) is determined by the condition \( \mathbf{n}^2 = 1 \). This gives the actual equation of motion of the director

\[
\frac{\partial \mathbf{n}}{\partial t} = \frac{\partial \mathbf{n}}{\partial \tau} + \lambda \mathbf{n},
\]

\[
\frac{\partial \mathbf{n}}{\partial t} = -\frac{\partial \Phi}{\partial \mathbf{n}} + \lambda \mathbf{n}.
\]

The linear stability analysis of Eq. (4) reveals that when \( E > E_c \), which will be defined later, the homeotropic state \( \mathbf{n} = \mathbf{z} \) becomes unstable. It is then natural to make a vertical Fourier expansion in which we retain only the first unstable mode. It can easily be shown that in the vicinity of the Fredericksz transition the higher order vertical distortion modes are damped and therefore follow adiabatically the first unstable mode. Let us choose for an order parameter, \( A(x,y) = X(x,y) + i Y(x,y) \), which measures the deviation from the homeotropic state, where \( X(x,y) \) and \( Y(x,y) \) are defined by:

\[
A(x,y) = X(x,y) \cos(\pi d z) + Y(x,y) \sin(\pi d z), \quad A = 1 - (n_x^2 + n_y^2)/2, \quad d = \text{constant}.
\]

Then, by calculating \( \mathbf{n} \times \mathbf{n} = \mathbf{z} \) and inserting into Eq. (1), one finds

\[
\frac{\partial A}{\partial t} = -\frac{\partial \Phi}{\partial A} + \lambda A
\]

The linear stability analysis of Eq. (4) reveals that when \( E > E_c \), which will be defined later, the homeotropic state \( n = z \) becomes unstable. It is then natural to make a vertical Fourier expansion in which we retain only the first unstable mode. It can easily be shown that in the vicinity of the Fredericksz transition the higher order vertical distortion modes are damped and therefore follow adiabatically the first unstable mode. Let us choose for an order parameter, \( A(x,y) = X(x,y) + i Y(x,y) \), which measures the deviation from the homeotropic state, where \( X(x,y) \) and \( Y(x,y) \) are defined by:

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Then, by calculating \( \mathbf{n} \times \mathbf{n} = \mathbf{z} \) and inserting into Eq. (1), one finds

\[
\frac{\partial A}{\partial t} = -\frac{\partial \Phi}{\partial A} + \lambda A
\]
This equation is very similar to the one studied in Ref. 13 except for the term $\tilde{A}gK$ which is proportional to the anisotropy of elasticity ($K_1-K_2$).

For simplicity, we first discuss the case $K_1=K_2$. Equation (2) simplifies after the transformation $A=A e^{-i\omega t}$ to

$$\gamma_1 A = (\mu + i \gamma_1 \omega) A + \gamma \tilde{A} + K_1 \nabla^2 A - a |A|^2 A.$$  \hspace{1cm} (6)

Let us take $E$ to be slightly smaller than $E_c = \sqrt{\frac{K_3}{\gamma_1} - \epsilon_n d^2}$. Note that the dielectric anisotropy $\epsilon_n$ is negative. The linear stability analysis of Eq. (3) yields that the homeotropic state $A=0$ loses stability when $H > H_1(\omega, E)$ (Fig. 4).

Let us now place ourselves in the synchronous region (Fig. 4) in which the director rotates at the same frequency as the magnetic field ($\omega < x_e H^2 / 2 \gamma_1$). In this region there exist two stable homogeneous solutions of Eq. (6) which read

$$A = \pm \sqrt{\frac{(\mu + \gamma \cos 2\delta) \omega}{\mu + \gamma \cos 2\delta}} e^{i \delta}$$

where $\delta$ is defined by

$$\sin(2\delta) = \gamma_1 \omega / \gamma.$$  \hspace{1cm} (7)

The Ising wall (static domain), a heteroclinic solution which joins those two solutions, reads

$$A = \sqrt{\frac{(\mu + \gamma \cos 2\delta) \omega}{\mu + \gamma \cos 2\delta}} \tanb \left( \frac{(\mu + \gamma \cos 2\delta) 2K_1 x}{\mu + \gamma \cos 2\delta} \right) e^{i \delta}.$$  \hspace{1cm} (8)

It loses stability entering region C of Fig. 4 toward a moving soliton (Bloch wall) when

$$\omega > \sqrt{\frac{2x_e^2 H^2 + x_e \epsilon_n (E_2^2 - E_1^2) H^2 - \epsilon_n^2 (E_2^2 - E_1^2)^2}{3 \gamma_1}}.$$  \hspace{1cm} (9)

Proceeding as in Ref. 13, we obtained that the velocity $v$ of the Bloch wall is

$$v = \frac{\sqrt{\mu - 3 \gamma^2 - \gamma_1 \omega^2}}{\mu + \gamma \cos(2\delta)}.$$  \hspace{1cm} (10)

Note that when $\gamma_1^2 \omega^2 = \gamma^2$ the velocity cannot be defined since we get into the asynchronous regime in which the size of the wall diverges.

In the more realistic case when $K_1 \neq K_2$, there is no simple transformation that reduces Eq. (5) to a time-independent equation like Eq. (6). The anisotropy of elasticity induces an oscillation at the core of the static domain wall. This oscillation which changes the size of the wall can be understood as a continuous periodic change of splay-bend static domain walls to twist static domain walls. As yet un-
understood qualitatively in Ref. 6, a more dramatic implication of the anisotropy of elasticity is to differentiate the size and the shape of the umbilics of opposite topological charge. As a consequence, in the nonequilibrium case ($\omega \neq 0$) the $+1$ and $-1$ umbilic selects a different wave number. This seems to be a generic feature of a nonequilibrium system for which the wave-number selection mechanism is intimately related to the typical size of the vortices. We have investigated the asymmetry between the defects by placing ourselves in the parameter region corresponding to region C of Fig. 4. Numerical simulations of Eq. (5) shows that a $+1$ umbilic selects a larger (smaller) wave number than a $-1$ umbilic when $K_2 < K_1$ (when $K_2 > K_1$) (see Fig. 5). We now interpret the experimental results concerning the IB transition with a static magnetic field $\omega = 0$, $\mathbf{H} = H \hat{z}$, and $\mathbf{E} = E \hat{z}$. This static IB transition was introduced in Refs. 11 and 12 in the context of ferromagnetic field (play-bend static domain, $K = K_1$, $\xi = x$) or, parallel to the magnetic field (twist wall $K = K_2$, $\xi = y$) read $X = \mu + \gamma \tanh[\xi \sqrt{\mu + \gamma}/2K]$, $Y = 0$.

The play-bend static domain wall loses stability when $E^2 > (a-1)/2 \epsilon e_1 \epsilon_2 \epsilon_3 H^2 - (K_2/\pi^2 \epsilon_3 d^2)$ toward static Bloch wall where

$$\alpha = \frac{K_2 (\sqrt{1+8K_1/K_2}-1)^2}{16K_1 K_2 (\sqrt{1+8K_1/K_2}-1)^2}.$$ 

The stability condition for the twist wall is obtained by interchanging $K_1$ and $K_2$ in the expression for $\alpha$. The positivity of the elastic constant implies that $0 < \alpha < 1$. These predictions (Fig. 2) are in good agreement with the experimental measurement of the Ising–Bloch transition of a splay-bend static domain wall. To end this note we remark that far from the umbilics the phase approximation leads to

$$\gamma_1 \Theta = K_1 \nabla^2 \Theta - \gamma \sin(2\Theta) + \gamma_1 \omega,$$

where $A = Re^{i\omega}$, $R^2 = \mu + K_1 (\nabla \Theta )^2 + \gamma \cos(2\Theta)$, and $K_1 = K_2$. This last equation, used in Ref. 6, gives a good approximation of the dynamics of the Bloch walls but cannot describe the spiral wave due to a phase singularity which defines the center of the spiral.

We have described the dynamics of spiral waves in liquid crystals submitted to a rotating magnetic field in the framework of pure elastic theory. The main difference with the spiral waves observed in chemical and biological systems is due to the existence of domains with opposite value of the order parameter, which lead to the formation of two-armed spiral waves. Slight modifications of the experiment allows one to obtain one-armed spiral waves which are most closely analogous to chemical spiral waves. Basically it amounts to introducing a slight imperfection. A small tilt of the magnetic field from horizontal favors one of the two states of the director and thus induces a transition from the two-armed spirals to the one-armed spirals. A more dramatic is the effect of a slight tilt of the electric field, in the case of a rotating sample, without the magnetic field. In that case only one domain of orientation exists. As in chemical systems, retraction waves eventually become spiral waves when the rotation (excitability parameter in chemical systems) exceeds some critical value.

7 S. Nasuno and S. Kai (private communication).