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Commensurate and Incommensurate Structures in a Nonequilibrium System

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A layer of nematic liquid crystal that has undergone an electrohydrodynamic instability (and contains nonequilibrium structures with period l_0) is subjected to a spatially periodic electric field with periodicity l_1. The competition between these two length scales is found to result in novel ordered phases similar to those found in equilibrium condensed-matter systems. A simple model is proposed that accounts for most of the observations.

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When a periodic structure is modulated by a spatially periodic perturbation having a different length scale, a variety of ordered phases may result: commensurate phases in which the structural periodicity is rationally related to that of the perturbation, incommensurate phases characterized by a regular array of dislocations, and chaotic phases.[1] These structures have been detected experimentally in a number of condensed matter systems, including adsorbed krypton on graphite,[2] magnetically ordered systems[3] such as CeSb, and smectic liquid crystals.[4] The basic phenomenon of interest, competing periodicities, has been the subject of much recent theoretical work.[5]

In this Letter we describe the experimental discovery of commensurate phases and novel two-dimensional incommensurate structures in a *nonequilibrium* system consisting of an oriented nematic liquid-crystal layer containing electrohydrodynamic (Williams) domains. In the presence of a uniform electric field applied transverse to the layer, the director field (local direction of the rod-shaped molecules) becomes spatially periodic with a period l_0 that is roughly twice the layer thickness d. This well-known phenomenon[6] is associated with the formation of microscopic convection rolls similar to those resulting from the Rayleigh-Bénard instability. By applying in addition a *spatially periodic* electric field with periodicity l_1, we find that a variety of ordered states can be produced, including one-dimensional commensurate phase-locked patterns and two-dimensional incommensurate patterns consisting of regular lattices of kinks. This system provides an opportunity to study ordered states resulting from competing periodicities in a situation where the important parameters (such as the ratio l_1/l_0 and the strength of the periodic perturbation) can be more easily varied than is generally the case in condensed matter systems. We also propose a simple theoretical model, based on the minimization of a Liapunov functional, that accounts for the main features of our observations.

The experimental configuration is shown in Fig. 1. A layer of nematic liquid crystal (MBBA) occupies the gap between two transparent conductive In_2O_3 electrodes. In order to impose a spatially periodic voltage across the layer, the lower electrode is photolithographically separated into two interdigitated regions A and B. By maintaining the two regions at different potentials V_A

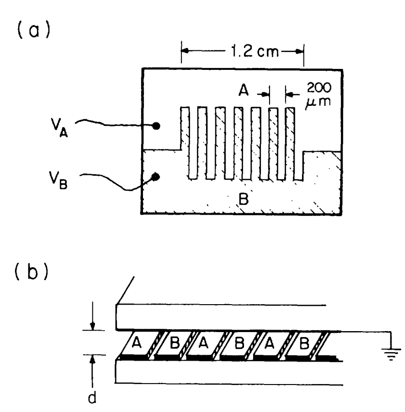

FIG. 1. (a) Schematic diagram of the interdigitated electrode whose 120 fingers provide a spatially periodic electric field. (b) Configuration of the cell.

and V_B with respect to the upper grounded electrode, a spatially periodic electric field (with periodicity $l_1 = 200$ μm) is applied across the layer. (To prevent electrolysis, 50-Hz ac voltages are actually used.)

In the absence of a spatially periodic perturbation, the director orientation is perpendicular to the electrode fingers and in the layer plane. This alignment is achieved by an oblique SiO evaporation onto both plates.[7] As a result, the electrohydrodynamic rolls are parallel to the electrode fingers, so that one might expect the problem to be basically one dimensional.

The patterns are observed by transmission microscopy with use of light polarized in a plane containing the director orientation. The unperturbed Williams domains are visible in the lower left half of Fig. 2(a) as a set of dark lines resulting from light refraction. There are two rolls per period l_0 of the unperturbed structure. Patterns are studied by means of a vidicon camera connected to a digital imaging system. Digitization with a resolution of 320×240 pixels and 8-bit accuracy allows us to compute numerically Fourier spectra of the optical intensity field with resolution better than 1%.

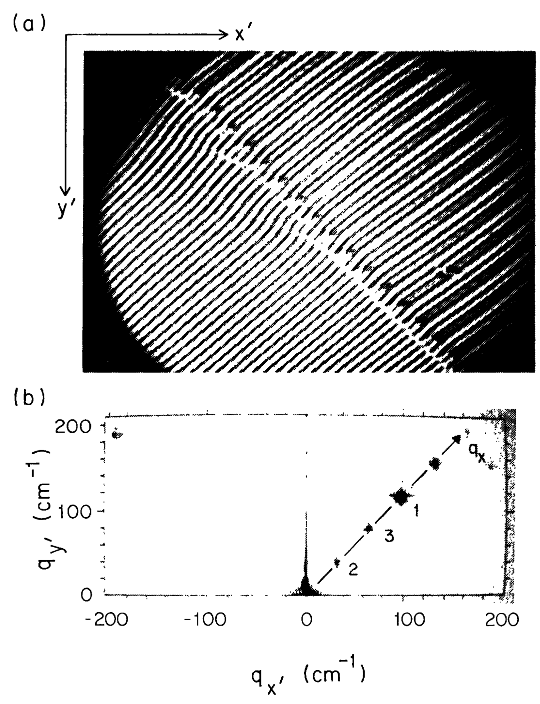

FIG. 2. (a) At the lower left, where the electrode is *not* interdigitated, the unperturbed Williams domains (roll size $l_0/2 = 65$ μm) are seen. At the upper right, the spatially periodic potential produces a one-dimensional commensurate or phase-locked structure (see text). (b) Two-dimensional Fourier spectrum (logarithmic intensity scale) of the pattern at the upper right of (a). The x direction is defined to be perpendicular to the electrode fingers (toward the lower right), and the q_x direction (arrow) is along the row of bright spots.

The patterns were studied as a function of roll size $l_0/2$ over the range 40 to 140 μm (by varying the layer thickness), while keeping l_1 fixed at 200 μm. The average voltage $(V_A + V_B)/2$ was set to 6.6 V (rms), about 10% higher than the threshold V_c for appearance of the Williams domains. For each of many values of the layer thickness, the steady-state patterns were studied for $V_A - V_B$ equal to 0.0, 0.2, and 0.6 V. In the discussion below, we focus on phenomena occurring in the range $l_0/2$ between 65 and 116 μm and $V_A - V_B = 0.6$ V.

When $l_0/2$ is within about 5% of $l_1/3$, we find [upper right portion of Fig. 2(a)] a one-dimensional phase-locked commensurate state in which there are three rolls per period of the potential; the rolls are parallel to the electrode fingers. The brightness of the rolls is modulated with period l_1, indicating a variation of the roll amplitude or the roll size or both.

In Fig. 2(b) we display the corresponding two-dimensional Fourier spectrum. It is most simply described in terms of rotated coordinates q_x and q_y (see caption). Most of the intensity is along the q_x direction in Fourier space, with the strongest peak (1) having its first moment at $q/2\pi = 150.5 \pm 1$ cm^{-1} and a linewidth of 4 cm^{-1}. This wave number corresponds to a perturbed roll size of 66.5 μm $= l_1/3$. We find that the position of this peak remains fixed even as the unperturbed roll size $l_0/2$ is varied by about 5%. The peak nearest the origin (2) is located at $q/2\pi = 1/l_1$, and corresponds to modulation of the structure by the perturbation. The power on the $q_{y'}$ (vertical) axis is instrumental.

For roll sizes in the range 84–116 μm we did not observe one-dimensional patterns, although it is possible that they would be induced for larger perturbations. Instead, we found that the favored structure for $V_A - V_B = 0.6$ V is a *two-dimensional* lattice, as shown in Fig. 3(a). The rolls are oriented predominantly along a line making an angle $\theta = 19°$ with respect to the electrode fingers (y axis). They are modulated by a regular lattice of kinks induced by the variation of the potential along x.

The corresponding power spectrum is shown in Fig. 3(b). In addition to the row of peaks along

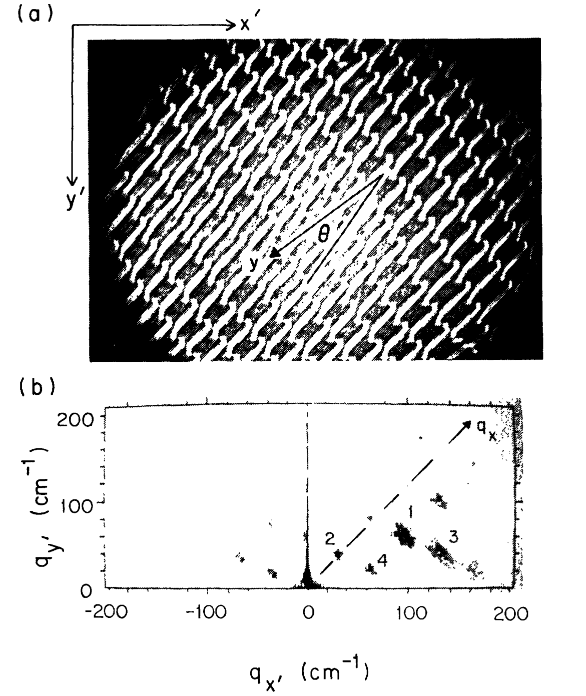

FIG. 3. (a) Two-dimensional pattern in the presence of a periodic perturbation, where the unperturbed roll size is $l_0/2 = 91$ μm. The system has a regular array of kinks. (b) Corresponding spectrum, showing that much of the power is off the q_x axis. Peaks are numbered in order of decreasing area.

the q_x direction, there are other rows parallel to it. The peak with the largest integrated area (1) is located at an angle θ below the q_x axis and corresponds to an optical intensity variation that is not orthogonal to the electrode fingers. This off-axis peak is somewhat broader than those in Fig. 2 in the azimuthal direction (about 7 cm^{-1}). We believe that irregularities in the lattice of kinks cause small spatial variations in the orientations of the rotated rolls, thus broadening the line. The next largest peak is the one nearest the origin (2), and is a response due to the perturbation at $q/2\pi = 1/l_1$. More than 60% of the total area lies under these two peaks. All peaks in the spectrum lie on a two-dimensional reciprocal lattice spanned by two basis vectors. Peak 3, for example, is located at twice the wave vector of peak 1 minus twice the wave vector of peak 2.

We characterize the two-dimensional patterns in Fig. 4 by plotting the wave number q of the dominant off-axis peak as a function of the wave number $2q_0$ corresponding to the unperturbed roll size. The solid line represents the condition $q = 2q_0$. The fact that the points lie close to the

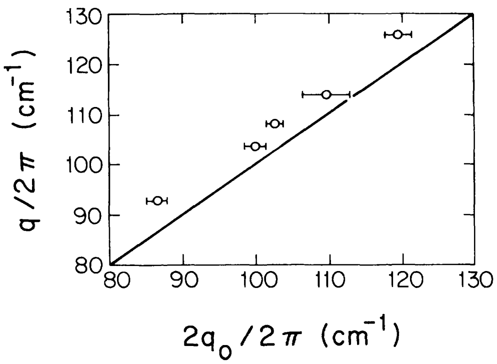

FIG. 4. Variation of the wave number q of the dominant off-axis peak with the unperturbed roll wave number $2q_0$. The solid line is the equation $q = 2q_0$.

line means that the off-axis peak corresponds approximately to a rotation of the unperturbed pattern. However, q is not exactly $2q_0$, indicating that the wave number is affected by the perturbation even away from lock-in. The smooth variation of one of the two basis vectors with q_0 supports the view that these modulated states are incommensurate.

We believe that it is possible to account qualitatively for there observations by introducing a Liapunov functional F that is minimized in the stable state. This approach is similar to that adopted by Cross[8] and others for understanding convective patterns. We decompose F into three parts:

$$F = F_R + F_S + F_P. \tag{1}$$

The first term describes the tendency to form rolls with wave number $q_0 = 2\pi/l_0$. It is identical to the functional used[8] to study Rayleigh-Bénard convection:

$$F_R = \int d^2r\, \tfrac{1}{2}[\epsilon^2 - \epsilon|\psi|^2 + g|\psi|^4 + C|(\nabla^2 + q_0^2)\psi|^2],$$

where ψ is a scalar amplitude from which all relevant dynamical variables can be obtained, $\epsilon = (V - V_c)/V_c$, g is a coupling constant, and C is a parameter with units of $(\text{length})^4$. The second term describes the alignment effect produced by surface treatment. It favors roll alignment along the y axis:

$$F_S = \int d^2r\, \tfrac{1}{2}\xi_y^2 |\nabla_y \psi|^2,$$

where ξ_y is a parameter having units of length. Finally, the last term represents the effect of the periodic perturbation:

$$F_P = -\sum_{n,m} \int d^2r\, U_{n,m} \operatorname{Re}[\exp(-imq_1 x)\psi^n],$$

where $U_{n,p}$ couples the pth harmonic of the perturbation to the nth harmonic of the amplitude. In the present case, the dominant term in the above sum for the commensurate structure is that with $p=3$ and $n=2$, favoring lock-in at $2l_1=3l_0$.

A model very similar to this has been used by Prost and Barois[4] to study equilibrium phases of smectic liquid crystals with competition between different length scales. For values of q_0 near lock-in (e.g., $q_0=3q_1/2$), they find a one-dimensional commensurate phase with Fourier intensities similar to those shown in Fig. 2. For q_0 away from lock-in, they find either one-dimensional or two-dimensional incommensurate structures (similar to those of Fig. 3), depending on the value of ξ_y^2.

In summary, we have observed both one-dimensional commensurate and two-dimensional incommensurate structures in a nonequilibrium system for which the important control parameters can be easily varied. These results are clearly preliminary and leave many questions unanswered. More detailed experimental and theoretical studies are in progress.

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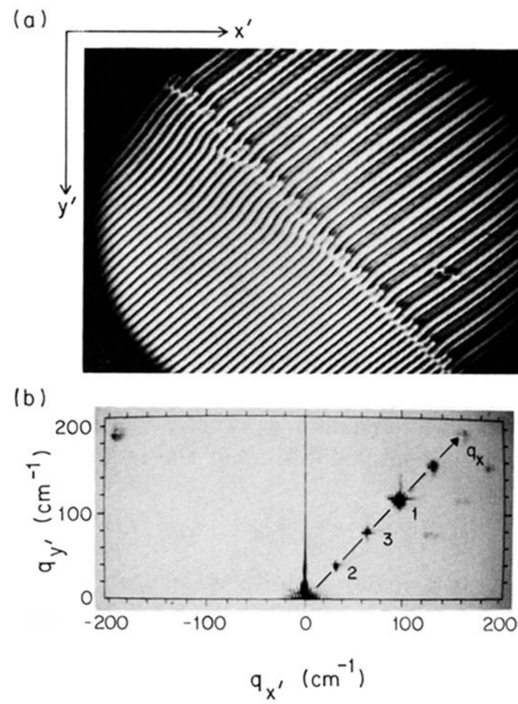

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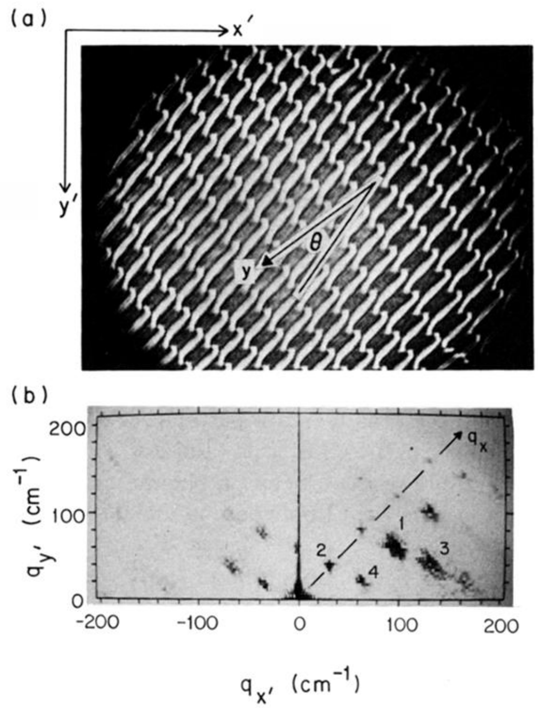

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