Field-Induced Transient Periodic Structures in Nematic Liquid Crystals: The Twist-Fréedericksz Transition

Franklin Lonberg, Seth Fraden, Alan J. Hurd, and Robert E. Meyer
The Martin Fisher School of Physics, Brandeis University, Waltham, Massachusetts 02254
(Received 2 January 1984)

Field-induced reorientations of liquid crystals, far from equilibrium, produce spatially periodic responses. The wavelength selected maximizes response speed. A detailed analysis of the effect in a novel geometry is presented, along with a discussion of its general importance in polymerlike liquid crystals.

PACS numbers: 61.30.Gd, 47.20.+m, 61.30.Eb, 62.10.+s

There are many important processes in nature in which far from equilibrium a dynamic periodic order emerges spontaneously from a homogeneous state.[1,2] In this Letter we present the view that reorientations of liquid crystals by electric or magnetic fields are, in general, members of this class of phenomena, and that such phenomena in liquid crystals, especially polymerlike liquid crystals, are much more important than has been realized until now. To support this view, we discuss the general nature of these phenomena and present an exact analysis of a case that has not been considered before, namely the twist-Fréedericksz transition, in which the sudden application of a large field results in the appearance of stripes in the sample (see Fig. 1).

The processes being considered have many similarities to other spatially periodic phenomena such as the Rayleigh-Bénard instability.[3] However, they are at the same time quite different in that they are transient responses rather than steady-state structures. Because of this, response speed is the characteristic which determines the mode that dominates. In these respects there are similarities to the spinodal-decomposition process.[4] As a brief statement of the physical origin of the effect discussed here, one can say that a periodic distortion can be a faster response than the uniform mode because it has a lower effective viscosity, as explained in detail below. Because of inherent nonlinearities, the distortion mode with the fastest initial growth rate suppresses all slower ones and becomes macroscopically observable.

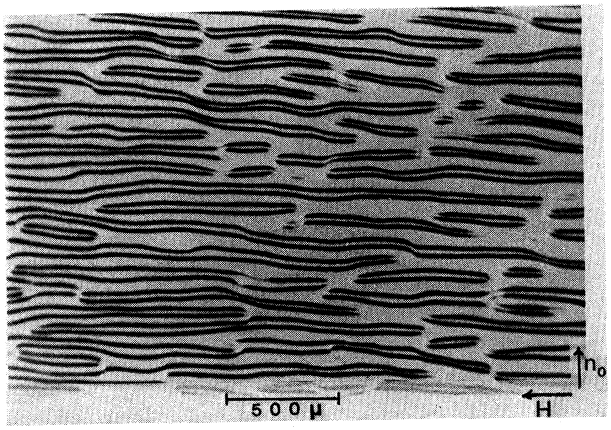

FIG. 1. Polarized light micrograph of a periodic twist-bend Fréedericksz distortion in MBBA. Sample thickness is 90 μm and field is $10H_c$.

With conventional liquid crystals composed of moderate length-to-width ratio molecules, the spatially periodic response to reorientating fields is not easily observed because it decays quickly to the homogeneous ground state. However, for nematics composed of very long molecules the large anisotropy in their viscosity[5] has two important consequences. First, the periodic instabilities appear under a very broad range of circumstances, and second the stripe patterns produced are very long-lived.

In subsequent sections of this Letter we will first present a theoretical analysis of this type of process in the particular geometry of the twist-Fredéericksz transition; second, experimental data will be cited to support this analysis; and finally the field-induced reorientation of liquid crystals in general will be discussed.

A nematic liquid crystal is a fluid phase composed of elongated molecules or particles which are oriented more or less parallel to one another in their ground state. The average orientation axis or director $\hat{n}$ can be aligned by surface interaction as well as by external magnetic (or electric) fields which interact with the anisotropy of the susceptibility χ_a. A single-crystal sample is formed by containing the nematic between parallel plates which are treated so as to align the director in a unique direction $\hat{n}_0$.

If a magnetic field $\vec{H}$, which tends to align $\hat{n}$

parallel to $\vec{H}$, is applied in the plane of the sample and perpendicular to $\hat{n}_0$ (see Fig. 2), a competition is set up between the field on the one hand, and the surface orientation forces mediated by the elasticity of the liquid crystal on the other. Above a threshold field $\vec{H}_c$, the interior of the sample begins to reorient toward $\vec{H}$, with twist elastic distortion being produced between the interior and the aligning walls, at which $\hat{n}$ remains parallel to $\hat{n}_0$ (the "strong anchoring" condition).[6] When a field above threshold is suddenly applied to the sample, any small perturbation in the initially uniform alignment will begin to grow exponentially with a rate that is inversely proportional to some effective viscosity for that particular reorientation process. For the simple twist-Fréedericksz transition described above, the uniform rotation of the director in the plane of the sample generates no flow and thus involves the pure rotational viscosity, γ_1. While this viscosity is only somewhat larger than the shear viscosities in low-molecular-weight nematics, it can be very much larger in the nematics composed of very long particles such as polymer chains or virus particles. Thus, especially in the latter case, the uniform reorientation rate can be very slow.

To see why a periodic pattern can give a faster response at high fields involves several considerations. First, since $\hat{n}$ and $-\hat{n}$ are equivalent, rotation in either direction from $\hat{n}_0$ lowers the magnetic free energy of the sample. Second, a nonuniform rotation pattern, e.g., oppositely rotating zones, produces a fluid flow response in addition to the molecular rotation. Third, this flow reinforces the opposite rotations of neighboring regions of the sample. Fourth, these oppositely rotating regions effectively replace γ_1 with a lower shear viscosity. These effects are summarized in Fig. 3. In low-molecular-weight nematics this trade-off of rotational for shear viscosity is not a large effect, since the coupling of rotation to shear flow is only of moderate strength. But in polymerlike nematics, in which this coupling is very strong, the viscosity reduction can be enormous[5]; therefore periodic responses abound.

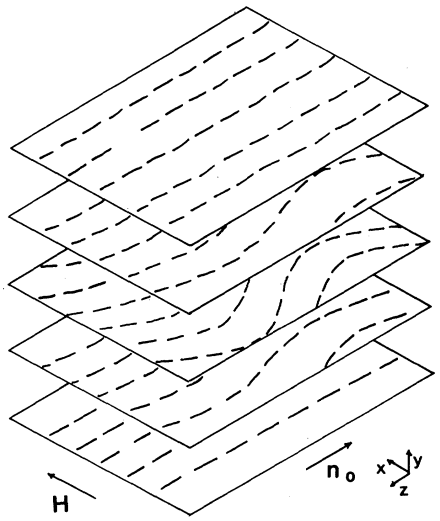

FIG. 2. Schematic representation of a periodic twist-bend Fréedericksz distortion.

Of course the nonuniform rotation modes involve additional internal elastic distortions which are absent in the uniform rotation mode. This leads to a compromise which determines the wavelength of the fastest-growing stripelike pattern; short wavelengths are energetically disfavored while long wavelengths have high viscosity. The optimum wavelength giving the fastest response is long, and perhaps infinite, for low fields near threshold; it grows shorter the higher the driving field.

The case we treat in detail is one in which a small-amplitude twist-Fréedericksz distortion is modulated in the z direction (see Fig. 2). The field has been turned on instantaneously at $t=0$. The following linearized calculation derives the relationship between the growth rate and the wavelength of an individual Fourier component of the modulation which is described by

$$\theta=\theta_0(t)\cos(k_y y)\cos(k_z z), \quad k_y\equiv\pi/d, \tag{1}$$

where θ is the angle between $\hat{n}$ and $\hat{n}_0$ and d is the sample thickness.

The balance-of-forces equation is the nematic form of the Navier-Stokes equation[7] for a system in which the inertial term is negligible:

$$\partial_\beta p+\partial_\alpha\sigma_{\alpha\beta}=0, \tag{2}$$

where the first term refers to the gradients of the pressure and the second to the gradients of the viscous-stress tensor. For a liquid crystal structure described by Eq. (1), only forces in the x direction

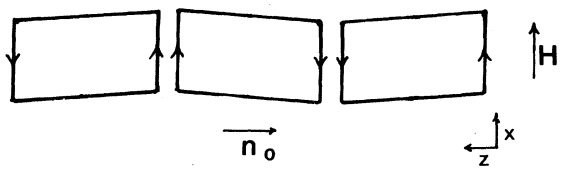

FIG. 3. Flows generated by oppositely rotating zones lower the rate of rotation of the director relative to the background fluid. Thus the highly dissipative pure rotation is replaced by flow gradients which have a lower viscosity.

are first order in θ. Since the system is invariant in the x direction the pressure gradient term in that direction is zero, and thus the only terms remaining are the gradients of the viscous-stress tensor. With use of the Leslie-Eriksen-Parodi formulation of nematodynamics[7] to evaluate the gradients, Eq. (2) for the forces in the x direction becomes

$$\eta_a \partial_y^2 V_x + \partial_z [\eta_c \partial_a V_x + \alpha_2 \dot{\theta}] = 0, \quad (3)$$

where V_x is the flow velocity in the x direction, α_2 is a Leslie viscosity coefficient, and η_a and η_c are the Meiscowitz combinations of Leslie viscosity coefficients describing simple shear viscosities.[7]

The balance-of-torques equation is

$$[K_2 \partial_y^2 \theta + K_3 \partial_z^2 \theta] + \chi_a H^2 \theta = \Gamma_v, \quad (4)$$

where the terms in the square brackets are the elastic torque with K_2 and K_3 being respectively the twist- and bend-Frank elastic constants. The second term is the field torque and Γ_v is the viscous-drag torque on the director. In the presence of shear flows established by Eq. (3), Γ_v is given by

$$\Gamma_v = \gamma_1 (\dot{\theta} - \tfrac{1}{2} \partial_z V_x) + \tfrac{1}{2} \gamma_2 \partial_z V_x. \quad (5)$$

Solving Eq. (3) using the θ field described in Eq. (1) yields, for the flow field,

$$V_x = \frac{-\alpha_2 k_z}{\eta_a k_y^2 + \eta_c k_z^2} \dot{\theta}_0(t) \cos(k_y y) \sin(k_z z). \quad (6)$$

Substituting Eq. (6) into Eq. (5) and solving yields

$$\theta_0 = \theta_{00} e^{st},$$
$$s(k_z) = \frac{\chi_a H^2 - K_2 k_y^2 - K_3 k_z^2}{\gamma_1 - \alpha_2^2 / [\eta_c + \eta_a (k_y / k_z)^2]}, \quad (7)$$

where θ_{00} is an initial amplitude produced by thermal fluctuations.

From Eq. (7) one can determine, for a given $\vec{H}$, a certain k_z which maximizes the growth rate s. This linearized analysis is valid as long as the amplitude of the response is sufficiently small. As they grow larger, the modes interact so that the fastest-growing mode will suppress all others and grow to an observable amplitude. The surprising result of this calculation is that it predicts a periodic response for almost any nematic at fields well above threshold, but no such observation has been reported for much-studied, common, thin-cell liquid crystals.

To test this calculation in a system for which all material parameters are known, we used a low-molecular-weight nematic, methoxybenzylidene-butylaniline (MBBA). A well-aligned sample was inserted rapidly into the field and the transient response was photographed. Figure 4 shows the agreement of the results with the calculation. The reason that such a simple phenomenon had been overlooked in years of research on ordinary nematics is probably that the stripes, which evolve into high mobility walls, recombine with one another within a few seconds.

The basic idea behind this calculation was applied to liquid crystals first by Guyon, Meyer, and Salan[8] in 1979, in the study of the splay Fréedericksz transition in very thick samples. Carr had also observed stripes induced in very thick nematic samples in a magnetic field.[9] More recently, there have been observations of similar stripe formation in lyotropic nematics,[10-12] and both Yu and Saupe[10] and Charvolin and Hendrikx[11] have suggested that the mechanism discussed here played a role in those cases. In our studies of nematics based on polymers and on tobacco mosaic virus,[13] we have found a rich variety of field-induced stripe patterns in splay-, bend-, and twist-Fréedericksz transitions, and in configurations with the field oriented obliquely to the director. Our analysis of several cases so far indicates that the dominance of the periodic response in the polymerlike systems is due to the very large anisotropy of the viscosity and the strong coupling between director rotation and shear flow in nematics composed of very long particles.[5] This accounts for both the induction of a periodic response in almost any geometry and the very long life of the resulting stripes. Once the physics was clear, we realized that with high driving fields, applied very quickly, stripes should appear in the commonly studied thin samples of ordinary nematics as well. We

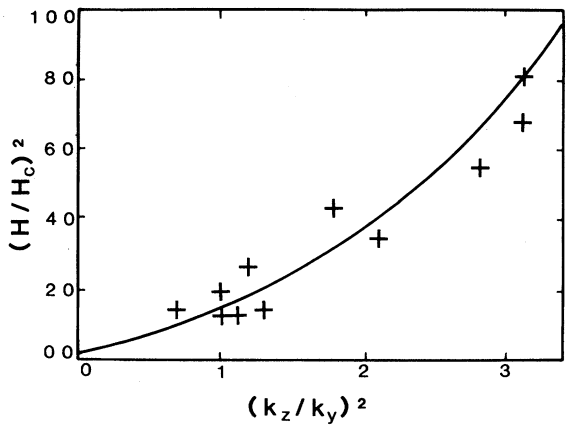

FIG. 4. Observed relative wave vectors vs relative field. Calculated curve based on literature values for MBBA. $K_3/K_2 = 2.5$; $\alpha_2^2/\gamma_1\eta_c = 0.74$; $\eta_a/\eta_c = 0.2$.

have found them for several configurations, including the one described here, in which they have been overlooked until now. The observation of easily induced, long-lived stripes in lyotropic nematics suggests that the micellar aggregates from which they are formed may act much like long polymer chains. Our observations on polymer and virus nematics, and their detailed analysis, will be presented elsewhere. These phenomena provide a useful means of studying both liquid crystals and dynamic periodic structures in general.

The authors wish to acknowledge many helpful discussions with Hiap Liew Ong, and thank Irving Epstein and Leigh Sneddon for critical readings of the manuscript. This work was supported by the National Science Foundation through Grant No. DMR-8210477-01, and by the Martin Fisher School of Physics of Brandeis University. One of us (F.L.) also thanks the IBM Corporation for a summer research fellowship.

[1]H. Haken, *Synergetics: An Introduction* (Springer-Verlag, New York, 1978).

[2]G. Nicolis and I. Prigogine, *Self-Organization in Nonequilibrium Systems* (Wiley, New York, 1977).

[3]S. Chandrasekhar, *Hydrodynamic and Hydromagnetic Instability* (Oxford Univ. Press, London, 1961).

[4]J. W. Cahn and J. E. Hilliard, J. Chem. Phys. **28**, 258 (1958).

[5]P. G. de Gennes, in *Polymer Liquid Crystals,* edited by A. Ciferri, W. R. Krigbaum, and R. B. Meyer (Academic, New York, 1982); R. B. Meyer, *ibid.*

[6]H. J. Deuling, in *Solid State Physics,* edited by H. Ehrenreich, F. Seitz, D. Turnbull, and L. Liebert (Academic, New York, 1978), Vol. 14, p. 77–107.

[7]P. G. de Gennes, *The Physics of Liquid Crystals* (Clarendon, Oxford, 1975).

[8]E. Guyon, R. Meyer, and J. Salan, Mol. Cryst. Liq. Cryst. **54**, 261 (1979).

[9]E. F. Carr, Mol. Cryst. Liq. Cryst. **34**, L159 (1977).

[10]L. J. Yu and A. Saupe, J. Am. Chem. Soc. **102**, 4879 (1980).

[11]J. Charvolin and Y. Hendrikx, J. Phys. (Paris), Lett. **41**, L597 (1980).

[12]H. Lee and M. M. Labes, Mol. Cryst. Liq. Cryst. **84**, 137 (1982).

[13]S. Fraden, F. Lonberg, A. J. Hurd, R. B. Meyer, in Proceedings of the GTE Symposium on Order in Polymeric Materials, Waltham, Mass., August 1983 (to be published), and in Proceedings of the Workshop on Colloidal Crystals, Les Houches, February 1984 (to be published).

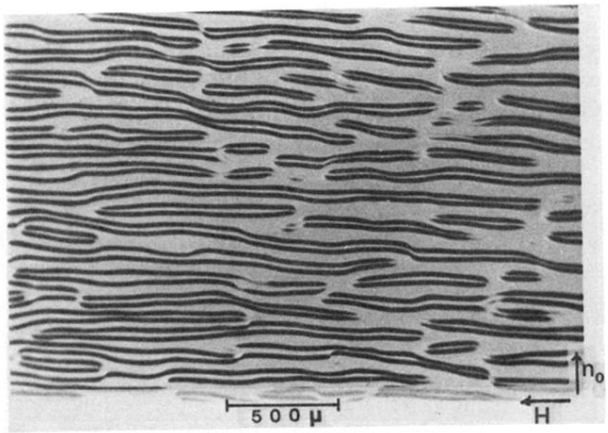

FIG. 1. Polarized light micrograph of a periodic twist-bend Fréedericksz distortion in MBBA. Sample thickness is 90 μm and field is $10H_c$.