Nonlinear effects in the dynamics of transient pattern formation in nematics

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(Received 25 June 1987)

A nonlinear calculation of the dynamics of transient pattern formation in the Fréedericksz transition is presented. A Gaussian decoupling is used to calculate the time dependence of the structure factor. The calculation confirms the range of validity of linear calculations argued in earlier work. In addition, it describes the decay of the transient pattern.

Liquid crystals exhibit extremely interesting nonequilibrium features in the presence of external electric or magnetic fields.[1] We are interested here in the description of pattern development associated with the Fréedericksz transition in nematics. This is a magnetic instability occurring when an initially well-aligned liquid-crystal sample is placed under a magnetic field $H > H_c$ applied perpendicular to the initial orientation of the molecules. There is now broad experimental evidence[2–6] of the occurrence of transient spatial structures during the early stages of the Fréedericksz transition. In this situation, hydrodynamical aspects seem to play a crucial role since the director field is dynamically coupled to a velocity flow. Such coupling accounts for the periodic pattern of opposite but equivalent reorientations. In the simplest cases the pattern consists of a collection of stripes perpendicular to the initial director.[2–4] Oblique[5] and two-dimensional[6] structures have also been observed.

A linear deterministic analysis of the nematodynamic equations has been broadly used to identify the most unstable mode whose characteristic wave number is then associated with the observed periodicity of the transient pattern.[2–7] We have recently[8] addressed the question of the theoretical description of the dynamics associated with this transient pattern development in a pure twist geometry. This was accomplished using a Langevin version of the nonlinear nematodynamic equations which include stochastic forces. The model we proposed is a generalized time-dependent Ginzburg-Landau model of the sort used previously in a variety of contexts (critical dynamics[9] or dynamics of phase transitions[10]). It takes consistently into account thermal fluctuations, associated with both the director and the velocity fields, satisfying appropriate fluctuation-dissipation relations. The initial stages of the decay from an unstable state are clearly governed by fluctuations, so that their consistent inclusion is necessary in any theory aimed to give a proper description of the time scales involved in the relaxation phenomena.

Once the general nonlinear stochastic nematodynamics model was introduced, we made, in Ref. 8, a linearized analysis in analogy with the Cahn-Hilliard-Cook (CHC) theory of spinodal decomposition.[10] However, we remarked that the range of validity of such a linear analysis seemed to be notably larger than in the case of spinodal decomposition of systems with short-range forces.[11–13] This conclusion, already noted in previous studies of the dynamics of the twist distortion neglecting hydrodynamic effects,[14,15] was supported by a calculation of the mean first-passage time (MFPT) the system takes to leave the unstable state.

Our purpose here is to go beyond the linear approximation. Nonlinear contributions in the dynamical equations for the time-dependent structure factor corresponding to the orientation of the director will be considered by means of a Gaussian decoupling.[10,13–15] This seems to be a first nonlinear description of the dynamics of the transient pattern formation associated with the Fréedericksz transition. The position of the maximum of the structure factor is used to characterize the observed periodicity and its time dependence to characterize the evolution of the pattern. Our results confirm the essential validity of the previous linear analysis over a time scale, accessible to experimentation, in which the pattern emerges and becomes well developed. At the same time, it gives a first description of the decay of the pattern to the final homogeneous state.

In our previous work[8] we derived coupled nonlinear stochastic equations for the director field **n** and velocity flow **v**. In the twist geometry, with a director initially aligned along the x axis, the magnetic field H along the y axis, and assuming macroscopic flow only in the y direction, these equations become, in a minimal coupling approximation,[8]

$$d_t \begin{bmatrix} \phi \\ v_y \end{bmatrix} = \begin{bmatrix} -1/\gamma_1 & \left[\dfrac{1}{2\rho}\right](\lambda+1)\partial_x \\ \left[\dfrac{1}{2\rho}\right](\lambda+1)\partial_x & \left[\dfrac{1}{\rho^2}\right](\nu_2\partial_z^2+\nu_3\partial_x^2) \end{bmatrix} \times \begin{bmatrix} \dfrac{\delta F}{\delta \phi} \\ \dfrac{\delta F}{\delta v_y} \end{bmatrix} + \begin{bmatrix} \xi \\ \partial_x \Omega_{yx} + \partial_z \Omega_{yz} \end{bmatrix} . \tag{1}$$

Here the director is assumed to reorientate in the x,y plane and $n_x(x,z)=\cos\phi(x,z)$. In (1), ρ is the mass density, γ_1, γ_2, ν_2, and ν_3 viscosity coefficients, $\lambda=-\gamma_2/\gamma_1$, and F the free energy,

$$\frac{\delta F}{\delta\phi}=-[K_{22}\partial_z^2\phi+K_{33}\partial_x^2\phi+\chi_a H^2(\phi-\tfrac{1}{3}\phi^3)] , \qquad (2)$$

$$\frac{\delta F}{\delta v_y}=\rho v_y , \qquad (3)$$

where K_{22} and K_{33} are elastic constants associated, respectively, with splay and bend deformations and χ_a is the anisotropic part of the magnetic susceptibility. The Gaussian random forces in (1) satisfy the following fluctuation dissipation relations:

$$\langle \xi(\mathbf{r},t)\xi(\mathbf{r}',t')\rangle=2\frac{K_BT}{\gamma_1 L}\delta(x-x')\delta(z-z')\delta(t-t') , \qquad (4)$$

$$\langle \Omega_{y\alpha}(\mathbf{r},t)\Omega_{y\beta}(\mathbf{r}',t')\rangle =2\frac{K_BT}{\rho^2 L}\nu_\alpha\delta_{\alpha\beta}\delta(x-x')\delta(z-z')\delta(t-t') , \quad \alpha,\beta=\{x,z\} \qquad (5)$$

with $\nu_x=\nu_3, \nu_z=\nu_2$, and L is the y-linear dimension of the sample.

Using the common approximation of negligible inertia,[2-4] we obtain a nonlinear equation for the Fourier amplitude[8] of the deformation angle $\theta_{m,q_x}(t)$,

$$\partial_t\theta_{m,q_x}(t)=\frac{1}{\bar{\gamma}_1}\Bigg[\left[\chi_a H^2-K_{22}(2m+1)^2\frac{\pi^2}{d^2}-K_{33}q_x^2\right]\theta_{m,q_x}(t) -\tfrac{2}{3}\chi_a H^2\sum_{q_{x_1},q_{x_2}}\sum_{n,l,p}\theta_{n,q_{x_1}}(t)\theta_{l,q_{x_2}}(t)\theta_{p,q_x-(q_{x_1}+q_{x_2})}(t) \times\frac{2}{d}\int_{-d/2}^{d/2}dz\cos(2m+1)\frac{\pi z}{d}\cos(2n+1)\frac{\pi z}{d}\cos(2l+1)\frac{\pi z}{d} \times\cos(2p+1)\frac{\pi z}{d}\Bigg]+\eta_{m,q_x}(t) . \qquad (6)$$

$\bar{\gamma}_1$ is the effective wave-number-dependent viscosity which includes hydrodynamics effects,[8]

$$\bar{\gamma}_1=\gamma_1-\frac{\alpha_2^2}{\eta_c+\eta_a Q^{-2}}, \quad Q=\frac{q_x}{(2m+1)\frac{\pi}{d}} , \qquad (7)$$

where α_2,η_a, and η_c are the usual Leslie and Meiscowitz coefficients

$$\alpha_2=-\tfrac{1}{2}\gamma_1(\lambda+1)^2 , \qquad (8)$$

$$\eta_a=\nu_2 , \qquad (9)$$

$$\eta_c=\nu_3+\tfrac{1}{4}\gamma_1(\lambda+1)^2 , \qquad (10)$$

and $\eta_{m,q_x}(t)$ is a Gaussian stochastic force which satisfies a consistent fluctuation-dissipation relation with $\bar{\gamma}_1$,

$$\langle \eta_{m,q_x}(t)\eta^*_{n,q_x'}(t')\rangle=2\frac{2K_BT}{\bar{\gamma}_1 V}\delta_{m,n}\delta_{q_x,q_x'}\delta(t-t') . \qquad (11)$$

The transient dynamics of the pattern formation and decay is followed by monitoring the evolution of the time-dependent structure factor

$$C_{m,q_x}(t)=\langle \theta_{m,q_x}(t)\theta_{m,-q_x}(t)\rangle .$$

Starting with (6) and using (11), standard methods lead to the evolution equation for the structure factor,

$$\frac{d}{dt}C_{m,q_x}(t)=\frac{e}{\bar{\gamma}_1}\Bigg[\left[\chi_a H^2-K_{22}(2m+1)^2\frac{\pi^2}{d^2}-K_{33}q_x^2\right]C_{m,q_x}(t) -\tfrac{2}{3}\chi_a H^2\sum_{q_{x_1},q_{x_2}}\sum_{n,l,p}\langle\theta_{n,q_{x_1}}(t)\theta_{l,q_{x_2}}(t)\theta_{p,q_x-(q_{x_1}+q_{x_2})}\theta_{m,-q_x}(t)\rangle \times\frac{2}{d}\int_{-d/2}^{d/2}dz\cos(2m+1)\frac{\pi z}{d}\cos(2n+1)\frac{\pi z}{d}\cos(2l+1)\frac{\pi z}{d} \times\cos(2p+1)\frac{\pi z}{d}\Bigg]+\frac{2}{\bar{\gamma}_1}\frac{2K_BT}{V} . \qquad (12)$$

Focusing on the most unstable mode $m=0$ which dominates the dynamics of the instability and decoupling it from other m modes, we obtain $[C_{m=0,q_x}(t)\equiv C_{q_x}(t);\ \theta_{m=0,q_x}(t)\equiv\theta_{q_x}(t)]$

$$\frac{d}{dt}C_{q_x}(t)=\frac{2}{\bar{\gamma}_1(m=0)}\left[\left[\chi_a H^2-K_{22}\frac{\pi^2}{d^2}-K_{33}q_x^2\right]C_{q_x}(t)-\tfrac{1}{2}\chi_a H^2\sum_{q_{x_1},q_{x_2}}\langle\theta_{q_{x_1}}(t)\theta_{q_{x_2}}(t)\theta_{q_x-(q_{x_1}+q_{x_2})}(t)\theta_{-q_x}(t)\rangle\right]+\frac{2}{\bar{\gamma}_1(m=0)}\frac{2K_BT}{V}\ . \qquad (13)$$

This equation is clearly the first of a hierarchy of equations which couples $C_{q_x}(t)$ to higher-order correlation functions. To obtain a solution of (13), an approximation is required in order to truncate this hierachy. We propose here the use of a Gaussian decoupling ansatz in which

$$\sum_{q_{x_1},q_{x_2}}\langle\theta_{q_{x_1}}(t)\theta_{q_{x_2}}(t)\theta_{q_x-(q_{x_1}+q_{x_2})}(t)\theta_{-q_x}(t)\rangle \simeq 3\left[\sum_k\langle\theta_k(t)\theta_{-k}(t)\rangle\right]C_{q_x}(t)\ . \qquad (14)$$

A Gaussian decoupling of this sort was already used[14] to describe the transient dynamics of orientational fluctuations for weak magnetic fields for which hydrodynamic coupling can be neglected. The nonlinearity which prevents the nonphysical unlimited growth of the unstable modes in a linear theory is introduced through the temporal evolution of

$$\sum_k\langle\theta_k(t)\theta_{-k}(t)\rangle=\langle\phi^2_{m=0}(x,z,t)\rangle\ .$$

This nonlinear scheme certainly gives a more accurate description of the early stages of relaxation from the unstable state. In the final approach to the stationary homogeneous distorted configuration, the mobility and recombination of defect walls should play an important role in the mechanisms of decay of the transient pattern. It is not clear that these effects are well described in a Gaussian approximation. However, this approximation does give a first picture of the decay of the transient pattern to a homogeneous state.[16]

A convenient way to rewrite (13) makes use of a dimensionless less time variable s, a reduced magnetic field h, and wave number Q ($Q=q_x/(\pi/d)$),

$$s=\tau_0^{-1}t,\quad \tau_0=\gamma_1/\chi_a H_c^2\ , \qquad (15)$$

$$h^2=H^2/H_c^2,\quad H_c^2=\frac{K_{22}\pi^2}{\chi_a d^2}\ , \qquad (16)$$

$$\frac{d}{ds}C_{q_x}(s)=\frac{2}{f(Q)}\left[h^2-1-\frac{K_{33}}{K_{22}}\left[\frac{q_x}{\pi/d}\right]^2-\frac{3}{2}h^2\left[\sum_k C_k(s)\right]\right]C_{q_x}(s)+\frac{2}{f(Q)}\varepsilon\ , \qquad (17)$$

where $f(Q)$ is the lowering factor for the effective viscosity,[8]

$$f(Q)=1-\frac{\alpha_2^2/\gamma_1\eta_c}{1+(\eta_a/\eta_c)Q^{-2}}\ , \qquad (18)$$

and ε measures the strength of thermal fluctuations

$$\varepsilon=2\frac{K_BT/V}{\chi_a H_c^2}\ . \qquad (19)$$

Equation (17) has been solved numerically by a self-consistent method after a limiting procedure ($V\to\infty$) has been applied to transform the k-mode summation into an integral,

$$\sum_k C_k(s)=(2\pi)^{-1}S^{1/2}\int_0^\infty dk C_k(s)\ . \qquad (20)$$

S stands for the sample surface in the x-y plane. Typical values for the material parameters corresponding to MBBA at room temperatures were used. The initial conditions appropriate to (17) were consistently obtained as a

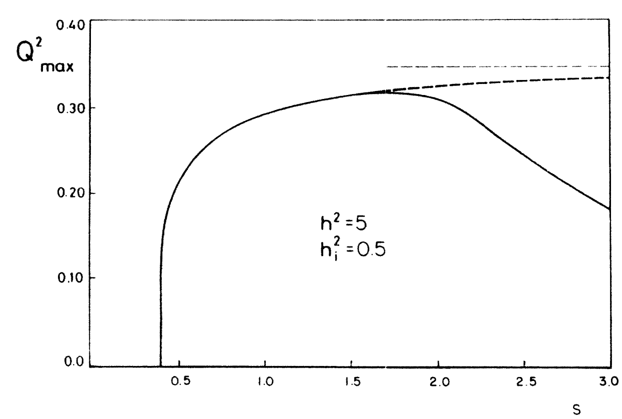

FIG. 1. Graph of wave number corresponding to the maximum of the structure factor vs time. Results of the linear theory (Ref. 8) are represented by the dashed line. Parameter values are those of MBBA (Ref. 1): $\alpha_2^2/(\gamma_1\eta_c)=0.74$, $\eta_a/\eta_c=0.40$, and $K_{33}/K_{22}=2.5$. The initial and final reduced magnetic fields are, respectively, $h_i^2=0.5$ and $h^2=5.0$. Times are measured in units of $\tau_0\simeq 10$ sec, corresponding to typical samples ($S=1$ cm^2, $d=10^{-2}$ cm). The asymptotic value obtained as the mode of fastest growth in a deterministic linear stability analysis is also depicted.

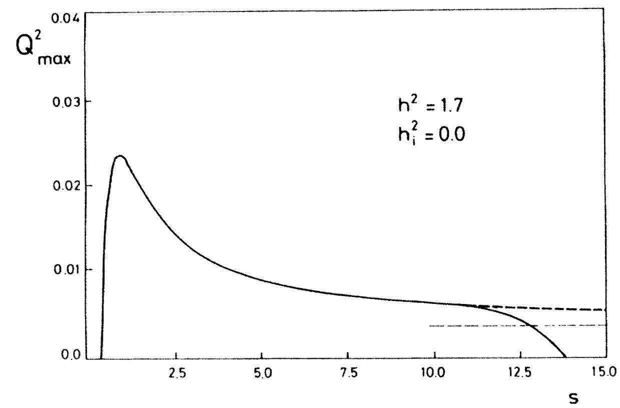

FIG. 2. Wave number corresponding to the maximum of the structure factor vs time for different values of material parameters (Ref. 3) and initial and final reduced magnetic fields: $\eta_a/\eta_c = 0.20$, $h_i^2 = 0$, and $h^2 = 1.7$.

linear stationary situation of (17) corresponding to $H_i < H_c$,

$$C_{q_x}(0) = \frac{\varepsilon}{1 + \frac{K_{33}}{K_{22}}\left[\frac{q_x}{\pi/d}\right]^2 - h_i^2} . \tag{21}$$

In Fig. 1 we describe the dynamics of the pattern formation by plotting $Q^2_{\max}$, the wave number corresponding to the maximum of the structure factor $C_{q_x}(s)$, as a function of time. The wavelength associated with $Q_{\max}$ corresponds to the periodicity of the pattern. Figure 1 shows a first stage of evolution ending with the emergence of the periodicity at $s \simeq 0.4$, a second stage of pattern formation and development up to $s \simeq 2$, and a last stage of decay of the pattern to a homogeneous sample $Q_{\max} \to 0$. The nonlinear evolution obtained from (17) is compared in Fig. 1 with the corresponding result of the linearized calculation.[8] As we anticipated, both calculations coincide extremely well during the stages of emergence and development of the pattern. The important fact is that those time intervals are easily accessible to usual experimental standards (for the parameters values of Fig. 1, $\tau_0 \sim 10$ sec, so that $s = 2.0$ corresponds to 20 sec). Notice that the linear theory fails to describe the decay of the pattern. An additional comment concerns our previous estimate[8] of the domain of validity of linear theory through the calculation of a mean first-passage time T. In the dimensionless time scale s, we obtained $T \simeq 2.5$, which gives an essentially correct upper bound for the time domain of validity.

In Fig. 2 we show the same calculation for different values of the material parameters. For these material parameters there are values of the initial and final magnetic fields for which $Q_{\max}$ is not a monotonously increasing function of time during the stage of pattern formation and development. An example of this situation is given in this figure: pattern formation and development manifest in a rapid growth of $Q_{\max}$ which then decays slowly to the asymptotic value obtained as the mode of fastest growth in a deterministic linear stability analysis. A later decay to $Q_{\max} = 0$ occurs when nonlinear effects come into play. The same comments made for the comparison of the linear and nonlinear calculation in Fig. 1 apply here.

This work has been supported by the Comisión Asesora para la Investigación Científica y Técnica Project 361/84 and by Dirección General de Investigación Científica y Técnica Project PB-86-0534.

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[16]In comparison with the shortcoming of the Gaussian approximation in the problem of spinodal decomposition (Ref. 10), we mention that, in our case, the final stationary state is not one of two-phase coexistence and therefore it is still described by a simple-peaked distribution.