

Transient dynamics of orientational fluctuations in the Fréedericksz transition

F. Sagués*

Departamento de Química Física, Universidad de Barcelona, Diagonal 647, 08028 Barcelona, Spain

M. San Miguel

Departamento de Física Teórica, Universidad de Barcelona, Diagonal 647, 08028 Barcelona, Spain

(Received 17 June 1985)

Transient dynamics of spatial fluctuations of the director field in the pure twist Fréedericksz transition is studied. A nonlinear calculation is presented. Anomalous transient fluctuations are shown. Different stages of evolution and the domain of validity of linear theories are discussed.

I. INTRODUCTION

Fréedericksz transition in a nematic slab occurs when the director reorientates in the direction of an applied magnetic field larger than a critical one H_c . The standard analysis of the transient dynamics (nonlinear relaxation) originated when the magnetic field is suddenly changed from a value $H_i < H_c$ to a final value $H > H_c$ is based on a mean field and deterministic model.¹⁻³ In this model spatial inhomogeneities and thermal fluctuations are not taken into account. However, a proper description of the decay of an unstable state requires the consideration of fluctuations. In addition a decay process is accompanied by anomalously large fluctuations as compared to those of equilibrium (transient anomalous fluctuation phenomenon).^{4,5} On the other hand, at the transition point a correlation length diverges in the plane in which the director reorientates, and spatial fluctuations associated with such two-dimensional-reorientation of the molecules are expected to be important. The purpose of this paper is to study the transient anomalous spatial fluctuations that appear in the dynamics of the Fréedericksz transition. We calculate the transient behavior of the structure factor of the director fluctuations.

Experimental work on the dynamics of the Fréedericksz transition^{6,7} shows the existence of transient spatial structures. In this context the analogy of the problem at hand with the problem of spinodal decomposition⁸ has been stressed.^{6,7} The analysis of Refs. 6 and 7 takes into account the essential spatial inhomogeneities but it is restricted to a linear theory and neglects thermal fluctuations. These approximations are known to be often unsatisfactory for the problem of spinodal decomposition.⁸ Partially motivated by these facts we present here a nonlinear calculation of the structure factor of the director fluctuations which consistently takes into account the thermal fluctuations of the system. In this paper we do not address the description of long-lived transient spatial structures,⁹ but our calculation should be a useful guide in more complicated situations, and in particular to analyze the domain of validity of linear approximations.

Our analysis is based on a model introduced earlier¹⁰ to justify a mean field study of the dynamics of the Fréedericksz transition in a random magnetic field.¹¹

Here we are not concerned with random fields but we go beyond the mean field description. Our results for the structure factor exhibit the anomalous fluctuation phenomenon for the spatial fluctuations. In its evolution, well-defined time scales can be distinguished. It is found that the time scale in which the system leaves the unstable state is reasonably described by linear theory. This time scale can be large enough to be of experimental relevance.

This paper is organized so that in Sec. II we define our stochastic dynamical model and we discuss the results of the linear calculation. In Sec. III we present a nonlinear calculation based on a Gaussian decoupling.

II. MODEL AND LINEAR THEORY

We consider a twist geometry for a nematic sample contained between two plates placed perpendicular to the z axis and separated a distance d . We assume strong anchoring at $z = \pm d/2$. Initially the molecules are, on the average, aligned along the x axis and a magnetic field $H_i < H_c$ (in particular $H_i = 0$) is directed along the y axis. At time $t = 0$ the magnitude of this magnetic field is instantaneously changed to a value $H > H_c$, so that the system becomes unstable and the molecules tend to align parallel to the magnetic field. We focus on the evolution of the spatial fluctuations during the decay of this unstable state. The model introduced in an earlier publication¹⁰ contains two main assumptions. The first assumption consists in neglecting hydrodynamical coupling of the director and velocity fields. This is reasonable for a twist geometry and magnetic fields not much larger than the critical one. Second, the model assumes that the director rotates in x - y plane

$$\begin{aligned} n_x(\mathbf{r}, t') &= \cos\phi(\mathbf{r}, t'), \\ n_y(\mathbf{r}, t') &= \sin\phi(\mathbf{r}, t'). \end{aligned} \quad (1)$$

The initial small z component of the director associated with equilibrium fluctuations remains stable during the transient evolution, while fluctuations in the x - y components become unstable and they are macroscopically amplified when switching on the magnetic field H in the

y direction. As a consequence, and in a first approximation, it seems reasonable to set $n_z=0$ when studying transient behavior. In these circumstances a dynamical model of the Ginzburg-Landau type for the evolution of $\phi(\mathbf{r}, t')$ can be introduced¹⁰ through

$$\partial_{t'}\phi(\mathbf{r}, t') = \frac{1}{\gamma_1} [K_{11}\partial_y^2\phi + K_{22}\partial_z^2\phi + K_{33}\partial_x^2\phi + \chi_a H^2(\phi - \frac{2}{3}\phi^3)] + \eta(\mathbf{r}, t'). \quad (2)$$

Here t' stands for time in the laboratory time scale, γ_1 is the twist viscosity, K_{11}, K_{22}, K_{33} are Frank's elastic constants associated, respectively, with splay, twist, and bend deformations, and χ_a is the anisotropic part of the magnetic susceptibility. The random force $\eta(\mathbf{r}, t')$ is such that the stochastic process defined by (2) has a canonical stationary solution $P_{st} \propto e^{-F/k_B T}$ (F is the free energy functional) describing equilibrium fluctuations. The process $\eta(\mathbf{r}, t')$ is then taken to be Gaussian of zero mean and satisfying a fluctuation-dissipation relation

$$\langle \eta(\mathbf{r}_1, t'_1) \eta(\mathbf{r}_2, t'_2) \rangle = 2 \frac{k_B T}{\gamma_1} \delta(\mathbf{r}_1 - \mathbf{r}_2) \delta(t'_1 - t'_2). \quad (3)$$

A linear stability analysis of (2) can be easily performed using a double Fourier transformation

$$\phi(\boldsymbol{\rho}, z, t') = \sum_m \cos\left[(2m+1)\frac{\pi z}{d}\right] \phi_m(\boldsymbol{\rho}, t'), \quad (4)$$

$$\phi_m(\boldsymbol{\rho}, t') = \sum_q e^{i\mathbf{q}\cdot\boldsymbol{\rho}} \theta_{m,\mathbf{q}}(t'). \quad (5)$$

$\boldsymbol{\rho}$ is the position vector in the x - y plane, and the first Fourier transform assures the fulfillment of the strong anchoring boundary condition $\phi(\boldsymbol{\rho}, z = \pm d/z, t') = 0$. It is found¹⁰ that $\theta_{m,\mathbf{q}}(t')$ is unstable for

$$1 - (2m+1)^2 \left[\frac{H_c}{H} \right]^2 > 0, \quad q_c^2(m) > Q^2, \quad (6)$$

where the critical field H_c is given by

$$H_c \equiv \left[\frac{K_{22} n^2}{\chi_a d^2} \right]^{1/2}. \quad (7)$$

$q_c(m)$ is a critical wave number whose significance is discussed below:

$$q_c^2(m) \equiv \zeta_2^{-2} \left[1 - (2m+1)^2 \left[\frac{H_c}{H} \right]^2 \right]. \quad (8)$$

ζ_i are magnetic coherence lengths^{2,3}

$$\zeta_i^2 \equiv \frac{K_{ii}}{\chi_a H^2} \quad i=1,2,3 \quad (9)$$

and Q^2 is a wave number which averages the anisotropy in the x - y plane of the sample

$$Q^2 \equiv \frac{\zeta_1^2}{\zeta_2^2} q_y^2 + \frac{\zeta_3^2}{\zeta_2^2} q_x^2. \quad (10)$$

We will consider situations for which $H_c < H < 3H_c$. In this case only the mode $m=0$ becomes unstable. We study the spatial fluctuations in the x - y plane of the mode $m=0$. These are given by the \mathbf{q} modes of $\phi_0(\boldsymbol{\rho}, t')$, some of which are unstable. Leaving implicit the subindex $m=0$, the equation for $\phi_0(\boldsymbol{\rho}, t')$ reads

$$\partial_{t'}\phi(\boldsymbol{\rho}, t') = \frac{1}{\gamma_1} \left[K_{11}\partial_y^2\phi + K_{33}\partial_x^2\phi - K_{22}\frac{\pi^2}{d^2}\phi + \chi_a H^2(\phi - \frac{1}{2}\phi^3) \right] + \eta(\boldsymbol{\rho}, t'), \quad (11)$$

where

$$\langle \eta(\boldsymbol{\rho}_1, t'_1) \eta(\boldsymbol{\rho}_2, t'_2) \rangle = 2 \left[\frac{2}{d} \frac{k_B T}{\gamma_1} \right] \delta(\boldsymbol{\rho}_1 - \boldsymbol{\rho}_2) \delta(t'_1 - t'_2). \quad (12)$$

Fluctuations in the $\boldsymbol{\rho}$ plane are described by the correlation function $C_{11}(\boldsymbol{\rho}_1 - \boldsymbol{\rho}_2, t') \equiv \langle \phi(\boldsymbol{\rho}_1, t') \phi(\boldsymbol{\rho}_2, t') \rangle$. The dynamical equation for C_{11} follows from (11) and (12):

$$\begin{aligned} \partial_{t'} C_{11}(\boldsymbol{\rho}_1 - \boldsymbol{\rho}_2, t') = & \frac{2}{\gamma_1} \left[\left[K_{11}\partial_y^2 + K_{33}\partial_x^2 - K_{22}\frac{\pi^2}{d^2} \right. \right. \\ & \left. \left. + \chi_a H^2 \right] C_{11}(\boldsymbol{\rho}_1 - \boldsymbol{\rho}_2, t') \right. \\ & \left. - \frac{1}{2} C_{31}(\boldsymbol{\rho}_1 - \boldsymbol{\rho}_2, t') \right] \\ & + 2 \left[\frac{2}{d} \frac{k_B T}{\gamma_1} \right] \delta(\boldsymbol{\rho}_1 - \boldsymbol{\rho}_2), \quad (13) \end{aligned}$$

where $C_{31}(\boldsymbol{\rho}_1 - \boldsymbol{\rho}_2, t') \equiv \langle \phi^3(\boldsymbol{\rho}_1, t') \phi(\boldsymbol{\rho}_2, t') \rangle$. Introducing a dimensionless time

$$t \equiv \left[1 - \left[\frac{H_c}{H} \right]^2 \right] \tau t', \quad \tau \equiv \frac{\chi_a H^2}{\gamma_1},$$

using the notation introduced after (6) for $m=0$, and with

$$\bar{\epsilon} \equiv \frac{2k_B T}{d\chi_a H^2} \left[1 - \left[\frac{H_c}{H} \right]^2 \right]^{-1},$$

Eq. (13) is rewritten as

$$\begin{aligned} \partial_t C_{11}(\boldsymbol{\rho}_1 - \boldsymbol{\rho}_2, t) = & 2 \left[1 - \frac{1}{q_c^2 \zeta_2^2} (\zeta_1^2 \partial_x^2 + \zeta_3^2 \partial_y^2) \right] \\ & \times C_{11}(\boldsymbol{\rho}_1 - \boldsymbol{\rho}_2, t) \\ & - \left[1 - \left[\frac{H_c}{H} \right]^2 \right]^{-1} C_{31}(\boldsymbol{\rho}_1 - \boldsymbol{\rho}_2, t) \\ & + 2\bar{\epsilon} \delta(\boldsymbol{\rho}_1 - \boldsymbol{\rho}_2). \quad (14) \end{aligned}$$

Equation (14) is the first of a hierarchy of equations which couples C_{11} to higher-order correlation functions. We first consider the linear theory in which the nonlinear term C_{31} in (14) is neglected. This is the analogous of the Cahn-Hilliard-Cook theory for spinodal decomposition.⁸ Introducing the transient time dependent structure factor $C(\mathbf{q}, t)$ defined by the Fourier transform (5) of $C_{11}(\boldsymbol{\rho}_1 - \boldsymbol{\rho}_2, t)$ the linear equation becomes

$$\partial_t C(\mathbf{q}, t) = 2 \left[1 - \frac{Q^2}{q_c^2} \right] C(\mathbf{q}, t) + 2\epsilon, \quad (15)$$

where

$$\epsilon \equiv \frac{\bar{\epsilon}}{S} = \frac{2k_B T}{V \chi_a H^2 \xi_2^2 q_c^2} = \frac{2k_B T / V}{\chi_a H^2 \left[1 - \left(\frac{H_c}{H} \right)^2 \right]}. \quad (16)$$

S is the surface in the x - y plane and V the volume of the sample. The solution of (15) is given by

$$C(\mathbf{q}, t) = C(\mathbf{q}, 0) e^{2(1 - Q^2/q_c^2)t} + \frac{\epsilon}{1 - Q^2/q_c^2} (e^{2(1 - Q^2/q_c^2)t} - 1). \quad (17)$$

The same linear analysis can be carried out for $H < H_c$. The solution (17) is also valid in this other situation in which q_c^2 and ϵ become negative quantities, provided that a minus sign is included in the time scale. All the \mathbf{q} modes are then stable and the asymptotic solution for $t \rightarrow \infty$ gives the equilibrium fluctuations.¹² They have an Ornstein-Zernique form

$$C_{\text{eq}}(\mathbf{q}) = \frac{\epsilon}{Q^2/q_c^2 - 1} = \frac{2k_B T / V}{\chi_a H^2 \xi_2^2 (Q^2 + \kappa^2)}, \quad (18)$$

where $\kappa^2 = -q_c^2$. Equation (18) identifies κ^{-1} as a correlation length in the x - y plane. It diverges at $H = H_c$ so that the Fréedericksz transition is accompanied by divergent spatial correlations in the x - y plane. On the other hand, if we use Eq. (17) to describe the dynamical relaxation associated with a change of the magnetic field from $H_i < H_c$ to $H_f > H_c$, Eq. (18) with $H = H_i$ may provide the appropriate initial conditions. Indeed, $C(\mathbf{q}, 0)$ in (17) are the initial fluctuations at $t = 0$ of the director field in equilibrium with a field $H_i < H_c$. (In particular H_i can be zero.) In fact, if H_i is not extremely close to H_c , equilibrium fluctuations are well described by a linear theory as given by (18).

When $H > H_c$, (18) identifies stable ($q_c^2 < Q^2$) and unstable modes ($q_c^2 > Q^2$). The unstable modes grow very fast without limit. The fastest growing mode is $\mathbf{q} = 0$. This is the only mode considered in mean field theories.¹⁻³ The stable modes decay to a final finite value $\epsilon / (Q^2/q_c^2 - 1)$. It is interesting to compare (17) with the result of a linear but deterministic theory ($\epsilon = 0$). In a deterministic theory, $C(\mathbf{q}, t)$, for the stable modes, decays to zero as $t \rightarrow \infty$ while for $\epsilon \neq 0$ it has a final finite value reflecting the existence of fluctuations in the final state. However, the asymptotic value of (17) as $t \rightarrow \infty$ is not the correct equilibrium value, because it is associated with a linearization around $\phi = 0$. Final equilibrium fluctuations could be ob-

tained linearizing around the macroscopic final value of ϕ . For the unstable modes, the deterministic theory gives a pure exponential growth. This is not the case in (17). For instance, an effective amplification factor defined by $(1/t) \ln C(\mathbf{q}, t)$ is not a linear function of Q^2 for $\epsilon \neq 0$.

The structure factor $C(\mathbf{q}, t)$ given by (17) is shown in Figs. 1 and 3 for various values of Q and t (Ref. 13). In these figures it is compared with the results of a nonlinear theory which we consider next. The domain of validity of the linear approximation is discussed below in connection with this nonlinear theory.

III. GAUSSIAN DECOUPLING

We now return to Eq. (14) to take into account nonlinear effects. The solution of (14) requires some approximation to truncate the hierarchy of equations for the correlation functions. In a first approximation to this problem we use here a Gaussian decoupling ansatz in which

$$C_{31}(\boldsymbol{\rho}_1 - \boldsymbol{\rho}_2, t) \simeq 3 \langle \phi^2(t) \rangle C_{11}(\boldsymbol{\rho}_1 - \boldsymbol{\rho}_2, t). \quad (19)$$

A Gaussian ansatz for the decay of an unstable state of a spatially homogeneous system has been discussed by Suzuki.⁴ It has also been used in the problem of spinodal decomposition.^{8,14,22} The nonlinearity is introduced through the dynamical evolution of $\langle \phi^2(t) \rangle$ in (19). The merit of the Gaussian theory as compared with the linear one is that $\langle \phi^2(t) \rangle$ incorporates a saturation effect which prevents the unphysical unlimited growth of the unstable modes. The theory gives then a good description of the early stages of the decay of the unstable state and a qualitatively correct description of the approach to equilibrium.¹⁵ Substituting (19) in (14) the equation for the structure factor becomes

$$\partial_t C(\mathbf{q}, t) = 2 \left\{ 1 - \frac{Q^2}{q_c^2} - \frac{3}{2} \left[1 - \left(\frac{H_c}{H} \right)^2 \right]^{-1} \langle \phi^2(t) \rangle \right\} \times C(\mathbf{q}, t) + 2\epsilon, \quad (20)$$

where

$$\langle \phi^2(t) \rangle = \sum_{\mathbf{q}} C(\mathbf{q}, t). \quad (21)$$

Equation (20) shows how the Gaussian approximation leads to saturation. Since $\langle \phi^2(t) \rangle$ grows with time, the range of unstable modes determined by (20) shrinks to zero as $t \rightarrow \infty$. Equations (20) and (21) form a set of coupled self-consistent equations which could be solved numerically. Instead of this we resort here to an approximation in which the sum over \mathbf{q} in (21) is replaced by the leading term $\mathbf{q} = 0$ (Ref. 16): $\langle \phi^2(t) \rangle \simeq C(0, t)$. With this approximation (20) and (21) become

$$\partial_t C(\mathbf{q}, t) = 2 \left[1 - \frac{Q^2}{q_c^2} - 3C_0(t) \right] C(\mathbf{q}, t) + 2\epsilon, \quad (22)$$

$$\partial_t C_0(t) = 2[1 - 3C_0(t)]C_0(t) + 2\epsilon_0, \quad (23)$$

where

$$C_0(t) \equiv \frac{C(\mathbf{q}=0,t)}{2[1-(H_c/H)^2]}, \quad \epsilon_0 \equiv \frac{\epsilon}{2[1-(H_c/H)^2]}.$$

The approximation leading from (20) and (21) to (22) and (23) is based on the fact that $\mathbf{q}=0$ is the fastest growing mode and that the initial condition $C(\mathbf{q},0)$ has also its maximum at $\mathbf{q}=0$, so that $\mathbf{q}=0$ gives the main contribution to (21) for all times. A related approximation based on the assumption of slow spatial variations of the order parameter was used,¹⁷ but within a deterministic analysis, to study the nonlinear relaxation of a spin system when a magnetic field is switched off. Also in this other situation the main contribution to the initial condition (aligned spins) is given by the $\mathbf{q}=0$ mode.¹⁸ Equation (23) gives a mean field Gaussian approximation and (22) gives the first contribution of spatial inhomogeneities within the Gaussian approximation.

The solution of (22) and (23) is given by

$$C_0(t) = C_0(\infty) + \alpha \left[1 - \left[1 + \frac{\alpha}{C_0(\infty) - C_0(0)} \right] e^{\delta_0 t} \right]^{-1}, \tag{24}$$

where

$$C_0(\infty) = \frac{1}{6} [1 + (1 + 12\epsilon_0)^{1/2}], \tag{25}$$

$$\alpha = -\frac{1}{3} (1 + 12\epsilon_0)^{1/2}, \tag{26}$$

$$\delta_0 \equiv 2(1 + 12\epsilon_0)^{1/2}, \tag{27}$$

$$C_0(0) = \frac{\epsilon}{2} \frac{(H/H_c)^2}{1 - (H_i/H_c)^2}, \tag{28}$$

and

$$C(\mathbf{q},t) = C(\mathbf{q},0) \frac{(1-a)}{(1-ae^{-\delta_0 t})} e^{\alpha_1 t} + 2\epsilon \frac{(\alpha_1 + \delta_0)(e^{\alpha_1 t} - 1) - \alpha_1 a(e^{\alpha_1 t} - e^{-\delta_0 t})}{\alpha_1(\alpha_1 + \delta_0)(1 - ae^{-\delta_0 t})}, \tag{29}$$

$$\alpha_1 \equiv 1 - 2 \frac{Q^2}{q_c^2} - (1 + 12\epsilon_0)^{1/2}, \tag{30}$$

$$a \equiv \frac{C_0(\infty) - C_0(0)}{\frac{1}{6} [1 - (1 + 12\epsilon_0)^{1/2}] - C_0(0)}, \tag{31}$$

$$C(\mathbf{q},0) = \frac{\epsilon_i}{\frac{Q^2}{q_{c,i}^2} - 1} = \left[\frac{1 - \left[\frac{H_i}{H_c} \right]^2}{\left[\frac{H}{H_c} \right]^2 - 1} \right]^{-1} \frac{\epsilon \kappa_i^2}{Q^2 + \kappa_i^2}, \tag{32}$$

with

$$q_c^2 = \frac{\chi_a H_c^2}{K_{22}} \left[\left[\frac{H}{H_c} \right]^2 - 1 \right], \quad \kappa_i^2 = \left[\frac{1 - \left[\frac{H_i}{H_c} \right]^2}{\left[\frac{H}{H_c} \right]^2 - 1} \right] q_c^2. \tag{33}$$

The subindex i in (28), (32), and (33) refers to the initial value H_i of the magnetic field. Our solution is thus expressed in terms of ϵ , H_i/H_c , H/H_c , and q_c^2 . Plots of (24) and (29) are shown in Figs. 1–3 for $\epsilon \approx 2 \times 10^{-10}$, $H_i/H_c = (\frac{1}{2})^{1/2}$, $H/H_c = (\frac{3}{2})^{1/2}$, and $q_c^2 \approx 5 \times 10^4$. These

values correspond to typical parameters for a sample of MBBA at room temperature with $T \approx 300$ K, $\chi_a \approx 10^{-7}$, $K_{22} \approx 10^{-6}$ dyn, $S = 1$ cm², $d = 100$ μ m.

We first analyze the behavior of $C(\mathbf{q},t)$ vs t for fixed \mathbf{q} (Fig. 1). In the deterministic limit ($\epsilon=0$), (24) essentially¹⁹ reproduces the standard mean field deterministic result for $\phi^2(t)$ (Refs. 1–3). For a finite value of ϵ , (24) gives a monotonous growth of $C_0(t)$ which saturates at the macroscopic equilibrium value of the angle ϕ . For the other \mathbf{q} modes $C(\mathbf{q},t)$ exhibits a maximum and then decays to a

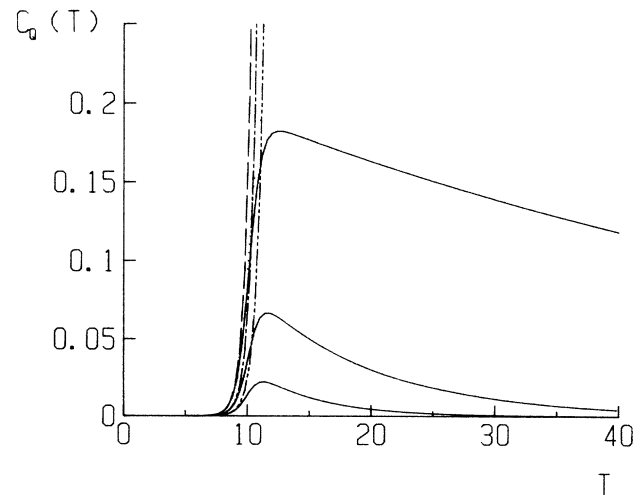


FIG. 1. Structure factor $C(\mathbf{q},t)$ vs time. The dotted line corresponds to the homogeneous mode ($Q=0$). Solid lines from top to bottom correspond to $Q=20, 50,$ and 70 . Results of the linear theory are shown for $Q=20$ (---), $Q=50$ (----), and $Q=70$ (—).

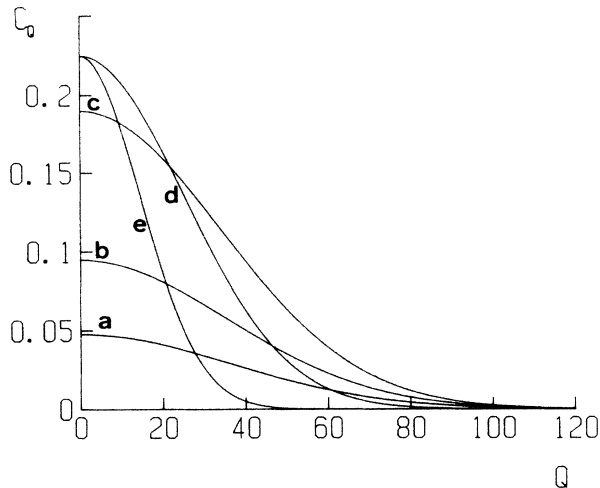


FIG. 2. Structure factor $C(\mathbf{q},t)$ vs Q . (a) $t=9.5$, (b) $t=10$; (c) $t=11$, (d) $t=20$; (e) $t=60$.

value which corresponds to the equilibrium fluctuations in the final state. The maximum of $C(\mathbf{q},t)$ reflects the phenomenon of anomalously large transient fluctuations associated with the decay of an unstable state. In fact it is possible to distinguish in Fig. 1 three characteristic time scales. The first stage of evolution is characterized by a time lag in which the system remains in the vicinity of the unstable state. This is followed by a very fast evolution in which the system leaves the unstable state. In this regime the $\mathbf{q}=0$ mode essentially reaches saturation at a time t_{\max} . For the parameters chosen here, $t_{\max} \simeq 12$ in our dimensionless time scale (see Fig. 1). Also at this time $C(\mathbf{q},t)$ reaches the maximum associated with the anomalous fluctuation phenomenon. The time at which $C(\mathbf{q},t)$ reaches its maximum has a small \mathbf{q} dependence becoming smaller for larger \mathbf{q} . In the final regime there is a slow evolution in which the system attains an equilibrium state characterized by the value of the macroscopic mode and the equilibrium fluctuations for other \mathbf{q} modes.

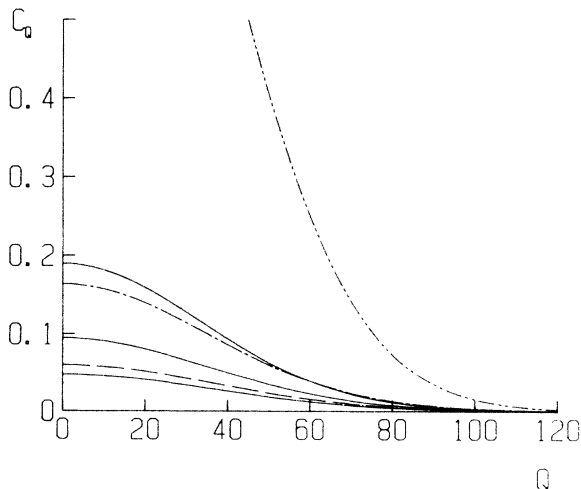


FIG. 3. Structure factor $C(\mathbf{q},t)$ vs Q for the linear and nonlinear theories. Solid lines correspond to the nonlinear theory. Times shown from top to bottom are $t=11$, $t=10$, $t=9.5$. Results of the linear theory are shown at the same times. $t=11$ (—), $t=10$ (---), and $t=9.5$ (-.-.-).

The time t_{\max} can be identified in order of magnitude, with a time slightly larger than the time needed by the system to leave the unstable state. This last time is mathematically characterized as the mean first passage time T to leave the immediate vicinity of the unstable state. This time was calculated in Ref. 11 for the $\mathbf{q}=0$ mode using a scaling theory for the decay of the unstable state. In the asymptotic limit of small ϵ and in the time units used here T is given by

$$T = \frac{1}{2} \ln \left\{ 2\epsilon \left[\left(\frac{H}{H_c} \right)^2 - 1 \right] \right\}^{-1}.$$

For the values of the parameters chosen here $T \simeq 11$ in agreement with our characterization of t_{\max} . In the original laboratory time units

$$T = \frac{\gamma_1}{\chi_a H_c^2} \left\{ 2 \left[\left(\frac{H}{H_c} \right)^2 - 1 \right] \right\}^{-1} \ln \frac{\chi_a H_c^2 V}{4k_B T} \simeq 220 \text{ sec}.$$

In Fig. 1 we also compare the results of our nonlinear calculation with those of the linear theory (17). It is seen that although the linear theory lacks the essential saturation effect it gives a rather good representation of the evolution of $C(\mathbf{q},t)$ roughly up to times $t \lesssim T \lesssim t_{\max}$. This indicates that, as it should be expected, the regime in which the system leaves the unstable state is dominated by linear terms. Related analysis of the validity of linear theory in the problem of spinodal decomposition^{20,21} give a time domain essentially unobservable for spin systems with short-range forces. In the problem discussed here, linear theory is appropriate in a time scale of experimental relevance. We finally note that the equilibrium correlation length κ^{-1} at zero field is d/π . For fixed H and H_c , T grows with d , that is, with the zero field correlation length. However since H_c depends on d , T becomes smaller for larger d and fixed H .

An alternative representation of our results useful in the analysis of scattering experiments is shown in Fig. 2. The analysis of $C(\mathbf{q},t)$ vs \mathbf{q} for fixed t also shows the different stages of evolution discussed earlier. The second stage of evolution associated with fast evolution and anomalous fluctuations is here characterized by a fast growth of the peak of $C(\mathbf{q},t)$ up to $t \simeq 12$ where the smallest \mathbf{q} modes saturate. This saturation time coincides with the time t_{\max} introduced earlier. In the last stage of evolution the height of the peak of $C(\mathbf{q},t)$ remains essentially fixed, but the width of $C(\mathbf{q},t)$ narrows with time indicating the decay of fluctuations to the equilibrium value. The maximum of $C(\mathbf{q},t)$ as a function of time implies a pulse structure which gives rise to the crossing of the tails of the structure function that appears in Fig. 2. Such crossing of tails is characteristic of nonlinear effects.^{21,22} In Fig. 3 we also plot the linear results indicating again the validity of linear theory for $t \lesssim T \lesssim t_{\max}$ with larger discrepancies for small \mathbf{q} . No crossing of tails exists for the linear theory since $C(\mathbf{q},t)$ grows monotonically with t for any \mathbf{q} .

We finally note that the presence of possible dust particles or defects in wall anchoring might modify the ideal situation described here. It is also interesting to note that an important amplification of thermal fluctuations has been discussed in the related problem of a Fréedericksz transition involving splay and bend in relating magnetic field.²³ Such steady-state periodic amplification of the fluctuations of the amplitude of the most unstable mode seem, however, to be related to the important combined

effect of a periodic driving force and small thermal fluctuations.²⁴

ACKNOWLEDGMENTS

Partial financial support from CAICYT Project 361/84 (Spain) and from United States—Spanish Cooperative Research Program Grant No. CCB8402025 is acknowledged.

*Present address: Physics Department, University of Texas at Austin, TX 78712.

¹F. Brochard, P. Pieranski, and E. Guyon, *Phys. Rev. Lett.* **28**, 1681 (1972); *J. Phys. (Paris)* **34**, 35 (1973).

²P. G. de Gennes, *The Physics of Liquid Crystals* (Clarendon Press, Oxford, 1975).

³S. Chandrasekhar, *Liquid Crystals* (Cambridge University Press, Cambridge, 1977).

⁴M. Suzuki, in *Order and Fluctuations in Equilibrium and Nonequilibrium Statistical Mechanics*, G. Nicolis, G. Dewel, and J. W. Turner (Wiley, New York, 1981).

⁵P. Tartaglia, in *Stochastic Processes Applied to Physics*, edited by L. Pesquera and M. A. Rodriguez (World Scientific, Singapore, 1985).

⁶F. Lonberg, S. Fraden, A. J. Hurd, and R. T. Meyer, *Phys. Rev. Lett.* **52**, 1903 (1984). Y. W. Hui, M. R. Kuzma, M. San Miguel, and M. M. Labes, *J. Chem. Phys.* **83**, 288 (1985).

⁷E. Guyon, R. Meyer, and J. Salan, *Mol. Cryst. Liq. Cryst.* **54**, 261 (1979).

⁸J. D. Gunton, M. San Miguel, and P. S. Sahni, in *Phase Transitions and Critical Phenomena*, edited by C. Domb and J. L. Lebowitz (Academic, New York, 1983), Vol. 8.

⁹We restrict ourselves here to situations in which the coupling of the director and velocity fields can be neglected. In the analysis of Refs. 6 and 7, this coupling plays a similar role to the conservation law of the number of atoms of a given species in the problem of the spinodal decomposition of an alloy. The coupling seems to be responsible for the existence of transient spatial structures. In our analysis with no coupling of director and velocity fields the problem is analogous to the dynamics of an order-disorder transition with no conservation law for the order parameter (see Ref. 8).

¹⁰See the Appendix of Ref. 11.

¹¹F. Sagués and M. San Miguel, *Phys. Rev. A* **32**, 1843 (1985); see also M. San Miguel, *ibid.* **32**, 3811 (1985).

¹²We recall that as mentioned before the equilibrium fluctuations of n_z are not taken into account in our approach which focuses primarily on unstable behavior.

¹³ $C(\mathbf{q}, t)$ is not an isotropic function since it depends separately on q_x and q_y . In practice $K_{11} \simeq K_{33}$ and we average out the

small anisotropy by plotting $C(\mathbf{q}, t)$ as a function of Q .

¹⁴J. S. Langer, *Ann. Phys. (N.Y.)* **65**, 53 (1971).

¹⁵As discussed elsewhere (see Ref. 8) it is obvious that a quantitatively correct description of the late approach to equilibrium cannot be based on a single peaked Gaussian distribution. In systems without spatial inhomogeneities the Gaussian decoupling is equivalent to a perturbation in ϵ to order ϵ^2 . This is known to be a singular perturbation theory which breaks down at late times. A resummation of dominant terms in the expansion can be obtained within a scaling theory (Ref. 4). It should also be mentioned that within the Gaussian approximation, the transient fluctuations of the $\mathbf{q}=0$ mode only exhibit the first part of the anomalous fluctuation phenomenon, namely, the variance of $\theta_{m=0, q=0}^{2(t)}$ shows a large growth up to a maximum value. The second stage in which this variance decreases to a small final equilibrium value is not reproduced by a Gaussian approximation.

¹⁶In fact, $\mathbf{q}=0$ only appears in the limit $S \rightarrow \infty$.

¹⁷K. Binder, *Phys. Rev. B* **8**, 3423 (1973).

¹⁸This approximation could be less justified in the study of the transient dynamics of a spin system quenched at zero field from a high temperature phase to a final temperature below the critical one. In this case, the initial structure factor has not a maximum at $\mathbf{q}=0$, but rather it is independent of \mathbf{q} . Initial fluctuations on a small spatial scale are important in the description of the initial stages of the process in which such fluctuations become amplified.

¹⁹A difference in numerical factors in α and $C_0(\infty)$ is due to the Gaussian decoupling. This difference disappears if the factor 3 in (19) is replaced by 1.

²⁰K. Binder, *Phys. Rev. A* **29**, 341 (1984).

²¹M. Grant, M. San Miguel, J. Viñals, and J. D. Gunton, *Phys. Rev. B* **31**, 3027 (1985).

²²M. San Miguel, in *Stochastic Processes Applied to Physics* (Ref. 5).

²³F. Brochard, L. Léger, and R. B. Meyer, *J. Phys. (Paris)* **C1**, 209 (1975).

²⁴F. de Pasquale, Z. Racz, M. San Miguel, and P. Tartaglia, *Phys. Rev. B* **30**, 5228 (1984).