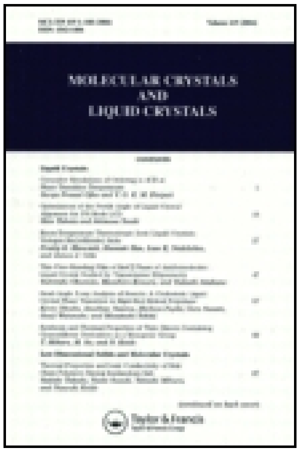

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A Stochastic Approach to Transient Structures and Hydrodynamic. Effects in the Magnetic Freedericksz Transition

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A STOCHASTIC APPROACH TO TRANSIENT STRUCTURES AND HYDRODYNAMIC. EFFECTS IN THE MAGNETIC FREEDERICKSZ TRANSITION

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Abstract The role of hydrodynamic couplings and fluctuations on the transient dynamics of the magnetic Freedericksz transition is analyzed for three different geometries. An anisotropic response for the transverse orientational fluctuations of the director at finite wave numbers and for an arbitrary velocity field is also described.

INTRODUCTION

There is broad experimental evidence of the formation of transient spatial structures during the magnetic Freedericksz transition in nematic liquid crystals[1,2]. This phenomenon is an interesting case of the more general problem of pattern formation occurring during the decay of unstable states in which the interplay between nonlinearities and fluctuations is crucial. This problem appears in a variety of systems and has been extensively studied using different techniques of nonequilibrium statistical mechanics[3,4,5]. Within this general context, the aim of this paper is use the formalism developed by San Miguel and Sagués[5] to describe the early stages of the transient dynamics of the magnetic Freedericksz transition of a nematic sample contained between two plates located at $z = \pm d/2$ for three different geometries.

In previous work[6] we have analyzed the possibility of occurrence of two-dimensional transient spatial structures as well as the effect of hydrodynamic couplings on the transverse spatial fluctuations for the twist (T) and homeotropic to planar (B) geometries. For the former case we found that such transient patterns do occur for large enough applied fields. Moreover, the pattern only formed in the x-direction in spite of the systematic inclusion of inhomogeneities in the y-direction. Contrary to what happens in this case, for the B geometry no transient pattern ever occurs. On the other hand, we explicitly found that the transient spatial fluctuations had a largely different dependence on x and y for both geometries yielding an anisotropic effective viscosity, a result that was expected from the validity of the fluctuation-dissipation theorem and that has been obtained with deterministic approaches [2]. This dynamical anisotropy appeared as a result of the hydrodynamical couplings involving the transverse response of the system at non-vanishing wave numbers. However, these results were obtained by imposing a restrictive assumption, namely, that the macroscopic flow occurred only in the direction of the applied magnetic field during the entire reorientation process. Under this restriction it was no longer clear whether the dynamical anisotropy exhibited by the viscosity and the fluctuations is simply a consequence of this assumed flow behaviour, or if it reflects an inherent dynamical anisotropy of the system.

In this paper we remove this restriction and consider an arbitrary velocity field with a full dependence on all the spatial coordinates. In addition to the T and B geometries, we also consider the splay (S) geometry and calculate analytically the behaviour of the transient structure factor in these three configurations. We still find that for B no transient periodic pattern ever occurs, but for T and S we find that a periodic structure emerges for finite wave numbers associated with the maxima of the corresponding dynamic structure factors. We find that the hydrodynamic couplings shift the position of these maxima toward lower values of the wave number and a dynamical anisotropic response for transverse orientational fluctuations is again found and expressed in terms of an effective viscosity with a different dependence on the transverse coordinates for each geometry. We also calculate the mean first-passage time (MFPT) for the three geometries with and without hydrodynamical effects and show that these couplings decrease the value of the MFPT.

The stochastic dynamics of transient pattern formation in nematics for the Splay geometry was first formulated by Sagués and Arias[7]. However, in their treatment there is an inconsistency in the choice of n_y and n_z as the relevant components of the director $\mathbf{n}$, and the assumption that the director's initial orientation $\mathbf{n}^0$ is along the x-axis and that the magnetic field **H** is in the z direction. Actually, if as usual one assumes that the whole reorientation process occurs in a plane, under the above conditions and according to the definition of the magnetic torque, this should be the xz plane and the variables describing the reorientation should be n_x and n_z, apart from the velocity field. The resulting dynamical equations for these two sets of components of $\vec{n}$ are in general different and they are equivalent only if the system exhibits an additional rotational symmetry around the z axis. Here we remove this inconsistency and use the stochastic description of Sagués and Arias for the variables n_x and n_z. As we show below the use of these variables does not alter the prediction of the existence of a pattern for this geometry; however, the values of the wave number associated with the maximum of the transient structure factor are different than those in reference 7.

BASIC EQUATIONS

Our general starting equations for the director $n_\beta(\mathbf{r},t)$ and velocity $v_\beta(\mathbf{r},t)$ fields are[5]

$$\partial_t n_\beta = (1/\gamma_1)(\delta F/\delta n_\beta) + \Gamma_{\beta\gamma}(\mathbf{n})(\delta F/\delta v_\gamma) + \zeta_\beta(\mathbf{r},t), \tag{1}$$

$$\partial_t v_\beta = L_{\beta\gamma}(\mathbf{n})(\delta F/\delta v_\gamma) - \Gamma_{\beta\gamma}(\mathbf{n})(\delta F/\delta n_\gamma) + \partial_\alpha \Omega_{\alpha\beta}(\mathbf{r},t), \tag{2}$$

where the symbols $\delta/\delta n_\alpha$, $\delta/\delta v_\alpha$ denote the functional derivatives of the free energy functional F

$$F = \tfrac{1}{2}\int d\mathbf{r}\ K_{\alpha\beta\gamma\delta}\ \partial_\beta n_\alpha \partial_\delta n_\gamma - \tfrac{1}{2}\int d\mathbf{r}\ \chi_a (n_\alpha H_\alpha)^2 + \tfrac{1}{2}\int d\mathbf{r}\ \rho v^2. \tag{3}$$

The first term expresses the Oseen-Frank distorsion free energy in terms of the Frank's elastic constants K_1, K_2 and K_3 associated, respectively, with splay, twist and bend elastic deformations. Here

$$K_{\alpha\beta\gamma\delta} = K_1(\delta_{\alpha\delta} - n_\alpha n_\delta)(\delta_{\beta\gamma} - n_\beta n_\gamma) + \\ + K_2 \varepsilon_{\alpha\beta\mu}\ \varepsilon_{\gamma\delta\nu}\ n_\mu n_\nu + K_3(\delta_{\alpha\gamma} - n_\alpha n_\gamma) n_\beta n_\delta \tag{4}$$

and $\varepsilon_{\alpha\beta\mu}$ is the totally antisymmetric Levi-Civita tensor. The second term is the magnetic free energy given in terms of the anisotropic part of the magnetic susceptibility χ_a and the magnetic field H_α. The hydrodynamic contribution is represented by the third term, where ρ is the mass density. Equations (1) and (2) describe the coupled dynamics of the director and the velocity in terms of the free energy (3) and of the kinetic operators $\Gamma_{\beta\gamma}$ and $L_{\beta\gamma}$, which in turn are given in terms of the viscosity coefficients γ_1, γ_2, ν_1, ν_2, ν_3, $\lambda = -\gamma_2/\gamma_1$:

$$\Gamma_{\beta\gamma}(\mathbf{n}) = (1/2\rho)((\lambda + 1)n_\alpha\partial_\alpha\delta_{\beta\gamma} + (\lambda - 1)n_\alpha\partial_\beta\delta_{\alpha\gamma}), \tag{5a}$$

$$L_{\beta\gamma}(\mathbf{n}) = \partial_\alpha M_{\alpha\beta\delta\gamma}(\mathbf{n})\partial_\delta, \tag{5b}$$

$$M_{\alpha\beta\gamma\delta}(\mathbf{n}) = (1/\rho^2)(2(\nu_1+\nu_2-2\nu_3)n_\alpha n_\beta n_\gamma n_\delta + \nu_2(\delta_{\beta\delta}\delta_{\alpha\gamma} + \delta_{\alpha\delta}\delta_{\beta\gamma}) + (\nu_3-\nu_2)(n_\alpha n_\gamma\delta_{\delta\beta} + n_\alpha n_\delta\delta_{\gamma\beta} + n_\beta n_\gamma\delta_{\delta\alpha} + n_\beta n_\delta\delta_{\gamma\alpha}). \tag{5c}$$

The noise sources $\zeta_\beta(\mathbf{r},t)$ and $\partial_\alpha\Omega_{\alpha\beta}(\mathbf{r},t)$ are Gaussian white noise with zero mean and they satisfy the following fluctuation-dissipation relations

$$\langle\zeta_\beta(\mathbf{r},t)\zeta_\gamma(\mathbf{r}',t')\rangle = (2K_BT/\gamma_1)\delta(\mathbf{r} - \mathbf{r}')\delta_{\beta\gamma}\ \delta(t - t'), \tag{6a}$$

$$\langle\partial_\alpha\Omega_{\alpha\beta}(\mathbf{r},t)[\partial_\delta\Omega_{\delta\gamma}(\mathbf{r}',t')]^T\rangle = -2K_BT\ L_{\beta\gamma}\delta(\mathbf{r} - \mathbf{r}')\delta(t - t')\ , \tag{6b}$$

which guarantee that the equilibrium distribution associated with Eqs.(1) and (2) has the canonical form

$$P_{EQ}[\mathbf{n}(\mathbf{r}),\mathbf{v}(\mathbf{r})] \propto \exp[-F/K_BT]\ . \tag{7}$$

AMPLITUDE EQUATION

The reorientational dynamics of the director will be described by the time evolution of the amplitudes of the discrete Fourier modes of an appropriate angle along the z-axis. The key idea of our development is to reduce the dynamical equations to a normal form amplitude equation for the amplitude of the unstable mode. In this way we reduce the problem posed by the system of stochastic partial differential equations (1) and (2) to an ordinary stochastic differential equation for a scalar variable. To this end we first specialize Eqs.(1) and (2) to the B, T and S geometries. For the T-geometry the director is initially aligned along the x-axis, $\mathbf{n}^0$ =

(1,0,0), and the magnetic field is applied along the y-axis. The angle between $\mathbf{n}^0$ and $\mathbf{n}$ at a later time is denoted by ϕ. In the B transition $\mathbf{n}^0$ = (0,0,1) and the magnetic field is applied along the x-axis, whereas for the S-geometry $\mathbf{n}^0$ = (1,0,0) and the magnetic field is along z-axis. For S and B, θ is the angle between $\mathbf{n}^0$ and $\mathbf{n}$. In all cases we assume strong anchoring conditions at the plates and stick boundary conditions for the velocity field which imply that $\mathbf{v}$ vanishes at the plates, but not its first time derivative. This boundary condition on $\mathbf{v}$ is not the most realistic one, but since it what follows we shall only consider the mode m = 0, this approximation will be sufficient for our purpose. As mentioned before, we keep the full dependence of $\mathbf{n}$ and $\mathbf{v}$ on (x,y,z) and the direction of $\mathbf{v}$ is arbitrary.

Following the method of reference 7, we make two approximations. First, we use a minimal coupling scheme of the dynamical equations in which the $\mathbf{n}$ dependence of $\Gamma_{\beta\gamma}$ and $L_{\beta\gamma}$ is approximated by replacing $\mathbf{n}$ by $\mathbf{n}^0$. This procedure retains the initial coupling between $\mathbf{n}$ and $\mathbf{v}$ which is essential in the initial stages of evolution, and amounts to linearize the dinamical equations. Second, we also make the approximation of negligible inertia in which the velocity follows instantaneously the director dynamics, i.e., $d_t v_i = 0$ (i = x, y, z). With these approximations Eqs.(1) and (2) are converted into a closed equation for the deformation angle ϕ or θ. This resulting equation is more conveniently expressed introducing discrete Fourier transforms of the type

$$v_\alpha(x,y,z,t) = \sum_{\mathbf{q}} \sum_{m} v_{\mathbf{q},m}(t)\, e^{i\mathbf{q}\cdot\rho} \sin(2m+1)(\pi z/d), \qquad (8a)$$

$$\theta(x,y,z,t) = \sum_{\mathbf{q}} \sum_{m} \theta_{\mathbf{q},m}(t)\, e^{i\mathbf{q}\cdot\rho} \cos(2m+1)(\pi z/d), \qquad (8b)$$

for ϕ, n and the noise sources, which are compatible with the assumed boundary conditions. Here ρ = (x,y) and the index m numbers the discrete modes in the z-direction and q is the continuous wave number in the transverse plane. For small deformation angles the resulting amplitude equation reads

$$\dot{\theta}^{\alpha}_{\mathbf{q}m}(t) = \Gamma^{\alpha}_{m}(\mathbf{q})\ \theta^{\alpha}_{\mathbf{q}m}(t) + \eta^{\alpha}_{\mathbf{q}m}(t), \qquad \alpha = S,\ T,\ B, \tag{9}$$

with the amplification factor $\Gamma^{\alpha}(\mathbf{q}) \equiv W^{\alpha}(\mathbf{q})/\bar{\gamma}^{\alpha}(\mathbf{q})$ where

$$W^{B}_{\mathbf{q}m} = \chi_a H^2 - K_1 q_x^2 - K_2 q_y^2 - K_3 (2m+1)^2 \frac{\pi^2}{d^2}, \tag{10a}$$

$$W^{T}_{\mathbf{q}m} = \chi_a H^2 - K_3 q_x^2 - K_1 q_y^2 - K_2 (2m+1)^2 \frac{\pi^2}{d^2}, \tag{10b}$$

$$W^{S}_{\mathbf{q}m} = \chi_a H^2 - K_3 q_x^2 - K_2 q_y^2 - K_1 (2m+1)^2 \frac{\pi^2}{d^2}. \tag{10c}$$

There are several features of Eq. (9) which is important to point out. First, the quantity $1/\bar{\gamma}^{\alpha} \equiv 1/\gamma_1 + f^{\alpha}(\mathbf{q},m)$ is an effective wave number dependent viscosity coefficient which contains the effect of the hydrodynamic coupling in the dynamics. It actually has an asymmetric dependence on q_x and q_y since the correction term $f^{\alpha}(\mathbf{q},m)$ turns out to have the general form

$$f^{\alpha}(\mathbf{q},m) = \frac{g_1^{\alpha} q_x^2 + g_2^{\alpha} q_y^2 + g_3^{\alpha}}{h_1^{\alpha} q_x^2 + h_2^{\alpha} q_y^2 + h_3^{\alpha}}, \qquad \alpha = S,\ T,\ B. \tag{11}$$

The coefficients g_i^{α}, h_i^{α} $(i = 1, 2, 3)$ depend only on the viscosity coefficients, are different for each geometry and their explicit form is irrelevant for our discussion. Using the values for the viscosities of MBBA at 25^0C[8], in Fig. 1 we have plotted $\bar{\gamma}^{\alpha}$ as a function of the dimensionless wave number $Q_{\beta} \equiv q_{\beta}/(2m+1)(\pi/d)$, ($\beta$ = x, y) for the three geometries. If we choose the value $Q_j^2 = 2$ the correction factor f^{α} is significant. Actually, $f^{\alpha}(\mathbf{q},m)$ may be as high as 50-60% for the B geometry and up to 14% and 26% for S and T, respectively. This suggest that the anisotropy in the effective viscosity may be measurable. It should be pointed out that these asymmetric effective viscosities have been obtained from purely deterministic treatments[2]; in this respect our results simply reflect that this stochastic description is consistent with the usual ones.

Secondly, one can show that the noise source $\eta_{m,\mathbf{q}}(t)$ in Eq. (9) is a linear combination of the Fourier amplitudes ζ_{α} and $\partial_{\alpha}\Omega_{\alpha\beta}$ and therefore it satisfies a fluctuation-dissipation relation of the same form as (6) but with the effective viscosity $1/\bar{\gamma}^{\alpha}$. This result shows the consistency of a description in terms of an effective viscosity. Third, the positivity of the amplification factor $\Gamma^{\alpha}_{m}(q)$ determines the stability of a given mode. There exists a range of q-modes associated with m = 0 which become unstable when H exceeds a critical value H_c obtained by setting the amplification factor $\Gamma^{\alpha}_{m}(q)$ equal to

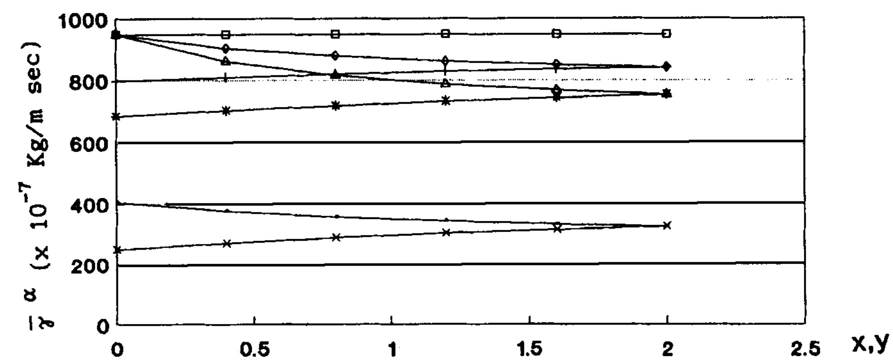

FIGURE 1 Inverse effective viscosity vs. X, Y. Here $X = Q_x^2$ and $Y = Q_y^2$. For $Q_x^2 = 2$, -o- B, -*- T, -+- S. For $Q_y^2 = 2$, -X- B, - - T, -<>- S, -□- γ_1

zero with the result $H_c \equiv K_i\pi^2/\chi_a\, d^2$, i = 1(B), 2(T), 3(S). A transient spatial pattern associated with the mode m = 0 with characteristic periodicity q^0 exists when $\Gamma^{\alpha}_{m}(q)$ has a maximum at $q = q^0 \neq 0$, within the range of unstable modes. This criterion defines the existence of a spatial structure.

DYNAMIC STRUCTURE FACTOR

Transverse spatial fluctuations are described by the time dependent structure factor $S_{mq}(t) \equiv \langle\theta_{m,q}(t)\ \theta_{m,-q}(t)\rangle$, which according to Eq. (9) obeys the equation

$$\partial_t S^{\alpha}_{m,q}(t) = 2\Gamma^{\alpha}_{m}(q)\ S^{\alpha}_{m,q}(t) + (2/\bar{\gamma}^{\alpha})(2K_BT/V), \quad \alpha = S,\ T,\ B. \qquad (12)$$

This equation shows that $\Gamma^{\alpha}_{m}(q)$ indeed determines the rate of initial amplification of fluctuations. The initial conditions $S_{mq}(0)$ are independent of $\Gamma^{\alpha}_{m}(q)$ and correspond to equilibrium fluctuations at t = 0 with an applied field $H<H_c$. They can be consistently obtained as

stationary solutions of Eq.(12) with the result $S_{mq}(0) = -\varepsilon^{\alpha}/W_{i}^{\alpha}(q)$. Here the quantities $W_{i}^{\alpha}(q)$ are obtained by replacing in Eqs.(10) the initial value of the magnetic field H and $\varepsilon^{\alpha} = 2\ K\ TV^{-1}[\chi_{a}H_{c}^{2\alpha}(2m+1)^{2}]^{-1}$, denotes the intensity of thermal noise. Solving Eq.(12) for this initial conditions $S_{mq}(0)$ and using the dimensionless quantities (α = S, T, B, and β = x, y)

$$\tau_{0}^{\alpha} \equiv \frac{\gamma_{1}}{\chi_{a}H_{c}^{2\alpha}\ (2m+1)^{2}}\ , \tag{13a}$$

$$h^{2\alpha}(m) = H^{2\alpha}/[(2m+1)\ H_{c}]^{2}, \qquad s^{\alpha} \equiv t/\tau_{0}^{\alpha}\ , \tag{13b}$$

the solution of Eq.(12) reads

$$S_{Qm}^{\alpha}(s) = S_{Qm}^{\alpha}(0)\ \exp\left\{\left[2\Gamma'^{\alpha}_{m}(Q)/\gamma_{1}\right]\ s\right\} + \\ + \varepsilon^{\alpha}/W'^{\alpha}_{mQ}\left\{\exp\left\{\left[2\Gamma'^{\alpha}_{m}(Q)/\gamma_{1}\right]\ s\right\} - 1\right\}. \tag{14}$$

Here $\Gamma'^{\alpha}_{m}(Q)$ and W'^{α}_{mQ} denote the quantities $\Gamma^{\alpha}_{m}(Q)$ and W^{α}_{mQ} written in terms of the above dimensionless variables and this expression describes the growth of the unstable mode leading to the Freedericksz transition.

If we plot Eq.(14) we get the curves plotted in Figs.2 and 3, for the S and B configurations where, again, we have used the viscosities of MBBA at 25^{o}C. From these graphs we conclude that the maximum of the structure factor for the B configuration is at the origin, whereas for S and T it is located at a finite value along the Q_{x}-axis: $Q^{2}_{max}(S) = 0.2$. Although for the T geometry it is not shown, the maximum is located at $Q^{2}_{max}(T) = 0.18$. This result holds even when the values of the parameters h_{i}^{2} and h^{2} are changed, which amounts to varying the magnitude of the applied field and the scaled time s. In Fig.4 we show the time evolution of the maximum of the structure factor for the S geometry is plotted for the scaled times s = 0.7, 0.8, which in real time correspond to 9.3 and 10.7 sec, respectively, in the presence and abscence of hydrodynamic effects. These values were chosen so that they are comparable with the corresponding MFPT for this geometry, as can be seen from Table I. These curves show that the amplitude of the structure factor increases with time (s)

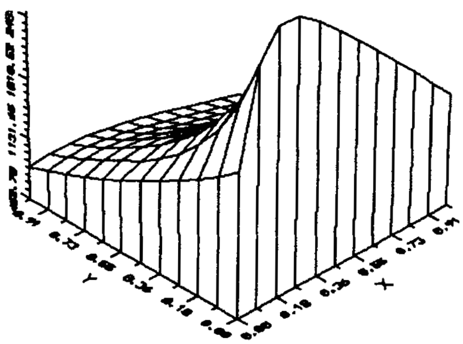

FIGURE 2 $S^{S}_{m=0,q}(t)/\varepsilon^{S}$ for $h_{1}^{2} = 0$, $h^{2} = 8.5$, $s = 0.5$. Here $X = Q_{x}^{2}$ and $Y = Q_{y}^{2}$.

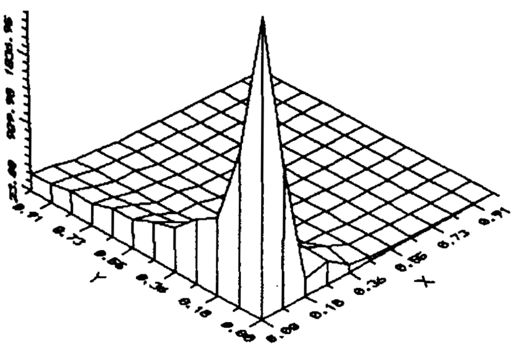

FIGURE 3 $S^{B}_{m=0,q}(t)/\varepsilon^{B}$ for $h_{1}^{2} = 0$, $h^{2} = 5$, $s = 0.2$. Here $X = Q_{x}^{2}$ and $Y = Q_{y}^{2}$.

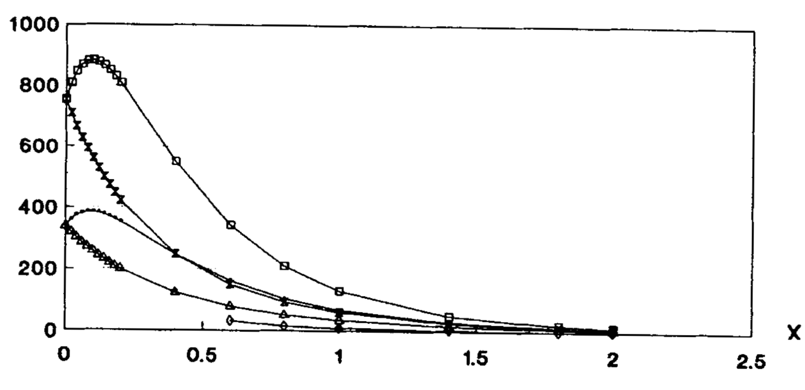

FIGURE 4 Time evolution of the maximum of $S^{s}_{m=0,q}(t)/\varepsilon^{s}$ projected on the Q_{x}^{2} plane, here $X = Q_{x}^{2}$ with $h_{1}^{2} = 0$ and $h^{2} = 5$. With hydrodynamic effects: -□- (s = 0.8), -•- (s = 0.7), Without hydrodynamic effects: -><- (s = 0.8), - - (s = 0.7).

and that when the hydrodynamic couplings are absent the maximum is at the origin, whereas the presence of hydrodynamic effects shift the maximum towards larger values of Q and increase its amplitude. A direct comparison of our result for $Q^2_{max}(S)$ with the values obtained in reference 7 is not possible since our dynamical equations differ from the ones used by Sagués and Arias, as has been pointed out in the Introduction. For their time values we find that the maximum of the transient structure is always at the origin.

Hydrodynamic effects are also manifested in the values of the mean first passage time (MFPT), which may be taken as a measure of the time needed to reach the unstable point for the first time. To calculate this quantity we use an asymptotic expression for the first-passage time distribution that was derived by Haake et al[9] for those cases where the random forces initiating the decay are weak and threshold is sufficiently far from the point of unstable equilibrium. By using this approximation in the Langevin equation (9), one arrives at the following expression for the mean first passage time:

$$T^{\alpha} \simeq (4W^{\alpha}_{qm} / \bar{\gamma}_{\alpha})^{-1} \ln (1/2\varepsilon_{\alpha}) , \quad \alpha = S, T, B. \qquad (15)$$

In TABLE I we give the numerical values of the MFPT obtained from Eq.(15) for MBBA with and without hydrodynamic effects.

TABLE I Mean First Passage Time (sec)

	T_1 (WITH HYD. EFFECTS)	T_2 (WITHOUT HYD. EFFECTS)	τ_{on}	τ_0
SPLAY	18.0991	20.066884	1.8057083	13.542812
TWIST	29.897616	36.19504	3.2051321	24.038491
BEND	3.4390	17.321673	1.3935357	10.451519

Note that the hydrodynamical couplings are present through the effective viscosity $\bar{\gamma}^{\alpha}(q)$. If we estimate numerically this quantity for MBBA, we get the values in column T_1. By replacing the effective viscosity by γ_1, we can calculate the value of the MFPT without hydrodynamic effects; this yields the column T_2. For comparison we have also included in this table the hydrodynamic onset time τ_{on} of the instability, $\tau^{\alpha}_{on} = \tau^{\alpha}_{o} [1/(h^2 - 1)]$ with α = S, T, B, obtained from the dynamical equations (1) by ignoring the fluctuating terms. From these numerical results we confirm that the hydrodynamic couplings make the instability to set in faster. We are not aware of experimental measurements of the MFPT in the magnetic transition to compare with, but it is interesting to note that these numerical estimations are at least comparable with the experimental mesurements of Buka et. al. for transient structures in a nematic sample between two electrodes[10], namely, elapsed times between 0.7 sec and about one minute.

Although for the purpose of determining the existence and the basic physical ingredients at the onset of the observed periodic structures the usual deterministic approach based on an eigenvalue analysis is sufficient, we believe that the stochastic approach introduced in references 5 and 7 is more convenient for describing the time dependence of the characteristic periodicity at later times starting from the homogeneous sample at the initial time.

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