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Metastability and front propagation in the first-order optical Fréedericksz transition

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A general dynamical model for the first-order optical Fréedericksz transition incorporating spatial transverse inhomogeneities and hydrodynamic effects is discussed in the framework of a time-dependent Ginzburg-Landau model. The motion of an interface between two coexisting states with different director orientations is considered. A uniformly translating front solution of the dynamical equations for the motion of that interface is described.

I. INTRODUCTION

The nematic phase of liquid crystals[1] is an interesting system for the theoretical and experimental study of nonequilibrium phenomena such as instabilities, interface motion, and pattern formation. In these systems the orientational dynamics of the molecules is coupled with velocity flows so that in addition to phenomena associated with the dynamics of the relevant macroscopic variable (director vector), the full richness of hydrodynamic instabilities is present. In this context, different aspects of the magnetic Fréedericksz transition have been broadly studied. When the nematic sample is contained between two plates perpendicular to the z axis, the consideration of transverse effects in the x-y director allows the study of interfaces between regions of different orientations. Interface formation and motion for Fréedericksz walls and inversion walls have been considered.[1] Hydrodynamic effects are known to be responsible for the occurrence of transient patterns when the magnetic field is switched to a value larger than the threshold value. Recent experimental[2] and theoretical[3,4] literature is devoted to this subject. Finally, the problem of front propagation into an unstable state[5] including the case[6] of front propagation leading to pattern formation can be also studied in the magnetic Fréedericksz transition.

The magnetic Fréedericksz transition is of second order (supercritical bifurcation), and as a consequence the possibility of metastable states does not exist. New interesting phenomena associated with metastability are expected to occur in the optical Fréedericksz transition (OFT). In this case, and as a manifestation of the large optical nonlinearity, the reorientation of the molecules in the nematic phase can be caused by laser irradiation. The possibility of a first-order transition (subcritical bifurcation) in the optical Fréedericksz transition was predicted by Ong.[7] Experimental observations of hysteresis cycles have been reported with the addition of a magnetic field[8] and also in an all-optical situation.[9] In comparison with the magnetic Fréedericksz transition, the effects of hydrodynamic coupling have not been considered in the theoretical description of the OFT. In addition, transverse effects have been considered[10] only in situations in which the OFT is of second order. In this paper, and paralleling our previous formulation of the magnetic Fréedericksz transition,[3(a),4] we introduce a dynamical model of the first-order OFT that includes transverse effects and hydrodynamic coupling. The problem addressed in this paper from this general formulation is related to metastability and transverse effects. We study the motion of an interface that separates, in the transverse direction, two states of different orientation of the molecules. These two states are within the hysteresis cycle, one being metastable and the other stable. The mathematical description of the equilibrium interface can be borrowed from earlier work on nucleation and phase separation in tricritical systems.[11,12] Interface motion is studied by analogy with other simpler physical situations involving metastability.[13] We find that this motion is described by a uniformly translating front solution of our general equations with a specified velocity. Hydrodynamic coupling is taken into account in a small-wave-number approximation. In the leading order of this approximation, and in our geometry, the hydrodynamic coupling amounts to the redefinition of a viscosity coefficient, so that its effect is a change of the time scale in which the propagating front evolves.

The situation considered in this paper gives an interesting physical realization of the general problem of front propagation in the domain of metastability of subcritical bifurcations.[14] A different but related problem, not considered here, is that of front propagation into an unstable state. Our model of the OFT provides an interesting physical realization for this problem. In fact, our reduction of hydrodynamic effects to an effective viscosity permits a direct application of the results of Ref. 15 to our case.

The outline of the paper is as follows. Section II describes our general formulation of the first-order OFT through stochastic nematodynamic equations and the approximate treatment of the hydrodynamic coupling. Section III contains the application of this formulation to interface motion in the metastable domain. Our main result for front propagation is discussed in Sec. III B. Some mathematical details about the general dynamical model are given in the Appendix.

II. MODEL EQUATIONS

We consider a homeotropically aligned nematic liquid-crystal layer of thickness d, measured along the z axis, and infinite transverse dimensions along the perpendicular directions x and y. A normally incident laser beam, polarized parallel to the place of incidence, is applied to the sample, as represented in Fig. 1. Inside the cell, the orientation of the nematic liquid crystal is given in terms of the director $\mathbf{n}(\mathbf{r})$. In the absence of radiation the director will remain everywhere parallel to the z axis: $\mathbf{n}(\mathbf{r})=\mathbf{n}^0=(0,0,1)$. The light beam causes reorientation assumed to take place essentially in the xz plane.

As is well known, the reorientation of the director is normally coupled to hydrodynamic effects expressed in terms of a velocity field $\mathbf{v}(\mathbf{r})$, which accounts for the translational motion of the molecules of the nematic liquid crystal. This enlarged description of the reorientational dynamics is conveniently analyzed in terms of a generic set of nematodynamic equations for both the director and velocity variables. In terms of a free-energy functional, written here uniquely in terms of the elastic and electromagnetic contributions,

$$F[\mathbf{n}(\mathbf{r})]=\int d\mathbf{r}\tfrac{1}{2}\{k_1(\nabla\cdot\mathbf{n})^2+k_2[\mathbf{n}\cdot(\nabla\times\mathbf{n})]^2 +k_3(\mathbf{n}\times\nabla\times\mathbf{n})^2+\mathbf{E}\cdot\mathbf{D}+\mathbf{B}\cdot\mathbf{H}\}\ , \quad (2.1)$$

these equations read[3(a)]

$$d_t n_\beta=-(1/\gamma_1)\frac{\delta F}{\delta n_\beta}+\rho\Gamma_{\beta\gamma}(\mathbf{n})v_\gamma+\xi_\beta(\mathbf{r},t)\ ,$$
$$d_t v_\beta=\rho L_{\beta\gamma}(\mathbf{n})v_\gamma-\Gamma^{+}_{\beta\gamma}(\mathbf{n})\frac{\delta F}{\delta n_\gamma}-\frac{1}{\rho}\partial_\beta P+\partial_\alpha\Omega_{\alpha\beta}(\mathbf{r},t)\ , \quad (2.2)$$

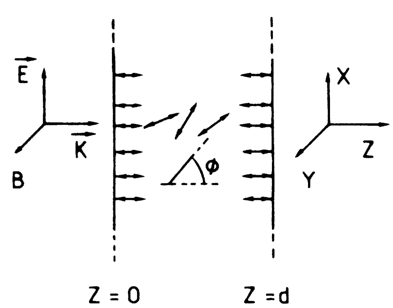

FIG. 1. Homeotropically aligned nematic liquid-crystal sample perpendicularly excited by a laser beam polarized parallel to the plane of incidence. The reorientation of the nematic material is measured in terms of the distortion angle ϕ.

where d_t is the total derivative including convective terms, ρ is the mass density, and P is the pressure. The operators $\Gamma_{\beta\gamma}$ and $L_{\beta\gamma}$ are given in terms of a convenient set of viscosity coefficients γ_1 and γ_2 and ν_1, ν_2, and ν_3 ($\lambda=-\gamma_2/\gamma_1$) as

$$L_{\beta\gamma}(\mathbf{n})=\partial_\alpha M_{\alpha\beta\delta\gamma}(\mathbf{n})\partial_\delta\ ,$$
$$M_{\alpha\beta\gamma\delta}(\mathbf{n})=\frac{1}{\rho^2}[2(\nu_1+\nu_2-2\nu_3)n_\alpha n_\beta n_\gamma n_\delta +\nu_2(\delta_{\beta\delta}\delta_{\alpha\gamma}+\delta_{\alpha\delta}\delta_{\beta\gamma}) +(\nu_3-\nu_2)(n_\alpha n_\gamma\delta_{\delta\beta}+n_\alpha n_\delta\delta_{\gamma\beta} +n_\beta n_\gamma\delta_{\delta\alpha}+n_\beta n_\delta\delta_{\gamma\alpha})]\ , \quad (2.3)$$
$$\Gamma_{\beta\gamma}(\mathbf{n})=\frac{1}{2\rho}[(\lambda+1)n_\alpha\partial_\alpha\delta_{\beta\gamma}+(\lambda-1)n_\alpha\partial_\beta\delta_{\alpha\gamma}]\ .$$

The adjoint operator Γ^+ is used here in the sense of integration by parts and transposing matrix indices. The noise sources ξ_β and $\partial_\alpha\Omega_{\alpha\beta}$ describe thermal fluctuations and have Gaussian white-noise statistics satisfying appropriate fluctuation-dissipation relations

$$\langle \xi_\beta(\mathbf{r},t)\xi_\gamma(\mathbf{r}',t')\rangle=2\frac{k_BT}{\gamma_1}\delta(\mathbf{r}-\mathbf{r}')\delta(t-t')\delta_{\beta\gamma}\ ,$$
$$\langle \partial_\alpha\Omega_{\alpha\beta}(\mathbf{r},t)[\partial_\delta\Omega_{\delta\gamma}(\mathbf{r}',t')]^+\rangle =-2k_BTL_{\beta\gamma}\delta(\mathbf{r}-\mathbf{r}')\delta(t-t')\ . \quad (2.4)$$

Under our assumption of planar reorientation of the director $\mathbf{n}(\mathbf{r})$, the actual state of orientation of the nematic liquid crystal may be described in terms of a distortion angle $\phi(\mathbf{r},t)$

$$\mathbf{n}(\mathbf{r},t)\equiv(\sin\phi(\mathbf{r},t),0,\cos\phi(\mathbf{r},t))\ . \quad (2.5)$$

In what follows we assume that any hydrodynamic flow, eventually originated in the reorientation of the director, will also be limited to the xz plane

$$\mathbf{v}(\mathbf{r},t)\equiv(v_x(\mathbf{r},t),0,v_z(\mathbf{r},t))\ . \quad (2.6)$$

Thermal noise terms introduced in (2.2) are essential in the early stages of the dynamics when the laser beam is switched to an intensity value that causes director reorientation. However, their effect is not important in the last-stage dynamics. We are concerned here with the study of propagating fronts occurring in the late stage of the reorientation process. As a consequence, and for simplicity, we neglect noise terms hereafter.

To proceed further with the analysis of the reorientational dynamics of the nematic liquid crystal, we make the common approximation of negligible inertia, i.e., $d_tv_x=d_tv_z=0$. A closed dynamical equation for the angular variable $\phi(\mathbf{r},t)$ may then be obtained after performing a few manipulations of the general scheme of Eqs. (2.2). First, we invoke the incompressibility condition of the nematic material to get rid of pressure terms. Additionally, we use a minimal coupling approximation[3(a)] that amounts to replacing the $\mathbf{n}$ dependence of the operators Γ and M in (2.3) by their value at the undistorted initial configuration of the director $\mathbf{n}^0$. After completing all this algebra we end up with a single equation for $\phi(\mathbf{r},t)$ written as

$$\partial_t \phi(\mathbf{r},t) = L\frac{\delta F}{\delta \phi} , \tag{2.7}$$

where L is a kinetic operator incorporating all the hydrodynamic effects. It admits the following formal expression:

$$L = -1/\gamma_1 + \tfrac{1}{2}(\lambda+1)E(A^{-1}B\Xi^{-1}-1)A^{-1}C - \tfrac{1}{2}(\lambda-1)D\Xi^{-1}A^{-1}C , \tag{2.8}$$

in terms of the differential operators

$$\begin{aligned} A &\equiv \partial_z[\nu_2(2\partial_x^2+\partial_y^2)+\nu_3(\partial_z^2-\partial_{xz}^2)] , \\ B &\equiv \partial_x[\nu_3(\partial_z^2-\partial_x^2-\partial_y^2)-2\nu_1\partial_z^2] , \\ C &\equiv \tfrac{1}{2}[(\lambda+1)\partial_z^2-(\lambda-1)\partial_x^2] , \\ D &\equiv \partial_x, \quad E \equiv \partial_z, \quad \Xi \equiv (A^{-1}B - D^{-1}E) . \end{aligned} \tag{2.9}$$

To render Eq. (2.7) amenable to solution we need to introduce several approximations. First of all we will restrict our description to only one transverse coordinate $\phi(\mathbf{r},t)=\phi(x,z,t)$. The complicated hydrodynamic coupling is here approximated by keeping only the dominant contribution to the kinetic operator L. To identify this dominant effect we take a small wave-number limit of L along the transverse direction x. It turns out that hydrodynamic effects already appear for distortions of zero wave number in the transverse direction. As a consequence the differential operator L becomes, in the lowest approximation, a pure numerical constant that can be easily interpreted in terms of an effective rotational viscosity γ_1^*

$$L \to -\frac{1}{\gamma_1^*} \equiv -\frac{1}{\gamma_1}\left[1+(1+\lambda)^2\frac{\gamma_1}{4\nu_3}\right] . \tag{2.10}$$

Equation (2.10) is obtained from (2.8) and (2.9) by formally setting $\partial_x=\partial_y=0$ in (2.9). With this limiting procedure, our treatment incorporates hydrodynamic effects in a way similar to what Brochard[16] did for the analogous problem of front waves in the magnetically induced Fréedericksz transition. Higher-order transverse effects due to hydrodynamic coupling can be incorporated in L by a straightforward calculation from (2.8) and (2.9). Such additional contributions to L provide other examples of different front-propagation dynamics. The different dynamics here would be related only to a different kinetic coefficient and not to a different free energy as in the cases considered in Ref. 15. In this paper we restrict ourselves to the lowest approximation for L in which hydrodynamics effects are incorporated by (2.10). In this approximation the hydrodynamic coupling only changes the time scale of evolution.

Before considering a more extensive analysis of the dynamics given by (2.7), we need to express the electromagnetic contribution to the free-energy functional in (2.1) in terms of the orientational angle ϕ. We adopt here the formulation proposed by Ong[7] and also followed by Marquis *et al.*[10] The contribution can be written in terms of the z component of the Poynting vector $S_z(x)$ as

$$F_{\rm em} = \tfrac{1}{2}(\mathbf{E}\cdot\mathbf{D}+\mathbf{B}\cdot\mathbf{H}) = -\frac{S_z(x)n_o}{c_o}\frac{1}{(1-r\sin^2\phi)^{1/2}} , \tag{2.11}$$

where $r\equiv 1-n_o^2/n_e^2$, is given in terms of the ordinary n_o and extraordinary n_e refractive indices of the liquid crystal and c_0 is the speed of light. In what follows we consider a situation corresponding to an infinite plane-wave illumination, so that $S_z(x)$ becomes a constant S. With all the previous considerations, the general equation (2.7) reduces to

$$\partial_t\phi(x,z,t) = \frac{1}{\gamma_1^*}\left[(k_1\partial_x^2+k_3\partial_z^2)\phi + \frac{Sn_o r}{c_o}\frac{\sin\phi\cos\phi}{(1-r\sin^2\phi)^{3/2}}\right] , \tag{2.12}$$

where according to the spirit of a Landau-type expansion only lowest-order terms in ϕ containing spatial derivatives are retained. In Eq. (2.12) k_1 and k_3 are elastic constants associated with splay and bend deformation, respectively. No use has been made here of the common approximation in which all elastic constants are taken to be equal.[10]

The complicated nonlinearities of (2.12) originate in the coupling between the director and the incident electromagnetic field. Equation (2.12) can be simplified by an expansion in powers of the distortion angle ϕ. This would be most adequate when referring to laser intensities close to the threshold of a second-order or weakly first-order Fréedericksz transition. In order to find typical features of the first-order transition in which we are interested here, we need to expand the nonlinear term in (2.12) in powers of ϕ up to ϕ^5. However, the expansion only makes sense for a restricted range of values of r, as discussed in Sec. III. We note that no approximation based on the value of r is invoked. Proceeding in this way, Eq. (2.12) is approximated as (see Appendix)

$$\partial_t\phi(x,z,t) = \frac{1}{\gamma_1^*}\left\{(k_1\partial_x^2+k_3\partial_z^2)\phi + \frac{Sn_o r}{c_o}\left[\phi+(\tfrac{3}{2}r-\tfrac{2}{3})\phi^3 + (\tfrac{2}{15}-\tfrac{3}{2}r+\tfrac{15}{8}r^2)\phi^5\right]\right\} . \tag{2.13}$$

A final transformation of this equation will enable us to concentrate specifically on transverse effects along the x direction by selecting a particular z dependence of the director angle $\phi(x,z,t)$. Prescribing strong anchoring boundary conditions at the limiting plates, i.e., $\phi(x,z,t)|_{z=o,d}=0$, and in the vicinity of the threshold intensity, we may assume that the z dependence is well approximated by the most unstable z-Fourier mode of $\phi(x,z,t)$. Thus, with the ansatz

$$\phi(x,z,t)=\theta(x,t)\sin\frac{\pi z}{d}\ , \tag{2.14}$$

Eq. (2.13) becomes (see Appendix)

$$\partial_t\theta(x,t)=\frac{1}{\gamma_1^*}\left[\left[k_1\partial_x^2-k_3\frac{\pi^2}{d^2}\right]\theta+\frac{Sn_o r}{c_o}[\theta+\tfrac{3}{4}(\tfrac{3}{2}r-\tfrac{2}{3})\theta^3+\tfrac{5}{8}(\tfrac{2}{15}-\tfrac{3}{2}r+\tfrac{15}{8}r^2)\theta^5\right]\ . \tag{2.15}$$

An appropriate dimensionless version of Eq. (2.15) will be the central equation used in our analysis of propagating fronts. Defining two basic length and time units, respectively, as

$$u\equiv x/(k_1d^2/k_3\pi^2)^{1/2},\quad \tau\equiv t/(\gamma_1^*d^2/k_3\pi^2)\ , \tag{2.16}$$

Eq. (2.15) becomes

$$\partial_\tau\theta(u,\tau)=\partial_u^2\theta+(s-1)\theta+s[\tfrac{3}{4}(\tfrac{3}{2}r-\tfrac{2}{3})\theta^3+\tfrac{5}{8}(\tfrac{2}{15}-\tfrac{3}{2}r+\tfrac{15}{8}r^2)\theta^5]\ , \tag{2.17}$$

where a reduced illumination parameter has been defined $s\equiv S/S_c$ in terms of the threshold value of the optical Fréedericksz transition S_c given by

$$S_c\equiv\frac{k_3\pi^2}{d^2}\frac{c_0}{n_o r}\ . \tag{2.18}$$

In the formulation of Eq. (2.17) the effect of hydrodynamic coupling is buried in the time scale considered. Such hydrodynamic effects can be quantitativly important. For example, for standard values[1] of γ_1, λ, and ν_3 for *N*-(*p*-methoxybenzylidine)-*p*- butylaniline (MBBA) one finds $(\gamma_1^*/\gamma_1)=0.23$, that is a viscosity reduction close to 80%.

III. INTERFACE PROFILE MOTION

Our model equation (2.17) for the reorientational dynamics admits the possibility of a first-order transition. Before such a transition occurs, the undistorted state becomes metastable for certain values of the incident laser intensity. The range of metastability is described in Sec. III A. Section III B considers the motion of a transverse interface created between the metastable undistorted state and a stable state of nonzero reorientation.

A. Phase diagram and metastability

Equation (2.17) is easily converted into a convenient potential form

$$\partial_\tau\theta(u,\tau)=-\frac{d}{d\theta}V(\theta)+\partial_u^2\theta\ , \tag{3.1}$$

in terms of a symmetric potential $V(\theta)$ generically written as

$$V(\theta)=-\frac{a}{2}\theta^2-\frac{b}{4}\theta^4+\frac{c}{6}\theta^6\ , \tag{3.2}$$

where the successive higher-order coefficients are given, in our particular situation, by

$$a\equiv s-1\ ,\quad b\equiv s\tfrac{3}{4}(\tfrac{3}{2}r-\tfrac{2}{3})\ ,\quad c\equiv -s\tfrac{5}{8}(\tfrac{2}{15}-\tfrac{3}{2}r+\tfrac{15}{8}r^2)\ . \tag{3.3}$$

The potential $V(\theta)$ enables us to examine the exchange of stability between the trivial solution $\theta=0$, corresponding to a nondistorted configuration, and those homogeneous stable or unstable stationary reorientational states characterized by nonzero values of the director angle amplitude θ. These states correspond to the extrema of $V(\theta)$.

The lowest-order coefficient a identifies the condition for instability of the state of zero distortion. The nature of the transition is then determined by the sign of the fourth-order parameter b. If b is negative, the Fréedericksz transition will be second order, the bifurcation of the reference solution $\theta=0$ being in this case supercritical. If b is positive, the bifurcation is subcritical, or in other words, we are facing a first-order transition with its eventual sequels of metastability and hysteresis effects. In our analysis we are interested in the propagation of fronts in situations of metastability. The condition $b>0$ for the existence of a first-order transition implies that $r>\frac{4}{9}$. In addition, stability considerations require $c>0$, so that

$$\tfrac{2}{15}(3+\sqrt{5})<r<\tfrac{2}{15}(3+\sqrt{5})\ . \tag{3.4}$$

Therefore a first-order transition is described by our model in a restricted range of values of the refractive index given by

$$\tfrac{2}{15}(3+\sqrt{5})>r>\tfrac{4}{9}\ . \tag{3.5}$$

This range of values for r is of the correct order of magnitude for common materials.[7] For many materials $r<\frac{4}{9}$, so that when a first-order transition occurs it will generally tend to be weakly first order. For these values of the refractive index metastability occurs for $\tilde{a}<a<0$, where $\tilde{a}=-b^2/4c$. In terms of the incident laser intensity, the domain of metastability is given by $S_i<S<S_c$, where

$$S_i=S_c\left[1+\frac{1}{4}\frac{[\frac{3}{4}(\frac{3}{2}r-\frac{2}{3})]^2}{\frac{5}{8}(-\frac{2}{15}+\frac{3}{2}r-\frac{15}{8}r^2)}\right]^{-1}\ . \tag{3.6}$$

Within the domain of metastability, the undistorted $\theta=0$ is globally stable for $a<a^*=-\frac{3}{4}(b^2/4c)$ and it is only metastable for $a>a^*$. In other words, tristability occurs at $a=a^*$ or

$$S_T=S_c\left[1+\frac{3}{16}\frac{[\frac{3}{4}(\frac{3}{2}r-\frac{2}{3})]^2}{\frac{5}{8}[-\frac{2}{15}+\frac{3}{2}r-\frac{15}{8}r^2]}\right]^{-1}\ . \tag{3.7}$$

For $S>S_T$ the undistorted state becomes metastable and at $S=S_c$ it becomes unstable. The above discussion is summarized in Fig. 2.

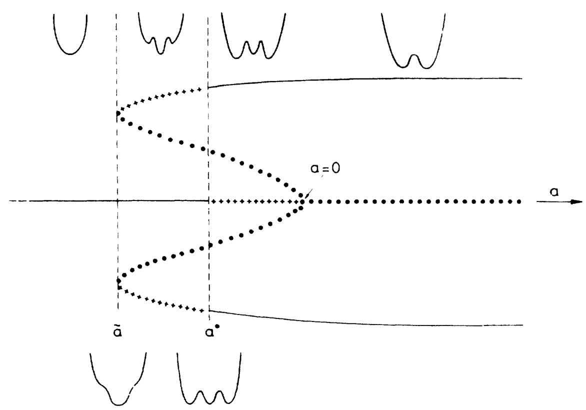

FIG. 2. Bifurcation diagram corresponding to the sixth-order potential $V(\theta)$ defined in (3.2). Solid lines denote stable states. Dots and crosses stand for unstable and metastable states, respectively. The region analyzed in the text corresponds to negative values of a up to $\bar{a}$.

B. Front propagation

Interfaces, metastability, and nucleation for a symmetric subcritical bifurcation as the one of Fig. 2 have been studied in Refs. 11 and 12. For the three-dimensional systems considered in these references, an interface defining a droplet of the state $\theta=0$ surrounded by $\theta\neq 0$ will grow or shrink even for $S=S_T$ due to curvature effects. In the optical Fréedericksz transition transverse effects are essentially one dimensional and such an interface at $S=S_T$ is stable. We will consider the form of such an interface and its motion for $S\neq S_T$ within the domain of metastability. Our analysis of one-dimensional interface motion follows closely that of Ref. 13. A uniformly translating front solution of Eq. (3.1) may be sought in the form $\theta(u-v\tau)=\theta(w)$, which satisfies

$$v\frac{d}{dw}\theta+\frac{d^2}{dw^2}\theta=\frac{d}{d\theta}V(\theta)\ , \tag{3.8}$$

or in terms of the nonzero stationary homogeneous solutions of Eq. (3.1), i.e., the extrema of the potential $V(\theta)$, denoted $=\pm\theta_\pm$,

$$v\frac{d}{dw}\theta+\frac{d^2}{dw^2}\theta -c\theta(\theta-\theta_+)(\theta+\theta_+)(\theta-\theta_-)(\theta+\theta_-)=0\ , \tag{3.9}$$

where

$$\theta_\pm^2\equiv\frac{b}{2c}\pm\frac{1}{2c}(b^2+4ac)^{1/2}\ . \tag{3.10}$$

$\pm\theta_+$ are minima of the potential associated with locally stable states of nonzero reorientation.

To avoid the trivial degeneracy originated in the symmetry of the potential $V(\theta)$, we arbitrarily refer to the positive branch of the bifurcation diagram, or in other words, to the positive states of stable (θ_+) and unstable reorientations (θ_-). Interface solutions connecting the two equivalent states $\pm\theta_+$ are not analyzed here.[17] At the equilibrium point $S=S_T$ where $V(0)=V(\theta_+)$, a stable interface profile connecting the two equivalent stable solutions $\theta=0$ and $\theta=\theta_+$, respectively, at $u=\pm\infty$, is given[11,12] by a solution of (3.9) with $v=0$:

$$\theta(u)|_{S=S_T}=\frac{1}{\sqrt{2}}\theta_+(S=S_T)[1+\tanh(u/\xi)]^{1/2}\ , \tag{3.11}$$

where the width of the interface is given by

$$\xi^2(S=S_T)=\frac{3}{(c\theta_+^4)|_{S=S_T}}=\left|\frac{S_T}{S_c}-1\right|^{-1}\ , \tag{3.12}$$

and $\theta_+^2(S=S_T)=3b/4c$. When $S>S_T$ one expects a motion of the interface (3.11) propagating the metastable state $\theta=0$ into the absolutely stable state $\theta=\theta_+$. The interesting feature is that such motion is described by a uniformly translating front of the form (3.11), which is a solution of (3.9). Specifically we find that a solution of (3.9) is

$$\theta(u=v\tau)=\frac{1}{\sqrt{2}}\theta_+(s)\{1+[\tanh(u-v\tau)]/\xi(s)\}^{1/2}\ , \tag{3.13}$$

where

$$\xi^2=(6c/b)[b+2ac/b+(b^2+4ac)^{1/2}]$$

and with a uniquely determined velocity

$$v/\xi=-(1/6c)[b^2+8ac+b(b^2+4ac)^{1/2}]\ ,$$

for all $S_i<S<S_c$. These quantities can be written as

$$\frac{v}{\xi}=\frac{c\theta_+^2}{3}(3\theta_-^2-\theta_+^2),\quad \xi^2=\frac{3}{c\theta_+^4}\ . \tag{3.14}$$

It is readily checked that v, as given by (3.14), is negative according to our physical picture of an advancing front moving to the left and shrinking the domain of the metastable solution $\theta=0$. At $S=S_T$, $3\,\theta_-^2=\theta_+^2$ and $v=0$. An analogous solution corresponds to the situation $S<S_T$, but now the velocity will be positive to describe the growth of the absolutely stable solution $\theta=0$. From (3.13) it can be seen that within the metastable region an interface profile will move without distortion with a well-defined velocity. The hydrodynamic coupling only modifies, in our approximation, the time scale of evolution.

A final remark concerning our propagating front result is in order. According to a recent analysis of a nonlinear marginal-stability criterion for velocity selection,[15] the result for the velocity derived above for a metastable situation would also apply to propagation into the unstable state $\theta=0$ for a finite range of positive values of $S>S_c$.

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APPENDIX

Equation (2.7) in the approximation (2.10) can be written as a stochastic time-dependent Ginzburg-Landau equation for the angular variable $\phi(\mathbf{r},t)$ as

$$\partial_t\phi(\mathbf{r},t)=-\frac{1}{\gamma_1^*}\frac{\delta F}{\delta\phi}+\eta(\mathbf{r},t)\ , \tag{A1}$$

where $\eta(\mathbf{r},t)$ is a new stochastic term which appears after the elimination of the velocity. It satisfies the appropriate fluctuation-dissipation relation with the effective viscosity γ_1^*,

$$\langle\eta(\mathbf{r},t)\eta(\mathbf{r}',t')\rangle=\frac{2k_BT}{\gamma_1^*}\delta(\mathbf{r}-\mathbf{r}')\delta(t-t'). \tag{A2}$$

Expanding $\delta F[\phi]/\delta\phi(\mathbf{r},t)$ for small values of $\phi(\mathbf{r},t)$ we obtain

$$\frac{\delta F[\phi]}{\delta\phi}=-(k_3-k_1)(\partial_x\phi)(\partial_z\phi)-[k_1(\partial_x^2\phi)+k_2(\partial_y^2\phi)+k_3(\partial_z^2\phi)] \\ -\frac{S_z(x)}{c_o}n_o r[\phi+(\tfrac{3}{2}r-\tfrac{2}{3})\phi^3+(\tfrac{2}{15}-\tfrac{3}{2}r+\tfrac{15}{8}r^2)\phi^5+O(\phi^6)]\ . \tag{A3}$$

In the following we neglect the quadratic term $(\partial_x\phi)(\partial_z\phi)$ (as usually done in the context of the Landau-Ginzburg theory). Fourier transforming the fields $\phi(\mathbf{r},t)$ and $\eta(\mathbf{r},t)$ we are able to identify the more unstable modes involved in the relaxation process triggered by $\eta(\mathbf{r},t)$. The Fourier expansion is given by

$$\phi(\boldsymbol{\rho},z,t)=\sum_{n=1}^{\infty}\sum_{\mathbf{q}}\sin\left[n\frac{\pi z}{d}\right]e^{i\mathbf{q}\cdot\boldsymbol{\rho}}\tilde{\phi}_{n,\mathbf{q}}(t),\quad \boldsymbol{\rho}=(x,y)\ . \tag{A4}$$

The fluctuation-dissipation relation (A2) becomes

$$\langle\tilde{\eta}_n\mathbf{q}_1(t_1)\tilde{\eta}_m\mathbf{q}_2(t_2)\rangle=\frac{2k_BT}{\gamma_1^*}\frac{2}{V}\delta_{m,n}\delta_{\mathbf{q}_1,-\mathbf{q}_2}\delta(t_1-t_2)\ , \tag{A5}$$

where here V is the volume of the system.

Substituting (A3)–(A5) in (A1) we obtain for the case of plane-wave illumination $S_z(x)=S$

$$\lambda d_t\tilde{\phi}_{m,\mathbf{q}}(t)=\left[1-m^2\frac{S_c}{S}-\alpha^2Q^2\right]\tilde{\phi}_{m,\mathbf{q}}(t) \\ +(\tfrac{3}{2}r-\tfrac{2}{3})\sum_{\mathbf{q}',\mathbf{q}''}\sum_{n,n',n''}\tilde{\phi}_{n,\mathbf{q}-(\mathbf{q}'+\mathbf{q}'')}\tilde{\phi}_{n',\mathbf{q}'}\tilde{\phi}_{n'',\mathbf{q}''} \\ \times\left[\frac{2}{d}\int_0^d\sin\left[m\frac{\pi z}{d}\right]\sin\left[n\frac{\pi z}{d}\right]\sin\left[n'\frac{\pi z}{d}\right]\sin\left[n''\frac{\pi z}{d}\right]dz\right] \\ +(\tfrac{2}{15}-\tfrac{3}{2}r+\tfrac{15}{8}r^2)\sum_{\mathbf{q}',\mathbf{q}'',\mathbf{q}''',\mathbf{q}^{\mathrm{IV}}}\sum_{n,n',n'',n''',n^{\mathrm{IV}}}\tilde{\phi}_{n,\mathbf{q}-(\mathbf{q}'+\mathbf{q}''+\mathbf{q}'''+\mathbf{q}^{\mathrm{IV}})}\tilde{\phi}_{n',\mathbf{q}'}\tilde{\phi}_{n'',\mathbf{q}''}\tilde{\phi}_{n''',\mathbf{q}'''}\tilde{\phi}_{n^{\mathrm{IV}},\mathbf{q}^{\mathrm{IV}}} \\ \times\left[\frac{2}{d}\int_0^d\sin\left[\frac{m\pi z}{d}\right]\sin\left[\frac{n\pi z}{d}\right]\sin\left[\frac{n'\pi z}{d}\right]\sin\left[\frac{n''\pi z}{d}\right] \\ \times\sin\left[\frac{n'''\pi z}{d}\right]\sin\left[\frac{n^{\mathrm{IV}}\pi z}{d}\right]dz\right]+\lambda\tilde{\eta}_{m,\mathbf{q}}(t)\ , \tag{A6}$$

where the following identifications have been made:

$$\lambda^{-1}\equiv\frac{Sn_or}{\gamma_1^*}c_o,\quad \alpha^2\equiv k_3\frac{c_o}{Sn_or},\quad S_c=\frac{k_3\pi^2c_o}{n_ord^2}\ , \\ Q^2=(k_1q_x^2+k_2q_y^2)/k_3\ . \tag{A7}$$

A linear version of (A6) gives

$$\lambda d_t\tilde{\phi}_{m,\mathbf{q}}(t)=\alpha^2[q_c(m)^2-Q^2\tilde{\phi}_{m,\mathbf{q}}(t)+\lambda\tilde{\eta}_{m,\mathbf{q}}(t),\quad m\geq 1 \tag{A8}$$

where $q_c(m)$ stands for the inverse of a characteristic length of the fluctuations of the mth mode in a plane perpendicular to the z axis,

$$q_c^2(m)=\frac{1}{\alpha^2}\left[1-m^2\frac{S_c}{S}\right]\ . \tag{A9}$$

The correlation length q_c^{-1} measures the spatial equilibrium fluctuations of the system in the x-y plane. For a given m and $\mathbf{q}$, the mode $\phi_{m,\mathbf{q}}(t)$ becomes unstable if the pair of inequalities

$$1-m^2\frac{S_c}{S}>0, \quad q_c^2>Q^2 \tag{A10}$$

are both satisfied. For each unstable z mode m, there is a range of unstable transverse modes **Q**. For example, for Poynting amplitudes $S_c<S<4S_c$, only the z mode $m=1$ is unstable.

The stochastic version of (2.13) is obtained from (A1) and (A3) assuming space homogeneity in the y-direction and neglecting the term $(k_3-k_1)(\partial_x\phi\partial_z\phi)$. From (A6) we can obtain a mean-field equation for the most unstable z mode $m=1$ in which spatial fluctuations in the plane x-y are averaged, by taking the limit $q\rightarrow 0$. In this context we can interpret (2.15) in the sense of a space-averaged equation in the y direction, for the mode $m=1$.

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