"Catastrophes" in orientational interaction of an optical wave with a nematic liquid crystal

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A theoretical investigation is reported of abrupt changes in orientational states of nematic liquid crystals when external parameters such as the intensities and propagation directions of optical waves interacting with a nematic liquid crystal vary smoothly. An analysis of these processes on the basis of the theory of catastrophes has made it possible to predict new types of instability and hysteresis when an orientational state of a nematic liquid crystal is governed by two parameters, such as the intensity of an optical wave and the angle at which it is incident on this liquid crystal. Estimates show that the predicted effects should be easily detected experimentally.

INTRODUCTION

Threshold and hysteresis effects in reorientation of liquid crystals in external fields are of considerable theoretical and practical interest.[1] Recent years have seen major attention given to orientational interactions of optical waves with liquid crystals. These interactions have many special features distinguishing them from orientational effects in quasistatic (electric, magnetic) fields and are of considerable interest from the point of view of the physics of liquid crystals and establishing various nonlinear processes and interactions between optical waves in liquid crystals.[2]

It is pointed out below that the published investigations of threshold and hysteresis phenomena in light-induced orientational processes in liquid crystals represent in fact very special cases of the theory of catastrophes. We shall show that using the approach of this theory, which relates possible types of instabilities in a system to the number of the controlling parameters,[3] makes it possible to predict and interpret new instability phenomena.

Light-induced effects in liquid crystals are characterized by a large number of parameters such as the intensity, state of polarization, and angle of incidence of optical waves, the number of such waves and their nature (ordinary and extraordinary waves), material parameters of a liquid crystal, its orientational state, etc. This circumstance, together with the fact that the orientation of the director is usually governed by two parameters (angles) leads us to expect the occurrence of various types of catastrophes.

We shall consider the clearest situation represented by the case of two controlling parameters, such as the intensity of an optical wave and its angle of incidence, while the other parameters are kept constant. A similar situation, i.e., the case of two controlling parameters is discussed in Refs. 4, 5 in the case of static electric and magnetic fields.

1. CONTROL OF THE ORIENTATION OF A NEMATIC LIQUID CRYSTAL BY ONE OPTICAL WAVE

In the present section we shall discuss the situation which can obviously be achieved most easily in experiments: we shall assume that the parameters controlling the orientation of the director are the intensity and angle of incidence of an optical wave on a nematic liquid crystal (NLC).

1. Principal equations

We shall consider a cell containing a homeotropically oriented NLC, on which a linearly polarized (xz plane) optical wave is incident at a small angle. The z axis of a Cartesian coordinate system is directed along the normal to the plates of the cell and this axis coincides with the unperturbed orientation $\mathbf{n}^0$ of the director. It is known[6] that when the intensity of such a wave is $P\sim P_F$, where P_F is the threshold intensity for the optical Fréedericksz transition, and the wave is incident normally on a cell with a homeotropically oriented NLC or if the angle of incidence is small so that $\alpha \ll 1$, a small perturbation of the director field is excited:

$$\mathbf{n}=\{\sin\varphi(z),\ 0,\ \cos\varphi(z)\},\quad \varphi(z)\approx\varphi_m\sin(\pi z/L)$$

(L is the thickness of the cell), which is described by the following equation:

$$B\varphi_m^3-{}^1/_2(\rho-1)\varphi_m+2\alpha\rho/\pi\varepsilon_\perp^{1/2}=0, \tag{1}$$

where

$$B={}^1/_4(1-9\varepsilon_a/4\varepsilon_\parallel-k),\quad k=1-K_1/K_3,\quad \rho=P/P_F,$$

K_1 and K_3 are the Frank constants such that for the usual thermotropic NLCs we have $K_1/K_3<1$, i.e., $k>0$, and $\varepsilon_a=\varepsilon_\parallel-\varepsilon_\perp$ is the anisotropy of the permittivity of the NLC at optical frequencies.

Equation (1) was obtained in Ref. 6 by simultaneous solution of a variational equation valid for small perturbations of the director and of the wave equation, retaining terms up to third order in φ_m and up to first order in the angle of incidence α.

For given values of the parameters of an NLC, i.e., for a fixed B, the dependence of φ_m on the parameters ρ and α is described by a smooth surface in the $\varphi_m\rho\alpha$ space, known as Whitney's fold[3] (Fig. 1a). The projection of the surface on the plane of the controlling parameters has a singularity.

The existence of the singularity is associated with the fact that the $\rho\alpha$ plane separates into two regions: in each of them every point (ρ,α) has only one inverse image, whereas in the other region each point has three inverse images. The other region is defined by the condition

$$\frac{1}{B}\left(\frac{1-\rho}{6}\right)^3+\left(\frac{\alpha\rho}{\pi\varepsilon_\perp^{1/2}}\right)^2\leqslant 0, \tag{2}$$

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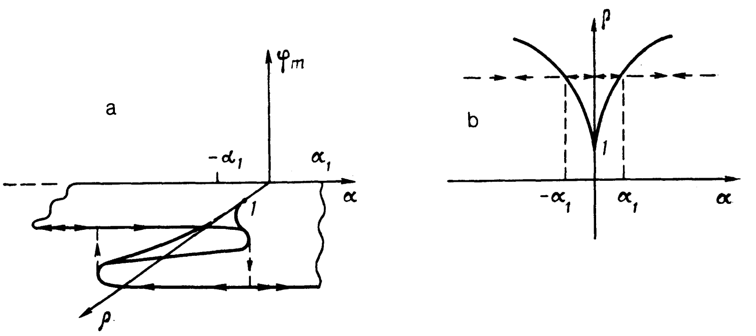

FIG. 1. Surface of equilibrium of perturbations of the director (a) and the bifurcation curve in the plane of the controlling parameters (b). The arrows show the direction of variation of the angle α. Abrupt changes in φ_m occur at the points α_1 and $-\alpha_1$ as we move from left to right and vice versa.

which follows from Eq. (1).

An investigation of the nature of the equilibrium can be carried out by constructing the free energy function $\Phi(\varphi_m)$; minimization of this function yields Eq. (1) for φ_m (see Ref. 6):

$$\Phi = {}^1/_4 B\varphi_m^4 - {}^1/_4(\rho-1)\varphi_m^2 + (2\alpha\rho/\pi\varepsilon_\perp^{1/2})\varphi_m. \quad (3)$$

Having determined the sign of the second derivative of Φ with respect to φ_m for different solutions of Eq. (1), we can readily show that the upper and lower surfaces in Fig. 1a correspond to stable states, whereas the middle surface corresponds to unstable equilibrium states.

The dependence $\Phi(\varphi_m)$ plotted in Fig. 2 shows that for $\rho < 1$ (Fig. 2a) there is only one stable equilibrium state and, consequently, in this approximation there should be no abrupt changes in the state. For $B < 0$, in principle abrupt changes may take place but in discussing them we need to allow for terms of higher orders in φ_m than φ_m^3. However, for $\rho > 1$ (Fig. 2b) and $B < 0$, there is only one unstable equilibrium state, as can be deduced directly from the condition that the inequality (2) is disobeyed. Once again the possibility of catastrophes, i.e., the appearance of stable states, is described by terms which are of higher orders than φ_m^3.

It therefore follows that the inequality of Eq. (2) should be considered subject to the conditions $B > 0$ and $\rho > 1$ and it can be transformed to

$$\rho > \rho_0(\alpha) \approx 1 + 6(B\alpha^2/\pi^2\varepsilon_\perp)^{1/3}. \quad (4)$$

A bifurcation curve separating regions with different numbers of inverse images in the $\rho\alpha$ plane is shown in Fig. 1b.

2. Instabilities due to changes in the angle of incidence of a wave

For a fixed value of the optical wave intensity $\rho < 1$ we find that variation of the angle α from, for example, its negative values to the range $\alpha > 0$ causes smooth reorientation of the director from $\varphi_m > 0$ to $\varphi_m < 0$. However, for $\rho > 1$, then the change in φ_m considered as a function of α becomes abrupt (fold catastrophe), as shown in Fig. 3. It is assumed that the angle α varies adiabatically slowly compared with the characteristic relaxation time of the director orientation. Abrupt changes occur at the boundaries of folds of the equilibrium surface projected onto the controlling surface as a cusp; they amount to

$$|\Delta\varphi_m| = [{}^3/_2(\rho-1)/B]^{1/2}. \quad (5)$$

The physical nature of this new type of instability can be explained as follows. If we have $\rho > 1$ and the value of α is negative and it is located outside a cusp, the director becomes reoriented in a direction which reduces the angle between **n** and **E** (**E** is the vector representing the intensity of the electric field of an optical wave), i.e., $\varphi_m > 0$ (Fig. 4a). Where α is reduced to zero, the value of φ_m remains positive, so that by definition we have $P > P_F$ (Fig. 4b). Clearly, when the sign of α is reversed, the absolute minimum of the free energy corresponds to a state with a negative φ_m. However, a state with $\varphi_m > 0$ remains stable up to a certain value of α (Figs. 4c and 4d). When α is varied in the opposite direction, an abrupt change in φ_m occurs at a different point and hysteresis occurs.

3. Instabilities associated with successive changes in the intensity and angle of incidence of a wave

We shall now consider a situation in which, with α constant (for example, $\alpha < 0$), the value of ρ is varied. If the initial point (ρ, α) is inside a cusp, then there is an abrupt change at $\rho = \rho_0(\alpha)$.

This type of instability may be observed, for example, in the following process. If the initial value is $\alpha > 0$ and a given value of $\rho > 1$ is retained, we can reduce α to a negative value α_i, which however is less than the bifurcation angle α_b. Sub-

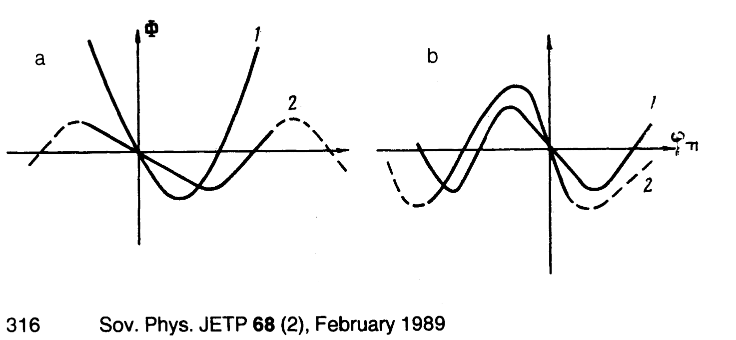

FIG. 2. Dependence of the free energy Φ on φ_m in the case when $\alpha < 0$: a) $\rho < 1$; b) $\rho > 1$. Curves 1 and 2 correspond to $B > 0$ and $B < 0$. The dashed parts of the curves represent the range of the dependence $\Phi(\varphi_m)$, corresponding to allowance for terms of higher order than φ_m^4.

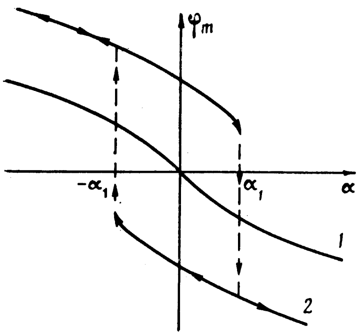

FIG. 3. Dependence $\varphi_m(\alpha)$ for a fixed ρ: 1) $\rho < 1$; 2) $\rho > 1$.

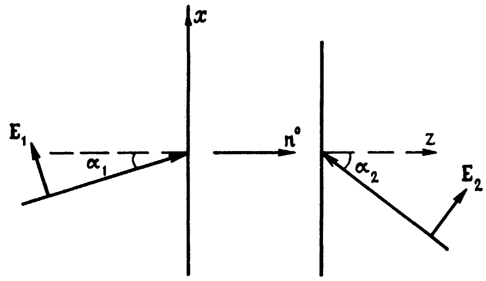

FIG. 5. Cell with a homeotropically oriented nematic liquid crystal subjected simultaneously to two light beams incident in the xz plane at angles α_1 and α_2. The polarizations of the two beams lie in the same plane.

sequent reduction in the intensity of the wave for a fixed value of α_i abruptly alters φ_m when $\rho_i = \rho_0(\alpha_i)$ is altered.

2. CONTROL OF THE ORIENTATIONAL STATE OF A NEMATIC LIQUID CRYSTAL BY TWO OPTICAL BEAMS

Some effects of reorientation of the director of an NLC interacting simultaneously with two optical beams were predicted and investigated in Ref. 7. The compensation effect, i.e., the disappearance of threshold-free perturbations of the director was observed subsequently experimentally.[8] Effects of this kind represent in fact the process of control of the orientational state of an NLC by two beams. This increases greatly the number of controlling parameters. In the present section we shall reexamine the effects which appear when the orientation of a liquid crystal is controlled by just two parameters and the other parameters are fixed.

We shall consider a cell with a homeotropically oriented NLC on which optical waves are incident at small angles from opposite directions (Fig. 5). The effects associated with interference of these waves will be ignored, because the characteristic scale on which the interference pattern varies is of the order of the wavelength and much smaller than the distance over which perturbations of the director vary, i.e., much smaller than the thickness of the NLC cell.

Using the results of Refs. 6 and 7, we can obtain an equation for φ_m which, to the required accuracy, is of the form

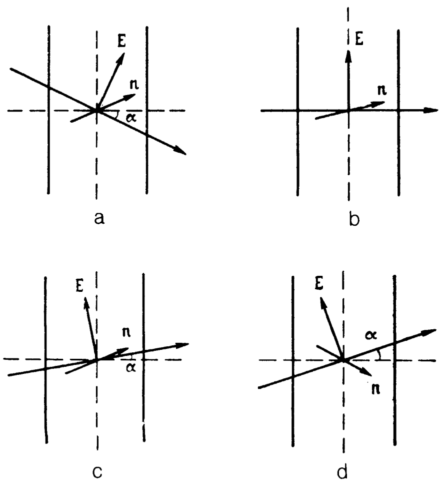

FIG. 4. Behavior of the director when the angle α is varied and $\rho > 1$.

$$B\varphi_m^3 - {}^1/_2(\rho_1+\rho_2-1)\varphi_m + 2(\alpha_1\rho_1 - \alpha_2\rho_2)/\pi\varepsilon_\perp^{1/2} = 0. \qquad (6)$$

where $\rho_{1,2} = P_{1,2}/P_F$. The above equation is derived ignoring the terms proportional to higher powers of the angles of incidence α_1 and α_2.

1. Abrupt changes for constant angles of incidence of waves on a nematic liquid crystal

We shall consider the specific situation when the angles of incidence α_1 and α_2 are equal: $\alpha_1 = \alpha_2 = \alpha$, when the control is provided by varying the beam intensities P_1 and P_2. The bifurcation curve is then described by the following expression which is derived from Eq. (6):

$$\rho_1+\rho_2 = 1+6(B\alpha^2/\pi^2\varepsilon_\perp)^{1/3}(\rho_1-\rho_2)^{2/3}. \qquad (7)$$

It describes a semicubic parabola, which is symmetric relative to the bisector of the first quadrant of the Cartesian coordinate system $\rho_1\rho_2$ (Fig. 6).

Therefore, if the intensity of one of the waves exceeds $0.5P_F$ and it is fixed, variation of the intensity of the other wave can result in abrupt changes of the orientation of the director. These abrupt changes can also occur when the intensities of both waves are altered simultaneously, i.e., for an oblique section of the cusp in Fig. 6.

2. Abrupt changes in the case of equal intensities

For $P_1 = P_2 = P$, it is found that the second controlling parameter is the difference between the angles of incidence

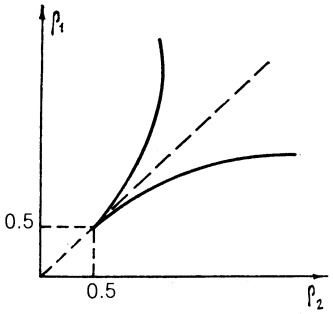

FIG. 6. Bifurcation curve for equal angles of incidence of the waves when the control is due to variation of their intensities.

of the waves $\Delta\alpha = \alpha_1 - \alpha_2$. The bifurcation curve is then described by

$$2\rho = 1 + 3(2B/\pi^2\varepsilon_\perp)^{1/3}\Delta\alpha^{2/3}. \quad (8)$$

3. Abrupt changes due to variation of the angles of incidence of waves

We shall assume that P_1 and P_2 are fixed. Then Eq. (6) yields the following expression for the bifurcation region:

$$|\alpha_1\rho_1 - \alpha_2\rho_2| \leqslant (\pi\varepsilon_\perp^{1/2}/6^{3/2}B^{1/2})(\rho_1+\rho_2-1)^{3/2}. \quad (9)$$

In the $\alpha_1\alpha_2$ plane there is a band with a slope governed by the ratio P_1/P_2; the width of this band is determined by the factor representing the excess above the threshold intensity.

Equation (9) is in fact an inequality needed to define the bifurcation region in the most general case of different values of α_1, α_2, P_1, and P_2 when two out of four parameters are fixed.

Clearly, the abrupt change in the orientation of the director due to a change in the intensity of one of the waves when the intensity of the second wave is constant and the angles of incidence are equal and fixed, was observed in the investigation reported in Ref. 8.

CONCLUSIONS

The catastrophes predicted in the present study for the orientational interaction of an optical wave with a liquid crystal can easily be detected experimentally. This is indicated by the following numerical estimates. A typical value of P_F is $P_F \sim 0.1$ kW/cm^2. In the case of parameters of an NLC layer (for example, MBBA) of thickness $L \sim 10^{-2}$ cm and for $P/P_F \approx 1.1$, we find that abrupt changes in the orientation due to a change in the angle of incidence of the wave occur between states with $\varphi_m^{(1)} \approx -0.6$ and $\varphi_m^{(2)} \approx 0.35$ (or $\varphi_m^{(1)} \approx 0.6$ and $\varphi_m^{(2)} \approx -0.35$), when $|\alpha| = |\alpha_b| \approx 1.5°$.

A nonlinear phase advance at low values of φ_m and α is given by the expression[6]

$$\psi = \frac{\omega}{c}\frac{\varepsilon_a\varepsilon_\perp^{1/2}}{4\varepsilon_\parallel}L\left(\varphi_m^2 - \frac{8}{\pi}\alpha\varphi_m\right) \quad (10)$$

and it then undergoes a jump equal to $\delta\psi = \psi_1 - \psi_2 \approx 36$ rad. Such a change can easily be detected, for example, by determination of the change in the number of rings observed in aberration self-focusing of light, amounting to $\Delta N = \delta\psi/2\pi \approx 6$. It should be pointed out that in the processes under discussion the important factor is not simply the absolute value of the intensity of an optical wave incident on an NLC, but the ratio of this intensity to P_F, which is governed by the parameters of the NLC cell. Bearing in mind that for a fixed value of the intensity of incident light we have $\rho \sim \varepsilon_a L^2/K$, we can see why instabilities will occur also when we vary the thickness of the NLC layer or the frequency of light in the dispersion region.

The present paper thus discusses the simplest and most accessible experimental situation when we initially have homogeneously oriented NLC and control is provided by just two parameters. It is shown above that some interesting new types of instability can then be observed.

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