# Fredericks transitions induced by light fields

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The reorientation of the director of a nematic liquid crystal induced by the field of a light wave is considered. An oblique (with respect to the director) extraordinary wave of low intensity yields the predicted and previously observed giant optical nonlinearity in a nematic liquid crystal. For normal incidence of the light wave on the cuvette with a homeotropic orientation of the nematic liquid crystal, the reorientation appears only at light intensities above a certain threshold, and the process itself is similar to the Fredericks transition. The spatial distribution of the director direction is calculated for intensities above and below threshold. Hysteresis of the Fredericks transition in a light field, which has no analog in the case of static fields, is predicted.

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## 1. INTRODUCTION

The nonlinear optics of liquid crystals has recently received a great deal of attention (see the review[1]). In addition to the quite interesting quadratic nonlinearities (generation of the second harmonic[2]), recently there have been predictions[3–8] and experimental discoveries[4,9] of giant cubic optical nonlinearities of liquid crystals (nematic, cholesteric, and smectic), which are due to the reorientation of the director by light fields. Succeeding experimental studies[10,11] have confirmed the presence of giant optical nonlinearities for a number of specific nematic liquid crystals (NLC) and experimental geometries. In particular, a nonlinear interaction was found[11] between a normally incident light wave and a homeotropically oriented cell of the liquid crystal (OCBP), and had a characteristic threshold dependence on the intensity of the laser beam. This effect was explained qualitatively in Ref. 11 on the basis of an analogy with the Fredericks transition in the field of a light wave.

In the present study we construct a quantitative theory of the Fredricks transition in the field of a light wave. In contrast to the simplest model of the Fredericks transition in static fields (see Refs. 12 and 13), here we take into account the following two facts: 1) the amplitude for the deformation of the director above threshold must be found taking into account the distortion of the longitudinal profile of the light wave itself as it propagates in an inhomogeneous anisotropic medium; 2) if the transverse dimension of the beam is significantly smaller than the cuvette thickness, the threshold intensity itself depends on the transverse distribution of the intensity in the beam. In addition, the theory predicts that for certain liquid crystals the Fredericks transition is accompanied by hysterisis.

## 2. THE SYSTEM OF BASIC EQUATIONS

We shall take the free energy per unit volume of an NLC in the presence of a light field of complex amplitude $\mathbf{E}$ to be of the form

$$F\,[\mathrm{erg/cm^3}] = {}^1/_2 K_{11}(\operatorname{div}\mathbf{n})^2 + {}^1/_2 K_{22}(\mathbf{n}\operatorname{rot}\mathbf{n})^2 + {}^1/_2 K_{33}[\mathbf{n}\operatorname{rot}\mathbf{n}]^2 - \varepsilon_a(\mathbf{nE})(\mathbf{nE}^*)/16\pi - \varepsilon_\perp|\mathbf{E}|^2/16\pi. \quad (1)$$

Here $K_{ii}$ are the Frank constants, $\mathbf{n}$ is a unit vector in the direction of the director ($\mathbf{n}$ and $-\mathbf{n}$ will be assumed equivalent), and the permittivity tensor of an NLC at the frequency of the light field $\omega$ is

$$\varepsilon_{ik} = \varepsilon_\perp\delta_{ik} + (\varepsilon_\parallel - \varepsilon_\perp)n_i n_k.$$

In addition, we have used in (1) the notation $\varepsilon_a = \varepsilon_\parallel - \varepsilon_\perp$.

The complex amplitude of a quasimonochromatic field $\mathbf{E}(\mathbf{r}, t)$ is determined in a self-consistent manner from the solution of Maxwell's equations with

$$\varepsilon_{ik}(\mathbf{r}, t) = \varepsilon_\perp\delta_{ik} + \varepsilon_a n_i(\mathbf{r}, t)n_k(\mathbf{r}, t).$$

However, we emphasize that to obtain the variational equations for the director $\mathbf{n}(\mathbf{r}, t)$ it is necessary to assume that the amplitude of the electric field $\mathbf{E}(\mathbf{r}, t)$ is fixed. We shall take the density of the dissipative function $R$ (in erg/cm$^3\cdot$sec) to be

$$R = {}^1/_2\eta(\dot{\mathbf{n}}\dot{\mathbf{n}}). \quad (2)$$

The variational equations for $\mathbf{n}(\mathbf{r}, t)$ have the form

$$\frac{\delta F}{\delta n_i} - \frac{\partial}{\partial x_k}\frac{\delta F}{\delta(\partial n_i/\partial x_k)} = \lambda n_i - \frac{\delta R}{\delta \dot{n}_i}, \quad (3)$$

where $\lambda(\mathbf{r})$ is an undetermined Lagrange multiplier that ensures that the condition $|\mathbf{n}| = 1$ is satisfied.

## 3. THE FREDERICKS TRANSITION IN BROAD LIGHT BEAMS

Let us consider a homeotropically oriented NLC cell occupying a layer $0 \leqslant z \leqslant L$. We shall assume that the light field has nonzero components $E_x$ and $E_z$, while $E_y = 0$. The unperturbed direction of the director in $\mathbf{n}^0 = \mathbf{e}_z$. We shall also assume that the perturbation of the director does not move it out of the $xz$ plane. Then

$$\mathbf{n}(\mathbf{r}) = \mathbf{e}_z\cos\varphi + \mathbf{e}_x\sin\varphi,$$

where $\varphi = \varphi(\mathbf{r})$. If the transverse dimensions of the beam $a_\perp$ in the $xy$ plane are much larger than the cuvette thickness $L$ and the beam itself is almost a plane wave, the field components $E_x$ and $E_z$ can be assumed to depend only on $z$; more precisely,

$$\mathbf{E} = [\mathbf{e}_x E_x(z) + \mathbf{e}_z E_z(z)]\exp(i\omega s x/c + i k_z z). \quad (4)$$

Here $s = \sin\alpha$ and $\alpha$ is the angle of incidence of the


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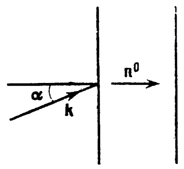


FIG. 1. Unperturbed director vector $\mathbf{n}^0$ perpendicular to the cell walls. The wave vector $\mathbf{k}$ of the light field makes an angle $\alpha$ with the normal to the cell walls.

light wave on the cell in air (see Fig. 1). Under these conditions the slope of the director $\varphi$ also depends only on $z$. Now the variational equations (3) take the form

$$(K_{11}\sin^2\varphi+K_{33}\cos^2\varphi)\frac{\partial^2\varphi}{\partial z^2}-(K_{33}-K_{11})\sin\varphi\cos\varphi\left(\frac{\partial\varphi}{\partial z}\right)^2+\frac{\varepsilon_a}{16\pi}[\sin 2\varphi(|E_x|^2-|E_z|^2)+\cos 2\varphi(E_xE_z^*+E_x^*E_z)]=\eta\frac{\partial\varphi}{\partial t}. \quad (5)$$

Let us first give a qualitative picture of the Fredericks transition. When a plane light wave falls at normal incidence on a homeotropic cell the electric field of the wave is exactly perpendicular to the director. At a positive value of $\varepsilon_a$ it would be energetically favorable to orient the director in the direction of the field. However, this is prevented by the homeotropic orientation of the director by the curvette walls. Furthermore, in the first approximation in the light intensity the orientational effect of the field on the unperturbed director is absent, in other words, for $\mathbf{E}=\mathbf{e}_x E_x$ the function $\varphi(z)\equiv 0$ is an exact solution to equation (5).

Above a certain threshold value of the intensity the solution $\varphi(z)\equiv 0$ will no longer be stable. To determine this threshold it is convenient to consider the linearized [that is, at small $\varphi(z)$] equation (5). Here we must take into account the fact that from the equation $\operatorname{div}\mathbf{D}=0$ we have

$$k_zD_z=k_z(\varepsilon_{zx}E_x+\varepsilon_{zz}E_z)=0.$$

Therefore, to terms linear in $\varphi$ we have $E_z=-\varepsilon_a\varphi E_x/\varepsilon_\parallel$ and equation (5) takes the form

$$K_{33}\frac{\partial^2\varphi}{\partial z^2}+\frac{\varepsilon_a}{4\pi}\frac{\varepsilon_\perp}{\varepsilon_\parallel}\frac{|E_x|^2}{2}\varphi=\eta\frac{\partial\varphi}{\partial t}. \quad (6)$$

Equation (6) is analogous to the linearized equation for the behavior of the director near the Fredericks transition in a static field $\mathbf{E}_{\rm stat}=\mathbf{e}_x E_{\rm stat}$ (cf. Ref. 13, section 4.2). There are two differences, however. First, instead of the $E^2_{\rm stat}$ of the static case in (6) we have $|E_x|^2/2$—the mean square of the amplitude of the light field. Secondly, in a static field $\mathbf{E}_{\rm stat}$ the condition $\operatorname{div}\mathbf{D}=0$ has no effect on the field vector $\mathbf{E}$ when the director is inclined. In contrast to this, in the problem with a light field, a component $E_z$ appears because of the condition $\operatorname{div}\mathbf{D}=0$.

As a result, in equation (6) we have an additional factor $\varepsilon_\perp/\varepsilon_\parallel$, the role of which reduces to a certain increase of the threshold. Expanding $\varphi(z,t)$ in a Fourier series and taking the boundary conditions $\varphi(z=0,t)=\varphi(z=L,t)=0$ into account, we obtain from (6)

$$\varphi(z,t)=\sum_k\varphi_k\sin\frac{\pi kz}{L}\exp\Gamma_k t,$$

$$\Gamma_k=\eta^{-1}\left(\frac{\varepsilon_\perp\varepsilon_a|E|^2}{8\pi\varepsilon_\parallel}-\frac{k^2\pi^2K_{33}}{L^2}\right). \quad (7)$$

From (7) we find that for

$$|E|^2\geqslant|E|_{\rm Fr}^2=\frac{8\pi^3K_{33}}{\varepsilon_aL^2}\frac{\varepsilon_\parallel}{\varepsilon_\perp}$$

a perturbation of the form $\sin(\pi z/L)$ begins to grow exponentially.

The steady-state value of the amplitude $\varphi=\varphi_{\max}\sin(\pi z/L)$ in the regime above threshold must now be determined with allowance for the nonlinear terms in equation (5). It is very important that in this approximation we must include a solution consistent with $\varphi(z)$ to Maxwell's equations in an inhomogeneous anisotropic medium. For a wave of the form (4) these solutions can be obtained in the geometrical optics approximation (see Refs. 14 and 15):

$$E_x(z)=A(\varepsilon_{zz}-s^2)^{1/4}e^{i\psi(z)},$$

$$E_z(z)=-A\frac{\varepsilon_{xz}(\varepsilon_{zz}-s^2)^{1/2}+s(\varepsilon_\parallel\varepsilon_\perp)^{1/2}}{\varepsilon_{zz}(\varepsilon_{zz}-s^2)^{1/4}}e^{i\psi(z)}. \quad (8a)$$

The advance of the optical phase $\psi(z)$ is equal to

$$\psi(z)=\frac{\omega}{c}\int_0^L\frac{-s\varepsilon_{xz}(z')+[\varepsilon_\parallel\varepsilon_\perp(\varepsilon_{zz}(z')-s^2)]^{1/2}}{\varepsilon_{zz}(z')}dz'. \quad (8b)$$

As already noted, $s=\sin\alpha$, where $\alpha$ is the angle of incidence in air, and the $\varepsilon_{ik}$ are

$$\varepsilon_{zz}=\varepsilon_\perp+\varepsilon_a\cos^2\varphi(z),\quad \varepsilon_{xz}=\varepsilon_a\sin\varphi(z)\cos\varphi(z). \quad (9)$$

The constant $A$ can be expressed in terms of the power flux density; namely the $z$ component of the Poynting vector is

$$P_z=c(\varepsilon_\parallel\varepsilon_\perp)^{1/2}|A|^2/8\pi$$

and is independent of $z$.

In the case of exactly normal incidence ($s=0$) equation (5), taking into account (8) and (9), has the following form in the stationary case:

$$(K_{11}\sin^2\varphi+K_{33}\cos^2\varphi)\frac{d^2\varphi}{dz^2}-(K_{33}-K_{11})\sin\varphi\cos\varphi\left(\frac{d\varphi}{dz}\right)^2+\frac{\varepsilon_a\varepsilon_\parallel\varepsilon_\perp|A|^2\sin\varphi\cos\varphi}{8\pi(\varepsilon_\perp+\varepsilon_a\cos^2\varphi)^{3/2}}=0. \quad (10)$$

The exact solution of this equation will be given in Sec. 5 below. Here we shall restrict ourselves to only taking into account terms of order $\varphi$ and $\varphi^3$ in this equation, a procedure valid near the Fredericks threshold. The search for a solution of the form

$$\varphi=\varphi_1\sin(\pi z/L)+\varphi_3\sin(3\pi z/L)+\ldots,$$

which automatically satisfies the boundary conditions, leads to the following expression for $\varphi_1$:

$$\varphi_1^2=2\left(1-\frac{9}{4}\frac{\varepsilon_a}{\varepsilon_\parallel}-k\right)^{-1}\frac{P-P_{\rm Fr}}{P_{\rm Fr}}, \quad (11)$$

$$k=(K_{33}-K_{11})/K_{33},\quad P_{\rm Fr}\left[\frac{\rm erg}{\rm cm^2\cdot sec}\right]=\frac{c\varepsilon_\parallel K_{33}}{\varepsilon_a\varepsilon_\perp^{1/2}}\left(\frac{\pi}{L}\right)^2. \quad (12)$$

Here $\varphi_3\sim\varphi_1^3$. Therefore $\varphi_1$ is proportional to the square root of the excess of the intensity $P$ above threshold $P_{\rm Fr}$.

The advance of the optical phase $\psi(L)$ over the cuvette thickness $L$ can be obtained from (8b); expanding (8b) to terms $\sim\varphi_1^2$ we obtain

$$\psi(L)=\frac{\omega}{c}\varepsilon_\perp^{1/2}L\left[1+\frac{1}{2}\frac{\varepsilon_a}{\varepsilon_\parallel}\left(1-\frac{9}{4}\frac{\varepsilon_a}{\varepsilon_\parallel}-k\right)^{-1}\frac{P-P_{Fr}}{P_{Fr}}\right]. \tag{13}$$

## 4. THE FREDERICKS TRANSITION IN NARROW BEAMS

In the case where the transverse dimension of the beam $a_\perp$ is smaller than or on the order of the cuvette thickness $L$, the Frank energy due to the transverse gradients of the director becomes dominant. Let us first estimate the order of magnitude of the threshold power of the Fredericks transition. The energy of the perturbed state is

$$W=\frac{1}{2}\varphi_1^2a_\perp^2L\left[K\left(\frac{\pi^2}{L^2}+\frac{1}{a_\perp^2}\right)-\frac{\varepsilon_a|\mathbf{E}|^2}{8\pi}\right].$$

Here $a_\perp^2L$ is the volume occupied by the disturbance and $K$ is the Frank constant. This expression gives a stable state $\varphi_1=0$ at small $|\mathbf{E}|^2$, and instability sets in at

$$|\mathbf{E}|^2\geqslant\frac{8\pi K}{\varepsilon_a}\left(\frac{\pi^2}{L^2}+\frac{1}{a_\perp^2}\right). \tag{14}$$

As seen from expression (14), $|\mathbf{E}|^2a_\perp^2=\text{const}$ at $a_\perp\ll L$.

In order to exactly determine the threshold of the Fredericks transition, it is necessary to solve the three-dimensional problem of the stability of the solution $\varphi=0$. We shall consider this problem for several particular cases.

a. *The single-constant approximation.* In the single-constant approximation we can assume even for a narrow beam that the vectors $\mathbf{E}$ and $\mathbf{n}$ lie in the $xz$ plane. Then in the approximation linearized in $\varphi(\mathbf{r})$ we have

$$\Delta\varphi+\left(\frac{\pi}{L}\right)^2\frac{P(\mathbf{r}_\perp)}{P_{Fr}}\varphi=\frac{\eta}{K}\frac{\partial\varphi}{\partial t}. \tag{15}$$

Here $P_{Fr}$ is determined by formula (12) using the fact that $K_{ii}=K$.

With the substitution $\varphi(\mathbf{r},t)=\varphi'(\mathbf{r})\exp\Gamma t$, equation (15) becomes

$$\Delta\varphi'(\mathbf{r})+\left[\left(\frac{\pi}{L}\right)^2\frac{P(\mathbf{r}_\perp)}{P_{Fr}}-\frac{\Gamma\eta}{K}\right]\varphi'(\mathbf{r})=0. \tag{16}$$

Let us first consider the case where a "ribbon" light beam falls on the medium, $P(\mathbf{r}_\perp)=P(x)$. Then after separating the variables in equation (16) we find by means of the substitution $\varphi'(\mathbf{r})=\xi(x)\chi(z)$

$$\frac{d^2\xi(x)}{dx^2}+\left[\left(\frac{\pi}{L}\right)^2\frac{P(x)}{P_{Fr}}-q\right]\xi(x)=0, \tag{17a}$$

$$\frac{d^2\chi(z)}{dz^2}+p\chi(z)=0,\quad q-p=\Gamma\eta/K. \tag{17b}$$

Equation (17a) is the one-dimensional Schrödinger equation for the potential $U\sim-P(x)$. It is possible to solve this equation analytically for a relation, for example, of the form

$$P(x)=P_0/\mathrm{ch}^2\beta x, \tag{18}$$

that is, for a function which gives a good qualitative approximation of a Gaussian (Fig. 2). The eigenvalues of $q$ for a "potential" of the form (18) will be[16]

$$q_n=\frac{\beta^2}{4}\left\{-(1+2n)+\left[1+\frac{4}{\beta^2}\left(\frac{\pi}{L}\right)^2\frac{P_0}{P_{Fr}}\right]^{1/2}\right\}^2. \tag{19}$$

The eigenvalues of $p$ from (17), corresponding to the eigenfunction $\chi(z)\propto\sin(\pi z/L)$ is $p=\pi^2/L^2$. The perturbation of the director field $\varphi'(\mathbf{r})$ will grow exponentially in time at $\Gamma=\eta^{-1}K(q-p)>0$. Then, defining the threshold intensity of the Fredericks transition as that value of $P_0$ at which a bound state first appears in the potential well $U\propto-1/\cosh^2\beta x$, we obtain

$$P_{th}=P_{Fr}(1+\beta L/\pi). \tag{20}$$

The quantity $\beta L$ can be viewed as the ratio of the cuvette thickness $L$ to the beam diameter $d$: $\beta L\sim L/d$.

**b.** Another case which can be solved analytically and by which we can approximate the real distribution of the intensity in the beam of a light wave corresponds to a function $P(\mathbf{r})$ of the form

$$P(\rho)=\begin{cases}\text{const} & \text{for } \rho\leqslant\rho_0\\ 0 & \text{for } \rho>\rho_0\end{cases}, \tag{21}$$

where $\rho$ is the distance from the beam axis in the transverse direction. Writing equation (16) in cylindrical coordinates and making the substitution $\varphi'(\mathbf{r})=\zeta(\rho)\chi(z)$, we find

$$\frac{d^2\zeta}{d\rho^2}+\frac{1}{\rho}\frac{d\zeta}{d\rho}+\left[\left(\frac{\pi}{L}\right)^2\frac{P(\rho)}{P_{Fr}}-h\right]\zeta=0, \tag{22a}$$

$$\frac{d^2\chi}{dz^2}+g\chi=0,\quad h-g=\Gamma\eta/K. \tag{22b}$$

The solution of (22a) is a zeroth-order Bessel function. Let

$$P(\rho\leqslant\rho_0)>(L/\pi)^2hP_{Fr}=P_{Fr} \tag{23}$$

[in expression (23) we have set $h=\pi^2/L^2$, since director-field perturbations which increase with time exist only for $\Gamma=\eta^{-1}K(h-g)>0$, where $g$, just as $p$ in case ($a$), equals $\pi^2/L^2$]. It is easy to see that the condition (23) is necessary for the existence of a nontrivial, that is, not identically equal to zero, solution to the system of Eqs. (22), compatible with the boundary conditions. Then the solution of Eq. (22a) will be

$$\zeta(\rho)=\begin{cases}c_1J_0\left(\frac{\pi\rho}{L}\left(\frac{P}{P_{Fr}}-1\right)^{1/2}\right) & \text{for } \rho\leqslant\rho_0\\ c_2K_0\left(\frac{\pi\rho}{L}\right) & \text{for } \rho>\rho_0\end{cases}, \tag{24}$$

where $c_1$ and $c_2$ are constants and $J_\nu$ and $K_\nu$ are Bessel functions. The condition of continuity of the logarithmic derivative at the point $\rho=\rho_0$ gives

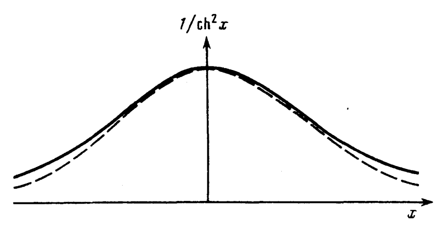


FIG. 2. Form of the function $1/\cosh^2x$. The dashed line shows the function $\exp(-x^2)$.

$$\left(\frac{P}{P_{\mathrm{Fr}}}-1\right)^{1/2} J_1\left(\frac{\pi\rho_0}{L}\left(\frac{P}{P_{\mathrm{Fr}}}-1\right)^{1/2}\right)\Big/ J_0\left(\frac{\pi\rho_0}{L}\left(\frac{P}{P_{\mathrm{Fr}}}-1\right)^{1/2}\right) = K_1\left(\frac{\pi\rho_0}{L}\right)\Big/ K_0\left(\frac{\pi\rho_0}{L}\right). \tag{25}$$

In the case $\pi\rho_0/L \ll 1$, if in addition we assume that

$$\frac{\pi\rho_0}{L}\left(\frac{P}{P_{\mathrm{Fr}}}-1\right)^{1/2} \ll 1, \tag{26}$$

we have from Eq. (25)

$$P_{\mathrm{th}} = P_{\mathrm{Fr}}\left[1-2L^2/\pi^2\rho_0^2 \ln(\pi\rho_0/L)\right]. \tag{27}$$

As seen from expression (27), the condition (26) is satisfied for $\ln(\pi\rho_0/L) \gg -2$, and this strengthens considerably the inequality $\pi\rho_0/L \ll 1$.

In the case where $\rho_0 \gtrsim L$, we obtain from Eq. (25)

$$P_{\mathrm{th}} = P_{\mathrm{Fr}}\left[1+\left(\frac{J_{01}}{\pi}\right)^2\left(\frac{L}{\rho_0}\right)^2\left(1-\frac{J_{01}LK_0(\pi\rho_0/L)}{\pi\rho_0 K_1(\pi\rho_0/L)}\right)^2\right]. \tag{28}$$

Here $J_{01} \approx 2.4$ is the first zero of the Bessel function $J_0(Z)$. In the limit $\rho_0 \gg L$ formula (28) gives $P_{\mathrm{th}} = P_{\mathrm{Fr}}$, in agreement with the result obtained in Sec. 3 for broad beams.

In the case where the beam radius is of the order of the cuvette thickness, $\pi\rho_0/L \approx 1$, Eq. (25) can be solved graphically. As a result we find

$$P_{\mathrm{th}} \approx 3.1 P_{\mathrm{Fr}}. \tag{29}$$

Therefore, in all the cases studied we have $P_{\mathrm{th}} \sim P_{\mathrm{Fr}} \sim 1/L^2$, whereas the coefficient of proportionality is determined by the ratio $\rho_0/L$ of the beam radius to the cuvette thickness.

In the case of more than a single constant the threshold of the Fredericks transition can be determined analytically for a ribbon beam of the form $P(\mathbf{r}_\perp) = P(y)$ polarized along the $x$ axis. The equation linearized in $\varphi(\mathbf{r})$ in this case has the form

$$K_{22}\frac{\partial^2\varphi}{\partial y^2} + K_{33}\frac{\partial^2\varphi}{\partial z^2} + \frac{\varepsilon_a\varepsilon_\perp^{1/2}}{c\varepsilon_\parallel}P(y)\varphi = \eta\frac{\partial\varphi}{\partial t}. \tag{30}$$

Introducing the new variable $y' = (K_{33}/K_{22})^{1/2}y$, Eq. (30) can be written in the form (17) by replacing $x$ by $y$ and using the definition of $P_{\mathrm{Fr}}$ from (12). After using the solution to (17), we obtain for a light wave of the form $P(y) = P_0/\cosh^2\beta y$

$$P_{\mathrm{th}} = P_{\mathrm{Fr}}\left[1+\frac{\beta L}{\pi}\left(\frac{K_{22}}{K_{33}}\right)^{1/2}\right]. \tag{31}$$

Therefore, when the beam dimensions are slightly smaller than the curvette thickness the inclusion of more than one constant leads to small corrections. We can expect this to hold for other beam shapes, also.

## 5. HYSTERESIS OF THE FREDERICKS TRANSITIONS IN BROAD BEAMS

Formula (11) becomes invalid for the maximum angle of deviation of the director from the unperturbed direction if the parameters of the liquid-crystal medium are such that

$$B = \frac{1}{4}\left(1-\frac{9}{4}\frac{\varepsilon_a}{\varepsilon_\parallel}-k\right) \leqslant 0.$$

This occurs, for example, for the nematic crystal PAA, for which $\varepsilon_a \approx 0.9$; $K_{11} = 4.5 \times 9.5 \times 10^{-7}$ dyne [at the temperature $T = 125$°C (Ref. 13)] and $B = -0.03$ ($B = 0.7$ for MBBA and for $B = 0.06$ for OCBP).

To study the Fredericks transition for $B \leqslant 0$ we integrate Eq. (10) with respect to $z$ after multiplying it by $2d\varphi/dz$. After determining the integration constant in terms of the maximum angle of deviation of the director from the unperturbed direction $\varphi_m$ we obtain

$$\left(\frac{d\varphi}{dz}\right)^2 = \frac{\varepsilon_\perp^{1/2}P}{cK_{33}}\frac{[1-(\varepsilon_a/\varepsilon_\parallel)\sin^2\varphi]^{1/2}-[1-(\varepsilon_a/\varepsilon_\parallel)\sin^2\varphi_m]^{1/2}}{(1-k\sin^2\varphi)[1-(\varepsilon_a/\varepsilon_\parallel)\sin^2\varphi]^{1/2}[1-(\varepsilon_a/\varepsilon_\parallel)\sin^2\varphi_m]^{1/2}}. \tag{32}$$

Taking into account the fact that the maximum angle of deviation $\varphi_m$ is reached at the center of the cell, that is, for $z = L/2$, we get from (32)

$$\int_0^{\varphi_m}\left\{\frac{(1-k\sin^2\varphi)[1-(\varepsilon_a/\varepsilon_\parallel)\sin^2\varphi]^{1/2}[1-(\varepsilon_a/\varepsilon_\parallel)\sin^2\varphi_m]^{1/2}}{[1-(\varepsilon_a/\varepsilon_\parallel)\sin^2\varphi]^{1/2}-[1-(\varepsilon_a/\varepsilon_\parallel)\sin^2\varphi_m]^{1/2}}\right\}^{1/2} d\varphi = \frac{L}{2}\left\{\frac{\varepsilon_\perp^{1/2}P}{cK_{33}}\right\}^{1/2}. \tag{33}$$

Assuming that the power density $P$ of the radition falling on a cell of the NLC is close to the threshold value for the Fredericks transition, that is, $\varphi_m \ll 1$, let us compute the integral in (33) up to terms $\sim\varphi_m^4$. The solution of the resulting biquadratic equation for $\varphi_m$ has the form

$$\varphi_m^2 = \frac{-B \pm (B^2-4GC)^{1/2}}{2G}, \tag{34}$$

where

$$G = \frac{1}{96}\left[\frac{11}{2}+\frac{9}{4}\frac{\varepsilon_a}{\varepsilon_\parallel}-k+\frac{63}{4}k\frac{\varepsilon_a}{\varepsilon_\parallel}-\frac{9}{2}k^2-\frac{261}{32}\left(\frac{\varepsilon_a}{\varepsilon_\parallel}\right)^2\right],$$

$$C = 1-(P/P_{\mathrm{Fr}})^{1/2}.$$

For the liquid crystals MBBA, OCBP, and PAA the values of $G$ are almost identical at $G \approx 0.06$. Before discussing formula (34), we note that it can in principle be obtained from the condition of the minimum of the free energy after expanding it in a series in $\varphi_m$ up to terms $\sim\varphi_m^6$. Apart from a coefficient that renders it dimensionless, the free energy has the form

$$\Phi = C\varphi_m^2 + {}^1/_2 B\varphi_m^4 + {}^1/_3 G\varphi_m^6. \tag{35}$$

At the point determined by formula (34) we have

$$d^2\Phi/d(\varphi_m^2)^2 = \pm(B^2-4GC)^{1/2}. \tag{36}$$

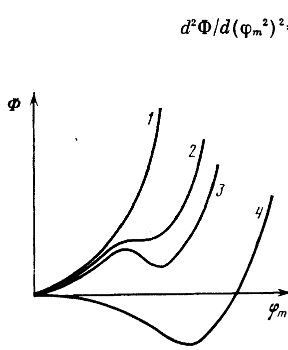


FIG. 3. Dependence of the free energy $\Phi$ on $\varphi_m$ for different values of the light field power $P$: 1—$P < P_{\mathrm{th}}$, 2—$P = P_{\mathrm{th}}$, 3—$P_{\mathrm{th}} < P < P_{\mathrm{Fr}}$, —$P > P_{\mathrm{Fr}}$.

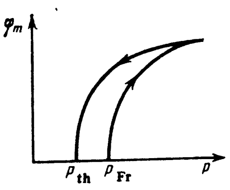


FIG. 4. Hysteresis of the Fredericks transition. The arrows indicate the direction of variation of the power of the light field $P$.

Therefore, we should use the plus sign in formula (34) (the condition that the free energy be a minimum).

If $B \geq 0$ the quantity $\varphi_m$ will be real at $C = 1 - (P/P_{Fr})^{1/2} \leq 0$, that is, $P_{th} = P_{Fr}$. If $B < 0$, then in order that $\varphi_m$ be real it is sufficient to require that the expression under the square root in (34) be positive, $B^2 - 4GC \geq 0$, from which we have

$$P_{th} = P_{Fr}(1 - B^2/4G)^2. \quad (37)$$

However, we see from (36) and (37) that when the power density of the incident radiation is $P = P_{th}$ the quantity $\varphi_m$ given by (34) is a point of inflection for the free energy function (see Fig. 3). As seen from Fig. 3, for values $P_{th} < P < P_{Fr}$ the function $\Phi(\varphi_m)$ has a local minimum at $\varphi_m \neq 0$, where

$$\Phi(\varphi_m \neq 0) > \Phi(\varphi_m = 0) = 0. \quad (38)$$

The point $\varphi_m = 0$ becomes unstable only at $P > P_{Fr}$. From the above discussion it is clear that as the power density of the light wave increases from zero the Fredericks transition occurs at $P = P_{Fr}$, so that a free-energy minimum corresponding to $\varphi_m \neq 0$ and occurring at $P_{th} < P < P_{Fr}$ is unattainable because of the presence of the potential barrier. For the inverse transition, that is, as $P$ decreases from the region of larger $P_{Fr}$, the free-energy minimum reached at $P < P_{Fr}$ and corresponding to $\varphi_m \neq 0$, while not an absolute minimum of the function $\Phi$ [as seen from (38)], is separated from it by the potential barrier. The barrier only disappears at $P = P_{th}$ and then we have $\varphi_m = 0$ (Fig. 4).

## 6. PROPAGATION OF A LIGHT WAVE AT SMALL ANGLES TO THE DIRECTOR

As already noted, an extraordinary wave propagating at an angle to the director induces strong nonlinear optical effects at very low powers $P \ll P_{Fr}$ (Refs. 3–9). When a light wave is normally incident on a cuvette with homeotropic orientation of the NLC, however, reorientation of the director is possible only above some threshold value of the power of the wave. The distortions of

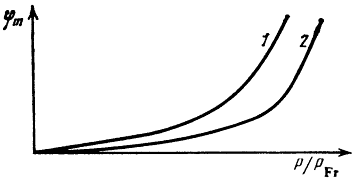


FIG. 5. Dependence of the maximum angle of deviation $\varphi_m$ on $P/P_{Fr}$ in the case of a light wave incident on a cell at small angles: 1—$\gamma = 10°$, 2—$\gamma = 5°$.

the field of the director in this case were discussed in Secs. 3–5. Here we shall study the nature of the reorientation of the director by the field of an extraordinary light wave in the intermediate region, that is, when the wave propagates at small angles to the director.

Substituting the expressions for the light fields (8a) into Eq. (5) and including terms up to third order in the small quantity $\varphi = \varphi_m \sin(\pi z/L)$ (that is, assuming that the power of the wave is less than or of order $P_{Fr}$) and of first order in $s = \sin\alpha$, we find

$$\left[\left(1 - \frac{9}{4}\frac{\varepsilon_a}{\varepsilon_\parallel}\right)\frac{P}{P_{Fr}} - k\right]\varphi_m^3 + 2\left(1 - \frac{P}{P_{Fr}}\right)\varphi_m + \frac{4\sin\gamma}{\pi}\frac{P}{P_{Fr}} = 0, \quad (39)$$

where $\sin\gamma = \varepsilon^{-1/2}\sin\alpha$ and $\gamma$ is the angle of refraction of the wave in the cell of the NLC.

In the case of weak fields $P \ll P_{Fr}$ and large anisotropy of the permittivity, Eq. (39) duplicates the results obtained earlier[4]:

$$\varphi_m = \varepsilon_a |E|^2 L^2 \sin\gamma / 2\pi^4 K_{33}.$$

Since the formulas are quite awkward we shall not give the analytic form of the solution of Eq. (39). The graph of the function $\varphi_m(P/P_{Fr})$ is shown in Fig. 5 for the parameters of the liquid crystal OCBP and for different angles $\gamma$.

The behavior of the cell in the oblique-incidence case discussed here is analogous to the case of a cell in an oblique magnetic field-in both cases there is no rigorous Fredericks transition (cf. Ref. 13, §4.2.3).

## 7. DISCUSSION

Let us estimate numerically the value of the threshold power for the Fredericks transition for the nematic crystal OCBP used in Ref. 11. The parameters of the liquid crystal are $\varepsilon_\parallel^{1/2} = 1.66$, $\varepsilon_\perp^{1/2} = 1.53$, $K_{11} = 4 \times 10^{-7}$ dyne, $K_{33} = 7 \times 10^{-7}$ dyne, the curvette thickness is $L = 150$ $\mu$m, and the beam radius is $\rho_0 = 50$ $\mu$m. Substituting the Frank constant $K = 0.5$ $(K_{11} + K_{33})$ into the expression for $P_{Fr}$, we find from formula (29)

$$P_{th} \approx 9{,}7 \cdot 10^2 \text{ Watt/cm}^2$$

This value is in very good agreement with the experimental value[11]

$$P_{th}^{exp} \geq 9 \cdot 10^2 \text{ Watt/cm}^2$$

For the cuvette thickness $L = 50$ $\mu$m and the same values of all the other parameters the value of the threshold power estimated from (28) is

$$P_{th} \approx 3 \cdot 10^3 \text{ Watt/cm}^2$$

This value is 3.1 times larger than that for the Fredericks transition in a cuvette of thickness $L = 150$ $\mu$m, which is also in very good agreement with the results of Ref. 11.

The temperature dependence of the threshold power is determined by the temperature dependence of the Frank constant and of the permittivity. Bearing in mind the fact that the Frank constant $K$ varies with temperature as the square of the order parameter $K \sim S^2$ while the anisotropy of the permittivity $\varepsilon_a \sim S$ (Ref. 12), we can separate the temperature-dependent part in expression (12) for $P_{Fr}$ in the form $P_{Fr} \propto \varepsilon_a \varepsilon_{\|} \varepsilon_{\perp}^{-1/2}$. As the temperature increases, $\varepsilon_a$ and $\varepsilon_{\|}$ decreases while $\varepsilon_{\perp}$ increases; from this we see that as the temperature increases the threshold power decreases. Introducing the notation

$$\varepsilon_0 = {}^1/_3 \mathrm{Sp}\,(\varepsilon_{ik}) = {}^1/_3(\varepsilon_{\|}+2\varepsilon_{\perp}),$$

we can write $P_{Fr} \propto \varepsilon_0^{1/2} \varepsilon_a$. Since $\varepsilon_0^{1/2}$ depends weakly on the temperature (for example, in the region of the MBBA mesophase $\varepsilon_0^{1/2}$ changes by only 0.01 of its value), as the point of the phase transition to an isotropic liquid is approached the threshold power decreases in proportion to $\varepsilon_a$, that is, to the order parameter $S$.

Here we note the following. At small values of $\varepsilon_a/\varepsilon_{\|}$ and $k$, as is the case near the critical point, Eq. (33) determining the maximum angle of deviation of the director $\varphi_m$ as a function of the power of the light field can be simplified. For this we must expand the integrand in powers of $\varepsilon_a/\varepsilon_{\|}$ and $k$. Then the integral on the left-hand side of Eq. (33) is expressed in terms of elliptic integrals and the resulting equation implicitly determines the function $\varphi_m = \varphi_m(P)$. In the limit where $P/P_{Fr} \ll 1$ (that is, $\varphi_m = \pi/2 - \delta, \delta \ll 1$) the function $\varphi_m(P)$ can be determined explicitly:

$$\delta \propto \exp\,[-(\pi/2)\,(P/P_{Fr})^{1/2}].$$

As was shown in Sec. 5, for certain types of liquid crystals the Fredericks transition can be accompanied by hysteresis. Let us calculate for the case of the well studied liquid crystal PAA the main characteristics of this phenomenon. As seen from formula (37) with $B = -0.03$ and $G = 0.06$, the powers at which the Fredericks effect in PAA is switched on and off differ little: $P_{th} \approx 0.992 P_{Fr}$. The angle corresponding to the appearance of a local minimum of the free-energy function [that is, $\varphi_m$ corresponding to the inflection point of the function $\Phi(\varphi_m)$] is $\varphi_m \propto (-B/2G)^{1/2} = 0.5$.

Let us demonstrate that thermal fluctuations of the direction of the director do not lead to surmounting of the potential barrier between the minima of the free energy function. For this we note that thermal fluctuations of the direction of the director would cause the hysteresis to disappear if the mean angle of the fluctuation deviations were on the order of or larger than the difference of the angles corresponding to the local minimum and maximum of the free energy. This difference is easily calculated using formula (34) for some value of the power $P$ such that $P_{th} < P < P_{Fr}$ (that is, $0 < C < B^2/4D$). For example, for $C = B^2/8D$ we find $\Delta\varphi = 0.4$, and since $\langle(\delta\varphi)^2\rangle^{1/2} \ll 1$ for thermal fluctuations of the director the local minimum of the free energy $\Phi(\varphi_m > 0)$ is stable to thermal fluctuations.

In concluding this discussion we note that a Fredericks transition induced by light fields in a NLC with $\varepsilon_a > 0$ can occur only when the light wave is normally incident on a homeotropically oriented NLC cell. It is impossible to have a Fredericks transition induced by an ordinary light wave normally incident on a planarly oriented NLC cell. This can be understood when it is realized that the electric field of an ordinary light wave in a NLC remains perpendicular to the director as it propagates through the medium. Strong nonlinear optical effects arise if the polarization of the normally incident cell makes some angle with the director. We have considered these effects in an earlier study.[6]

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*Note added in proof.* We have been kindly informed by the authors of a paper submitted to this journal that they have also studied the theory of light-induced Fredericks transitions (A. S. Zolot'ko, V. F. Kitaeva, N. N. Sobolev, and A. P. Sukhorukov, Zh. Eksp. Teor. Fiz. 81, 933 (1981) [Sov. Phys. JETP 54, No. 9 (1981)]).

[1]S. M. Arakelyan, G. A. Lyakhov, and Yu. S. Chilingaryan, Usp. Fiz. Nauk **131**, 3 (1980) [Sov. Phys. Uspekhi **23**, 245 (1980)].
[2]S. M. Arakelyan, G. L. Grigoryan, S. Ts. Nersisyan, M. A. Nshanyan, and Yu. S. Chilingaryan, ZhETF Pis. Red. **28**, 202 (1978) [JETP Lett. **28**, 186 (1979)].
[3]B. Ya. Zel'dovich and N. V. Tabiryan, ZhETF Pis. Red. **30**, 510 (1979) [JETP Lett. **30**, 478 (1980)].
[4]B. Ya. Zel'dovich, N. F. Pilipetskiĭ, A. V. Sukhov, and N. V. Tabiryan, ZhETF Pis. Red. **31**, 287 (1980) [JETP Lett. **31**, 263 (1980)].
[5]B. Ya. Zel'dovich and N. V. Tabiryan, Preprint FIAN, No. 61, 1980.
[6]B. Ya. Zel'dovich and N. V. Tabiryan, Preprint FIAN, No. 62, 1980.
[7]B. Ya. Zel'dovich and N. V. Tabiryan, Preprint FIAN, No. 63, 1980.
[8]B. Ya. Zel'dovich and N. V. Tabiryan, Kvant. Elektronika (Moscow) **7**, 770 (1980) [Sov. J. Quantum Electronics **10**, 440 (1980)].
[9]N. F. Pilipetski, A. V. Sukhov, N. V. Tabiryan, and B. Ya. Zel'dovich, Optical Communications **37**, 280 (1981).
[10]V. B. Pakhalov, A. S. Tumasyan, and Yu. S. Chilingaryan, Abstracts of Reports at the Tenth Int. Conf. on Coherent and Nonlinear Optics, Kiev, October 14–17, 1980, Part I, p. 18.
[11]A. S. Zolot'ko, V. F. Kitaeva, N. Kroo, N. N. Sobolev, and L. Chilag, ZhETF Pis. Red. 32, 170 (1980) [JETP Lett **32**, 158 (1981)].
[12]P. G. De Gennes, The Physics of Liquid Crystals, Oxford University Press, 1974 (Russ. transl., Mir, 1977).
[13]L. M. Blinov, Elektro- i magnitooptika zhidkikh kristallov (Electro- and Magneto-optics of Liquid Crystals), Moscow, Nauka, 1978.
[14]Analiticheskie metody v teorii rasprostraneniya i difraktsii) voln (Analytic Methods in the Theory of Wave Propagation and Diffraction), ed. S. V. Butakovaya, Moscow, Sov. radio, 1970.
[15]B. Ya. Zel'dovich and N. V. Tabiryan, Zh. Eksp. Teor. Fiz. 79, 2388 (1980) [Sov. Phys. JETP **52**, 1210 (1980)].
[16]L. D. Landau and E. M. Lifshitz, Kvantovaya mekhanika, Moscow, Nauka, 1974 (Engl. transl. Quantum Mechanics: Nonrelativistic Theory, New York, Pergamon, 1977).

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