Optically induced Freedericksz transition and bistability in a nematic liquid crystal

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An Euler equation, consistent with Maxwell's equations, for describing the optically induced spatial reorientation of the director of a homeotropically oriented nematic liquid crystal is obtained for the case of normal incidence. The exact solution describing the orientation of the director is obtained. By examining the maximum deformation angle near the threshold, the threshold intensity and the criterion for the physical parameters that indicate whether the transition is first- or second-order at the threshold are obtained. The hysteresis accompanying the first-order Freedericksz transition is discussed. By choosing a compound (PAA, *p*-azoxyanisole) with suitable material parameters from known nematic liquid crystals, an experiment is proposed to observe, for the first time, a first-order Freedericksz transition in nematic liquid crystals. The Zel'dovich approach [Sov. Phys.—JETP 54, 32 (1981)] based on the geometrical-optics approximation is shown to be internally inconsistent and also inconsistent with the geometrical-optics approximation. The Euler equation using a self-consistent geometrical-optics approximation is also obtained, and turns out to be identical to our exact Euler equation, but different from the infinite-plane-wave approximation used by Durbin *et al.* [Phys. Rev. Lett. 47, 1411 (1981)]. Detailed comparisons between our approach and the Durbin approach are made. The dynamics of the transition are discussed and an approximate solution is given. The transient responses to the laser switch-on and switch-off are shown to have exponential time dependence. Finally, the effects of surface interactions on the transition are discussed and the exact solution is given. The procedure for determining the threshold, the saturation, and the parallel-state-maintenance intensities is given. We also discuss the first-order transition and propose experimental methods manifesting the effects of surface interaction. The criterion for the transition to be first order at any intensity is given.

I. INTRODUCTION

The nonlinear optical properties of liquid crystals in the isotropic phase have been studied quite extensively in the past decade.[1-8] The liquid-crystal phase was considered by Shelton and Shen who studied the normal and umklapp optical third-harmonic generation in cholesteric liquid crystals.[8,9] Their experimental results agreed well with their theoretical predictions. In 1979, Herman and Serinko suggested that larger optical wave-mixing efficiency could be obtained in the nematic liquid-crystal (NLC) phase than in the isotropic phase, if the NLC is maintained in an external magnetic field near the Freedericksz transition.[10] The theory has been extended and verified experimentally by Khoo and Zhuang.[11] Recently, the purely optical-field-induced nonlinear interaction between a normally incident light wave and a homeotropically oriented NLC cell has received a great deal of attention.[12,13] Studies have shown that there exists a characteristic threshold intensity below which no molecular reorientation can be induced. This effect was explained qualitatively by Zolot'ko *et al.* in 1980.[14] A quantitative theory of the optically induced Freedericksz transition was constructed by Zel'dovich, Tabiryan, and Chilingaryan[15] (hereafter referred to as the Zel'dovich approach), using the geometrical-optics approximation in 1981.[16] They obtained the solution for the spatial distribution of the director and predicted that near the threshold the maximum deformation angle is proportional to the square root of the excess of the intensity above the threshold intensity and that for certain NLC's the Freedericksz transition is accompanied by hysteresis, which they claimed has no analog in the case of a static dc field.[15,17] In fact, it was shown by Deuling and Helfrich[18,19] in 1974 that for a twisted NLC cell and a conducting nematic cell with a NLC of strong conductive anisotropy and a dielectric anisotropy of opposite sign the Freedericksz transition can be first order and one can obtain hysteresis. In 1981, Durbin, Arakelian, and Shen[20] (hereafter referred to as the Durbin approach) reported the first observation of the optically induced Freedericksz transition in nematic 5CB (4-cyano-4′-pentylbiphenyl); their results were in quantitative agreement with their theoretical predictions using the infinite-plane-wave approximation. Also in 1982, Khoo presented an approximate solution that assumed equal elastic constant and small optical dielectric anisotropy.[12,13] Explicit analytic expressions were obtained in the small-angle linearized approximation. Khoo also made a quantitative experimental verification of the associated nonlinear optical processes. It is because of their large anisotropies and associated nonlinear effects that NLC's have received considerable attention. Experimental results show that the optical nonlinearity of NLC's has a value larger by eight to ten orders of magnitudes than that of carbon disulfide (CS_2) and a name "gigantic optical nonlinearity" (GON) has been given by Zel'dovich to such large nonlinearities.[12-16,20] While Zel'dovich and Durbin take these large anisotropies into account, approximations were made which do not properly describe the resulting nonlinear effects. Clearly, an exact theory is desirable for a rigorous study of the interaction between the optical field and the NLC.

The study of the action of an optical field on a NLC is complicated because the field is propagating in an inhomogeneous anisotropic medium having a dielectric tensor depending on both the intensity of the optical field and the position, and therefore the field and the Poynting vector vary in space. Moreover, as appreciated by Durbin, not only the electric energy, but also the magnetic energy, of the optical field depend upon the orientation of the NLC molecules. A theory should encompass all of these aspects of the problem. Zel'dovich[15] realized that the complex amplitude of the electric field is to be determined in a self-consistent manner from the solution of Maxwell's equations. However, in order to obtain the Euler equation for the director, Zel'dovich found it necessary to assume that the amplitude of the electric field is fixed when varying the total free energy. After obtaining the Euler equation for the director, the geometrical-optics approximation was then introduced to obtain the final equation describing the director. Moreover, the magnetic energy of the incident beam was neglected and only the electric energy of the incident beam was assumed to be responsible for the molecular reorientation by the optical field. Evidently, such an approach is internally inconsistent since the time-averaged electric and magnetic energy densities are equal even within the accuracy of geometrical-optics approximation,[21] and the amplitudes of the fields are not fixed but depend on the local director.[15–16,22] A different approach is used by Durbin, in which the total electromagnetic energy density is written as the ratio of the magnitude of the Poynting vector to the ray velocity.[20] The infinite-plane-wave approximation in which the magnitude of the Poynting vector is assumed to be a constant is then introduced.

In this paper, we present an exact solution, which is consistent with Maxwell's equation, for describing the optically induced spatial reorientation of the director of a homeotropically oriented NLC for the case of normal incidence. By first showing that the time average of the z component of the Poynting vector is a constant throughout the medium, the total electromagnetic energy density can then be expressed as the ratio of the intensity of the incident field to the phase velocity, where the z axis is normal to the NLC cell surfaces. This conclusion is shown to agree with the geometrical-optics approximation, but disagrees with the infinite-plane-wave approximation. The Euler equations for the director, using the exact approach as well as a self-consistent geometrical-optics approximation, will then be obtained and compared with those obtained from the Zel'dovich approach and the Durbin approach. The Euler equations obtained from the first three approaches turn out to be the same, although the Zel'dovich approach is shown to be internally inconsistent and also inconsistent with the geometrical-optics approximation. We also obtain the exact solution describing the orientation of the NLC. In particular, we examine the maximum deformation angle near the Freedericksz threshold and obtain the criterion for the optically induced Freedericksz transition to be first order. The criterion shows that for a NLC with large dielectric and elastic anisotropies, the transition can be first order; the characteristic of being first order is more favored in the Durbin approach than this approach. By examining the material parameters from known NLC's, we show that the observation of a first-order Freedericksz transition is possible in PAA (p-azoxyanisole) in this approach and is possible in many NLC's in the Durbin approach. The hysteresis accompanying the first-order Freedericksz transition is shown to be similar to that proposed by Deuling and Helfrich[18,19] for a twisted NLC and a conducting NLC cell in a static electric field, but is different from that proposed by Zel'dovich. In our prediction, the rising and falling transitions exhibit the same deformation for intensities greater than the rising threshold intensity, and the changes at both the rising and falling threshold intensities are discontinuous.

We also discuss the dynamics of the transition. The transient responses to the laser switch-on and switch-off are shown to have exponential time dependence. Finally, the effects of interfacial interactions between the NLC and the surfaces on the molecular reorientation are discussed and the exact solution is given. The results show that as the anchoring strength decreases, the threshold intensity will be lower and there will be a saturation intensity above which the NLC will orient parallel to the surfaces. Once the parallel state is attained, the minimum intensity needed to maintain the parallel state can be different from the saturation intensity and can even be less than the threshold intensity. The criterion for the existence of the first-order transition at the threshold intensity with finite anchoring is shown to be different from the case of rigid anchoring strength. By examining the maintenance intensity as a function of the maximum deformation angle, we obtain the general criterion for the existence of a first-order transition. Using the dependence of the maximum deformation on the anchoring strength and the cell thickness, we propose three simple experimental methods to manifest the effects of finite anchoring on the transition.

In the following sections we first discuss the exact Euler equation and the Durbin approach. We then make a detailed comparison between the self-consistent geometrical-optics approximation and the Zel'dovich approach in Sec. III. A section on the solution describing the orientation of the NLC follows, including a discussion of the first-order transition and the experimental observation. Finally, the dynamic behavior and the effects of the interfacial interaction on the transition are discussed.

II. EXACT EULER EQUATION AND DURBIN APPROACH

Let us consider a homeotropically oriented NLC cell of thickness d confined between the planes $z=0$ and $z=d$ of a Cartesian coordinate system. In the cell, the average direction of NLC orientation is given by the director $\hat{n}(\vec{r})$. The NLC director always lies in the xz plane and in the absence of a light beam, the directors are parallel to the z axis everywhere. We let $\theta(z)$ be the tilt angle between the director and the z axis. Then the director can be described by $\hat{n}(\vec{r})=\hat{n}(z)=(\sin\theta,0,\cos\theta)$. A harmonic time-dependent light beam of complex amplitude $\vec{E}$ is normally incident on the NLC medium with the polarization parallel to the plane of incidence, which is the xz plane.

We shall consider the equilibrium orientation of the director. Then the NLC can be considered as a nonconducting and nondissipative medium where no mechanical work is done. On taking the time average of the energy flow

$$\frac{\partial F_{\rm opt}}{\partial t}+{\rm div}\vec{S}=0 \,, \tag{2.1}$$

we find that

$$\mathrm{div}\langle\vec{S}\rangle=0 \,, \tag{2.2}$$

where $F_{\rm opt}$ is the energy and $\vec{S}=(c/4\pi)\vec{E}\times\vec{H}$ is the Poynting vector of the optical field. Therefore, we conclude that the time average of the z component of the Poynting vector is a constant throughout the medium. To show that the magnitude of the Poynting vector is not a constant, it is sufficient for us to show that $S_x(z)$ is not a constant even for the case of normal incidence. By Maxwell's equation $\mathrm{curl}\vec{H}=(1/c)\partial\vec{D}/\partial t$, we obtain $D_z=0$ for a normally incident wave. Consequently, using $\vec{D}=\hat{\epsilon}\cdot\vec{E}$, we have $E_z=-(\epsilon_{13}/\epsilon_{33})E_x$ and $\vec{S}=S_z(\epsilon_{13}/\epsilon_{33},0,1)$. Since $\epsilon_{13}/\epsilon_{33}$ varies with z, we conclude that S_x and hence S are not constant, but S_z is a constant. This conclusion agrees with the geometrical-optics approximation,[15,16,23] in which the time average of the z component of the Poynting vector is a constant, but disagrees with the infinite-plane-wave approximation in which the time average of the magnitude of the Poynting vector is a constant.

In general, the wave vector of the fields can be expressed as $\omega\vec{n}_p/c$, where $c/|\vec{n}_p|$ is the phase velocity and ω is the frequency of the incident wave. In terms of $\vec{n}_p$, the electric and the magnetic energy densities can be written, respectively, as

$$F_e=\vec{E}\cdot\vec{D}/8\pi=-\vec{E}\cdot(\vec{n}_p\times\vec{H})/8\pi \tag{2.3}$$

and

$$F_m=\vec{B}\cdot\vec{H}/8\pi=\vec{H}\cdot(\vec{n}_p\times\vec{E})/8\pi \,. \tag{2.4}$$

Consequently, F_e and F_m are both equal to $\vec{S}\cdot\vec{n}_p/2c$, so that F_e and F_m will make equal contributions to the free energy although the coupling to the director is entirely through a torque applied by the electric field. The total electromagnetic energy density can then be written as $F_{\rm opt}=\vec{S}\cdot\vec{n}_p/c$. Thus, for a normally incident wave, $\vec{n}_p=(0,0,n_p)$ and the total electromagnetic energy density can be expressed as the ratio of the z component of the Poynting vector to the phase velocity, i.e., $S_z n_p/c$ with

$$n_p=n_0n_e/(n_0^2\sin^2\theta+n_e^2\cos^2\theta)^{1/2} \,, \tag{2.5}$$

where $n_0=\sqrt{\epsilon_\perp}$ and $n_e=\sqrt{\epsilon_\parallel}$ are ordinary and extraordinary refractive indices respectively, and $\epsilon_\perp$ and $\epsilon_\parallel$ are the dielectric constants perpendicular and parallel to the local director at the optical frequency ω. Since $S_z n_p=Sn_r$, the electromagnetic energy density can also be written as Sn_r/c, where S is the magnitude of the Poynting vector, c/n_r is the ray velocity with

$$n_r=[(\epsilon_\perp+\epsilon_\parallel)/\epsilon_\perp\epsilon_\parallel-(\epsilon_\perp+\epsilon_a\cos^2\theta)^{-1}]^{-1/2} \,, \tag{2.6}$$

and $\epsilon_a=\epsilon_\parallel-\epsilon_\perp$ is the dielectric anisotropy at the optical frequency ω. The last expression is used by Durbin with the infinite-plane-wave approximation in which $\langle S\rangle$ is a constant throughout the medium. It is therefore of interest to compare the solutions obtained by our approach and that obtained by Durbin. In what follows, we shall refer to our approach as approach I, and the Durbin approach as approach II. In approach I, $\langle S_z\rangle$ was shown to be constant, whereas in approach II, $\langle S\rangle$ is assumed to be constant, and indeed the constant is the intensity of the incident field, I (if the scattering loss in traversing the medium can be neglected). Thus the total electromagnetic energy density will be written as

$$F_{\rm opt}=I\tilde{n}(\theta)/c \,, \tag{2.7}$$

where $\tilde{n}=n_p$ for the approach I and $\tilde{n}=n_r$ for approach II.

We shall first consider the static orientation and ignore the fluctuations of the field and the director. Therefore the total free energy per unit volume of the NLC can be written as

$$\mathscr{F}=\int\left[\tfrac{1}{2}k_{11}(\mathrm{div}\hat{n})^2+\tfrac{1}{2}k_{22}(\hat{n}\cdot\mathrm{curl}\hat{n})^2+\tfrac{1}{2}k_{33}(\hat{n}\times\mathrm{curl}\hat{n})^2-\frac{I}{c}\tilde{n}(\theta)\right]d^3r \,, \tag{2.8}$$

where k_{11}, k_{22}, and k_{33} are the splay, twist, and bend elastic constants. The Euler equation for $\hat{n}(z)$ resulting from the variation of the free energy has the following form in the stationary case:

$$(1-k\sin^2\theta)\frac{d^2\theta}{dz^2}-k\sin\theta\cos\theta\left[\frac{d\theta}{dz}\right]^2+\frac{In_0}{ck_{33}}\frac{(\beta-\alpha)\sin\theta\cos\theta}{(1-\alpha\sin^2\theta)^{1/2}(1-\beta\sin^2\theta)^{3/2}}=0 \,, \tag{2.9}$$

where $k=(k_{33}-k_{11})/k_{33}$ and α and β are defined in Table I. The exact solution of the Euler equation (2.9) will be given in Sec. IV.

III. GEOMETRICAL-OPTICS APPROXIMATIONS AND ZEL'DOVICH APPROACH

A. Electromagnetic field and energy

We first discuss the solutions of the fields in the geometrical-optics approximation in which the NLC medium is assumed to be a slowly varying dielectric medium so that

$$\frac{\lambda}{\pi}\left|\frac{d}{dz}\frac{1}{\sqrt{\epsilon_{ij}}}\right|\ll 1 \tag{3.1}$$

is satisfied, where λ is the wavelength of the incident light.

TABLE I. A comparison of the different parameters in approaches I and II. In the table, $k=(k_{33}-k_{11})/k_{33}$, $\epsilon_a=\epsilon_{\parallel}-\epsilon_{\perp}$, $u=1-\epsilon_{\perp}/\epsilon_{\parallel}$, $w=1-(\epsilon_{\perp}/\epsilon_{\parallel})^2$, and the CFFT stands for the criterion for first-order transition at the threshold intensity.

Parameter	Approach I	Approach II
$\tilde{n}$	$\dfrac{n_0}{(1-u\sin^2\theta)^{1/2}}$	$n_0\left[\dfrac{1-u\sin^2\theta}{1-w\sin^2\theta}\right]^{1/2}$
I	$\langle S_z\rangle$	$\langle S\rangle$
α	0	u
β	u	w
I_{Fr}	$ck_{33}(\epsilon_{\parallel}/n_0\epsilon_a)(\pi/d)^2$	$ck_{33}(\epsilon_{\parallel}^2/n_0\epsilon_{\perp}\epsilon_a)(\pi/d)^2$
B	$(1-k-9u/4)/4$	$(1-k-9w/4-3u/4)/4$
G	$(11/2-k+9u/4+63u/4-9k^2/2-261u^2/32)/96$	$(11/2-k+9w/4+3u/4+63kw/4+153wu/16$ $+3ku-9k^2/2-261w^2/32-189u^2/32)/96$
CFFT	$\dfrac{k_{11}}{k_{33}}+\dfrac{9}{4}\dfrac{\epsilon_{\perp}}{\epsilon_{\parallel}}<\dfrac{9}{4}$	$\dfrac{k_{11}}{k_{33}}+\dfrac{3}{4}\dfrac{\epsilon_{\perp}}{\epsilon_{\parallel}}+\dfrac{9}{4}\left[\dfrac{\epsilon_{\perp}}{\epsilon_{\parallel}}\right]^2<3$

Since the magnetic anisotropy for a typical NLC is of the order of 10^{-7} which is much smaller than the typical dielectric anisotropy ($\sim10^{-1}$), NLC's can be considered as nonmagnetic media and the magnetic permeability can be set to unity throughout all space.[24] By eliminating the magnetic field in the usual manner from Maxwell's equations, we obtain

$$\vec{H}=\frac{1}{ik_0}\mathrm{curl}\vec{E}\ , \tag{3.2}$$

and

$$\nabla^2\vec{E}-\mathrm{grad}\,\mathrm{div}\vec{E}+k_0^2\hat{\epsilon}\cdot\vec{E}=0\ , \tag{3.3}$$

where $k_0=\omega/c$. The dielectric tensor of the NLC is given by[24]

$$\epsilon_{ij}=\epsilon_{\perp}\delta_{ij}+\epsilon_a n_i n_j\ . \tag{3.4}$$

With $\hat{n}=(\sin\theta,0,\cos\theta)$, $\hat{\epsilon}$ can be written as

$$\hat{\epsilon}=\begin{bmatrix}\epsilon_{\perp}+\epsilon_a\sin^2\theta & 0 & \epsilon_a\sin\theta\cos\theta\\ 0 & \epsilon_{\perp} & 0\\ \epsilon_a\sin\theta\cos\theta & 0 & \epsilon_{\perp}+\epsilon_a\cos^2\theta\end{bmatrix}\ . \tag{3.5}$$

For a normally incident p-polarized wave, $\vec{E}=(E_x,0,E_z)$ and $\vec{H}=(0,H_y,0)$. Equations (3.2), (3.3), and (3.5) give

$$H_x=\frac{1}{ik_0}\frac{dE_x}{dz}\ ,\qquad E_z=-\frac{\epsilon_{13}}{\epsilon_{33}}E_x\ , \tag{3.6}$$

and E_x satisfies the wave equation

$$\frac{d^2E_x}{dz^2}+k_0^2\frac{\epsilon_{\perp}\epsilon_{\parallel}}{\epsilon_{33}}E_x=0\ . \tag{3.7}$$

In the geometrical-optics approximation, the fields are given by[25]

$$E_x(z)=A\epsilon_{33}^{1/4}\exp\left[ik_0n_0n_e\int\epsilon_{33}^{-1/2}(z)dz\right]\ ,$$

$$E_z(z)=-(\epsilon_{13}/\epsilon_{33})E_x\ ,$$

and

$$H_y(z)=(n_0n_e/\sqrt{\epsilon_{33}})E_x\ . \tag{3.8}$$

With the use of $\vec{S}=(c/4\pi)\vec{E}\times\vec{H}$, the time average of the Poynting vector is given by

$$\langle\vec{S}\rangle=\frac{cn_0n_e|A|^2}{8\pi}\left[\frac{\epsilon_{13}}{\epsilon_{33}},0,1\right]\ . \tag{3.9}$$

Thus, the magnitude of the time-averaged Poynting vector is not a constant since ϵ_{13} and ϵ_{33} vary with z. But the time average of the z component of the Poynting vector, $\langle S_z\rangle$, is a constant which is just the power flux density I since at $z=0$, $\epsilon_{13}=0$ and $\langle S_z\rangle=I$. The constant A relating to the amplitude of the electric field can be expressed in terms of I as follows:

$$|A|^2=8\pi I/cn_0n_e\ . \tag{3.10}$$

Equations (3.8) and (3.10) constitute the geometrical-optics approximation.

We now discuss the electromagnetic energy using the geometrical-optics approximation. To simplify the discussion, we let

$$F_{e1}=\frac{1}{16\pi}\epsilon_{\perp}|\vec{E}|^2\ ,$$

and

$$F_{e2}=\frac{1}{16\pi}\epsilon_a(\hat{n}\cdot\vec{E})(\hat{n}\cdot\vec{E}^*)\ . \tag{3.11}$$

Then the time-averaged electric energy density in the NLC is the sum of F_{e1} and F_{e2}. In the geometrical-optics approximation, F_{e1} and F_{e2} can be written as

$$F_{e1}=\frac{I}{2c}n_0n_e\epsilon_{\parallel}(1-w\sin^2\theta)/\epsilon_{33}^{3/2}\ ,$$

$$F_{e2}=\frac{I}{2c}n_0n_e\epsilon_{\perp}u\sin^2\theta/\epsilon_{33}^{3/2}\ ,$$

and

$$F_e=In_p/2c\ , \tag{3.12}$$

where $u=1-\epsilon_{\perp}/\epsilon_{\parallel}$ and $w=1-(\epsilon_{\perp}/\epsilon_{\parallel})^2$. By Eq. (3.8),

$H_y = n_p E_x$ and the time-averaged magnetic energy density $F_m = |H_y|^2/16\pi$ can also be written as $F_m = In_p/2c$. Therefore, within the accuracy of geometrical optics, the time-averaged electric and magnetic energy densities are equal. Such a result is true in general as we have shown in Sec. II.[21] Consequently, $F_{\rm opt}$ has the form

$$F_{\rm opt} = In_p/c \quad (3.13)$$

which is the same as Eq. (2.7) with $\tilde{n} = n_p$ that we obtained without the geometrical-optics approximation. This shows that $\langle S \rangle$ is not a constant and the infinite-plane-wave approximation fails even in the slowly varying media. Consequently, the equation describing the orientation of the director using the geometrical-optics approximation is the same as approach I.

B. Euler equation

In the following we discuss the Euler equation for the director using the self-consistent geometrical-optics approximation and compare with that obtained by Zel'dovich. The contributions to the Euler equation from the $F_{\rm opt}$ under the geometrical-optics approximation are of the form

$$\frac{d}{dz}\frac{\delta F_{e1}}{\delta\theta'}\bigg|_{\rm GOA} = \frac{d}{dz}\frac{\delta F_{e2}}{\delta\theta'}\bigg|_{\rm GOA} = 0 ,$$

$$\frac{\delta F_{e1}}{\delta\theta}\bigg|_{\rm GOA} = \frac{I}{4c} n_0 n_e \epsilon_{\parallel} \epsilon_a \sin 2\theta \times (2u - 1 - w\sin^2\theta)/\epsilon_{33}^{5/2} ,$$

$$\frac{\delta F_{e2}}{\delta\theta}\bigg|_{\rm GOA} = \frac{I}{4c} n_0 n_e \epsilon_{\perp} \epsilon_a \sin 2\theta (2 + u\sin^2\theta)/\epsilon_{33}^{5/2} , \quad (3.14)$$

and

$$\frac{\delta F_{\rm opt}}{\delta\theta}\bigg|_{\rm GOA} = \frac{I}{2c} n_0 n_e \epsilon_a \sin 2\theta/\epsilon_{33}^{3/2} ,$$

where $\theta' = \partial\theta/\partial z$ and GOA stands for the geometrical-optics approximation. The results (3.13) and (3.14) show that the Euler equation that obtained using the self-consistent geometrical-optics approximation is the same as the exact Euler equation that obtained using approach I.

In the Zel'dovich approach, the magnetic energy density is assumed not to affect the molecular orientation and the total electromagnetic energy density is set equal to the electric energy density. Such an approach is inconsistent with the geometrical-optics approximation and would predict only half of the orientational energy of an optical field. In addition, when varying the free energy, the amplitudes of the fields were assumed to be fixed and thereby the contribution of the electric energy density in the Euler equation for the director was overestimated by a factor of 2, which then canceled the factor of one-half induced by erroneously ignoring the contribution from the magnetic energy density. As a result, Zel'dovich was able to obtain the correct Euler equation using the geometrical-optics approximation.

The contributions from $F_{\rm opt}$ in the Zel'dovich approach (denoted by subscript Zel) are of the form

$$\frac{d}{dz}\frac{\delta F_{e1}}{\delta\theta'}\bigg|_{\rm Zel} = \frac{d}{dz}\frac{\delta F_{e2}}{\delta\theta'}\bigg|_{\rm Zel} = 0 ,$$

$$\frac{\delta F_{e1}}{\delta\theta}\bigg|_{\rm Zel} = 0 , \quad (3.15)$$

and

$$\frac{\delta F_{e2}}{\delta\theta}\bigg|_{\rm Zel} = \frac{1}{16\pi}\epsilon_a[\sin 2\theta(|E_x|^2 - |E_z|^2) + \cos 2\theta(E_x E_z^* + E_x^* E_z)] .$$

After obtaining the Euler equation for the director, Zel'dovich then used the geometrical-optics approximation, i.e., taking into account Eqs. (3.8) and (3.10), and obtained

$$\frac{\delta F_{e2}}{\delta\theta}\bigg|_{\rm Zel} = \frac{I}{2c} n_0 n_e \epsilon_a \sin 2\theta/\epsilon_{33}^{3/2} . \quad (3.16)$$

From Eqs. (3.14)–(3.16), we see that the contributions from F_e and F_m to the Euler equation in the Zel'dovich approach are different from those of the geometrical-optics approximation:

$$\frac{\delta F_{e1}}{\delta\theta}\bigg|_{\rm Zel} = 0 \neq \frac{\delta F_{e1}}{\delta\theta}\bigg|_{\rm GOA} ,$$

$$\frac{\delta F_{e2}}{\delta\theta}\bigg|_{\rm Zel} \neq \frac{\delta F_{e2}}{\delta\theta}\bigg|_{\rm GOA} , \quad (3.17)$$

$$\frac{\delta F_{m}}{\delta\theta}\bigg|_{\rm Zel} = 0 \neq \frac{\delta F_{m}}{\delta\theta}\bigg|_{\rm GOA} .$$

Table II summarizes the results. However, the overall contributions from $F_{\rm opt}$ have the form

$$\frac{\delta F_{\rm opt}}{\delta\theta}\bigg|_{\rm Zel} = \frac{\delta F_{e2}}{\delta\theta}\bigg|_{\rm Zel} = 2\frac{\delta(F_{e1}+F_{e2})}{\delta\theta}\bigg|_{\rm GOA} = \frac{\delta F_{\rm opt}}{\delta\theta}\bigg|_{\rm GOA} . \quad (3.18)$$

Therefore, the final Euler equation obtained by Zel'dovich is the same as the Euler equation obtained under the self-consistent geometrical-optics approximation, although the Zel'dovich approach is not internally consistent and also inconsistent with the geometrical-optics approximation.

TABLE II. A comparison of the contributions from the electric and magnetic energy densities to the Euler equation for the director between the self-consistent geometrical-optics approximation and the Zel'dovich approach. In the table, $\theta' = \partial\theta/\partial z$ and $M = (I/2c)n_0 n_e \epsilon_a \sin 2\theta/\epsilon_{33}^{3/2} = (I/c)(\delta n_p/\delta\theta)$.

Contribution	Geometrical-optics approximation	Zel'dovich approach
$\frac{d}{dz}\frac{\delta F_{\text{opt}}}{\delta\theta'}$	0	0
$\frac{\delta F_{e1}}{\delta\theta}$	$M\epsilon_{\parallel}(2u-1-w\sin^2\theta)/2\epsilon_{33}$	0
$\frac{\delta F_{e2}}{\delta\theta}$	$M\epsilon_{\perp}(2+u\sin^2\theta)/2\epsilon_{33}$	M
$\frac{\delta F_{e}}{\delta\theta}$	$M/2$	M
$\frac{\delta F_{m}}{\delta\theta}$	$M/2$	0
$\frac{\delta F_{\text{opt}}}{\delta\theta}$	M	M

IV. SOLUTION AND HYSTERESIS

A. Exact solution and threshold behavior

Now we consider the solution for the deformation angle of the NLC described by Eq. (2.9). In this section, rigid boundary conditions at the two interfaces are always assumed (i.e., $\theta=0$ at $z=0$ and $z=d$). The effects of finite anchoring on the transition are discussed in Sec. V B. In the rising transition, for $I<I_{\text{Fr}}$, $\theta=0$ is the solution which minimizes the free energy where I_{Fr} is the threshold intensity below which no molecular reorientation can be induced. By the symmetry of the problem, we look for solutions which are symmetrical with respect to the $z=d/2$ plane, that is, solutions satisfying

$$\theta(z)=\theta(d-z)\ . \tag{4.1}$$

Consequently $d\theta/dz=0$ at $z=d/2$, and minimization of total free energy requires that the maximum deformation angle θ_m is attained at the center of the cell, $\theta_m=\theta(z=d/2)$.

Together with the above conditions at $z=d/2$, Eq. (2.9) can be integrated to give for $d\theta/dz\neq 0$

$$\left[\frac{d\theta}{dz}\right]^2=\frac{2I}{ck_{33}}\frac{\tilde{n}(\theta_m)-\tilde{n}(\theta)}{1-k\sin^2\theta}\ . \tag{4.2}$$

With the use of the boundary conditions, integration of Eq. (4.2) yields for $0\leq z\leq d/2$,

$$z=\left[\frac{ck_{33}}{2I}\right]^{1/2}\int_0^\theta\left[\frac{1-k\sin^2\theta}{\tilde{n}(\theta_m)-\tilde{n}(\theta)}\right]^{1/2}d\theta\ . \tag{4.3}$$

We let

$$\sin\psi=\sin\theta/\sin\theta_m\ , \tag{4.4}$$

then the orientation of the director at a point $0\leq z\leq d/2$ is given by

$$z\left[\frac{2n_0 I}{ck_{33}}\right]^{1/2}=\sin\theta_m\left[\frac{1-\beta\sin^2\theta_m}{1-\alpha\sin^2\theta_m}\right]^{1/4}\int_{\psi_0}^{\psi}\left[\frac{1-k\sin^2\theta_m\sin^2\psi}{1-\left[\frac{1-\beta\sin^2\theta_m}{1-\alpha\sin^2\theta_m}\,\frac{1-\alpha\sin^2\theta_m\sin^2\psi}{1-\beta\sin^2\theta_m\sin^2\psi}\right]}\,\frac{1}{1-\sin^2\theta_m\sin^2\psi}\right]^{1/2}\cos\psi\,d\psi \tag{4.5}$$

with $\psi_0=0$.

The maximum deformation angle can be determined by evaluating Eq. (4.5) at $z=d/2$ and setting the upper limit in the integration to be $\psi=\pi/2$. The deformation angle at a point z can be expressed in terms of the maximum deformation angle using Eqs. (4.1) and (4.5). Therefore, Eqs. (4.1) and (4.5) describe completely the spatial orientation of the director as a function of the NLC parameters and the incident beam intensity.

To consider the threshold behavior, i.e., $I\gtrsim I_{\text{Fr}}$, we compute the integral in Eq. (4.5) up to and including terms $\sim\theta^2$, and obtain the following equation for the tilt angle:

$$\theta\simeq\theta_m\sin(\pi z/d)+0(\theta_m^3) \tag{4.6}$$

and

$$\theta_m^2\simeq(\sqrt{I/I_{\text{Fr}}}-1)/B$$
$$\simeq(I/I_{\text{Fr}}-1)/2B\ , \tag{4.7}$$

where the threshold intensity I_{Fr} and B are defined in Table I.[26] In the limit of a single elastic constant $k_{11}=k_{33}$ and small dielectric anisotropy $\epsilon_a\ll\epsilon_\perp\sim\epsilon_\parallel$, $B\approx\frac{1}{4}$ in both approaches and Eq. (4.7) reduces to those obtained by Khoo.[12,13] Notice that the threshold intensity predicted by approach I is smaller than that predicted by

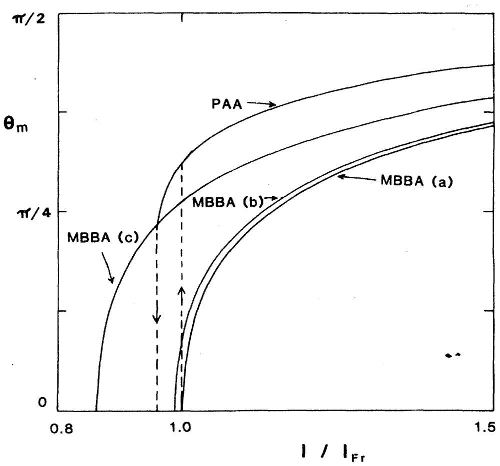

FIG. 1. Maximum deformation angle θ_m as a function of the reduced orientating beam intensity $I/I_{\rm Fr}$ for MBBA and PAA in approach I. For MBBA, we put $\lambda=6328$ Å, $n_0=1.544$, $n_e=1.758$, $k_{11}=6.95\times10^{-7}$ dyn, and $k_{33}=8.99\times10^{-7}$ dyn. Freedericksz transition is second order with $B=0.06$, $G=0.06$, and $I_{\rm Fr}=120.6$ W/cm^2 for a cell 250-μm thick. For PAA, we put $\lambda=4800$ Å, $n_0=1.595$, $n_e=1.995$, $k_{11}=9.26\times10^{-7}$ dyn, and $k_{33}=18.10\times10^{-7}$ dyn. Freedericksz transition is first order accompanied by hysteresis with $B=-0.08$, $G=0.07$, $I_{\rm Fr}=149.0$ W/cm^2, and $I'_{\rm Fr}=142.8$ W/cm^2 for a cell 250-μm thick. (a) For PAA and MBBA, the rigid anchoring condition at the two interfaces are assumed. Curve of θ_m vs $I/I_{\rm Fr}$ depends only on k and u, but is independent of the cell thickness for rigid anchoring. (b) and (c) For MBBA, a finite interfacial potential of $F_s=(11.7\sin^2\theta+7.8\sin^4\theta)$ merg/cm^2 is assumed at the two surfaces. (b) For MBBA, $d=250$ μm, (c) for MBBA, $d=50$ μm. Curve of θ_m vs $I/I_{\rm Fr}$ depends not only on k, u, and the anchoring, but also on the cell thickness for finite anchoring.

approach II. The exact maximum deformation angle as a function of the reduced intensity $I/I_{\rm Fr}$ is shown in Fig. 1.[27] Since

$$\frac{d}{2}\sqrt{2n_0I/ck_{33}}=\frac{\pi}{2}\sqrt{2I/uI_{\rm Fr}}\ ,$$

the curve for θ_m vs $I/I_{\rm Fr}$ depends only on k, u, and w, but is independent of the cell thickness.

B. First-order transition and hysteresis

From Eq. (4.7), we see that as $B>0$, the tilt angle remains zero for intensity below the threshold intensity and changes smoothly as $I\geq I_{\rm Fr}$. However, if $B<0$ which implies that $dI/d(\theta_m^2)<0$, then small distortions are not stable and the Freedericksz transition becomes a first-order transition accompanied by hysteresis: As the intensity increases from zero, the tilt angle remains 0 for $I<I_{\rm Fr}$ and the director switches at $I=I_{\rm Fr}$ from the undeformed state to a finite value of θ_m. But if the intensity decreases from above the threshold intensity, the director assumes a finite θ_m even at intensities below the rising threshold intensity. Upon reaching a lower critical threshold intensity $I'_{\rm Fr}$, the director switches again to the undeformed state. This falling threshold intensity can be determined by the condition

$$\frac{dI}{d(\theta_m^2)}=0\ . \tag{4.8}$$

The hysteresis associated with the first-order Freedericksz transition that we proposed is analogous to that proposed by Deuling and Helfrich[18,19] for a twisted nematic cell and a conducting nematic cell in a static electric field, but is different from that proposed by Zel'dovich. That is, with increasing intensity, the NLC director switches discontinuously at the rising threshold intensity from the undistorted homeotropic state to a state with a finite amount of distortion. With decreasing intensity the NLC director switches discontinuously back to the undistorted state at a lower falling threshold intensity as shown in Fig. 1. In our prediction the transitions associated with hysteresis are first order and the rising and falling transitions assume the same deformation for intensities greater than the rising threshold intensity, and the changes at both the rising and falling threshold intensities are discontinuous. But Zel'dovich proposed that the transitions associated with hysteresis are two second-order transitions and the rising and falling transitions assume different deformations even for intensities greater than the rising threshold intensity.

The criterion for the existence of the first-order optically induced Freedericksz transition at the threshold is given by $B<0$, i.e.,

$$\frac{k_{11}}{k_{33}}+\frac{9}{4}\frac{\epsilon_\perp}{\epsilon_\parallel}<\frac{9}{4} \tag{4.9}$$

for approach I; and

$$\frac{k_{11}}{k_{33}}+\frac{3}{4}\frac{\epsilon_\perp}{\epsilon_\parallel}+\frac{9}{4}\left[\frac{\epsilon_\perp}{\epsilon_\parallel}\right]^2<3 \tag{4.10}$$

for approach II. Figure 2 shows the nature of the transition at the threshold as a function of the NLC's parameters k_{11}/k_{33} and $\epsilon_\perp/\epsilon_\parallel$. The criterion shows that for a NLC with large dielectric and elastic anisotropies, the

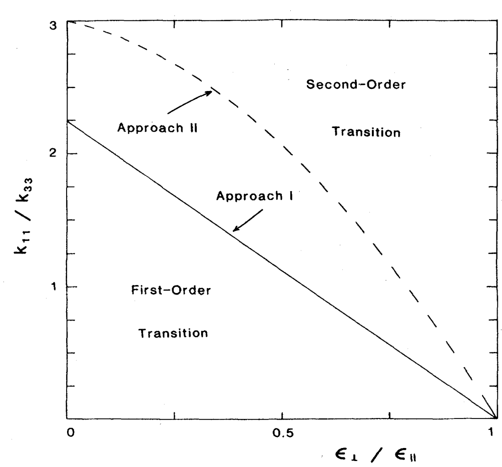

FIG. 2. Criterion for the existence of a first-order optically induced Freedericksz transition at the rising threshold intensity with rigid anchoring condition.

transition can be first order and that this is more favored in approach II than approach I. The nature of the transition at $B=0$ can be determined from the coefficient of θ^4 in the series expansion of Eq. (4.5), G, which will be determined later.

To determine the falling threshold intensity in the first-order Freedericksz transition, we compute the integral in Eq. (4.5) up to and including terms $\sim\theta^4$, and obtain the following equation for the maximum deformation angle:

$$\theta_m^2=\frac{-B+(B^2+4GC)^{1/2}}{2G}\,, \tag{4.11}$$

where $C=\sqrt{I/I_{\rm Fr}}-1$ and G is defined in Table I.[26] Therefore, for the first-order optically induced Freedericksz transition, the falling threshold intensity is given by

$$I'_{\rm Fr}=I_{\rm Fr}(1-B^2/4G)^2\,. \tag{4.12}$$

Since $G>0$ for known NLC's even when $B<0$, Eqs. (4.11) and (4.12) suffice for our present discussion of the first-order Freedericksz transition. In any case, if it happens that $G<0$, we need only to expand the solution up to the next higher-order term having a positive coefficient.

We can obtain a consistent formal thermodynamic description for the behavior of the transition by considering the expansion of the total free energy as a function of the maximum deformation angle. By expanding Eq. (2.8) up to and including terms θ_m^6, apart from a coefficient that renders it dimensionless, the total free energy has the form[28]

$$\mathcal{F}=-C\theta_m^2+B\theta_m^4/2+G\theta_m^6/3+O(\theta_m^8)\,. \tag{4.13}$$

The series (4.13) does not contain terms in odd powers of θ_m due to inversion symmetry for an ideal nematic and since the laws of electromagnetism conserve parity. The value of θ_m in thermal equilibrium is given by the minimum of $\mathcal{F}$ as a function of θ_m and satisfies

$$\frac{\partial\mathcal{F}}{\partial(\theta_m^2)}=-C+B\theta_m^2+G\theta_m^4+O(\theta_m^6)=0\,. \tag{4.14}$$

A negative value of C means that $\partial\mathcal{F}/\partial(\theta_m^2)>0$ for $\theta_m=0$, implying that the initial homeotropic state $\theta_m=0$ is stable, whereas a positive value of C means that the homeotropic state is unstable. The variation of the total free energy with the maximum deformation angle and intensity is shown in Fig. 3 for both $B>0$ and $B<0$.

We first consider the case where B is positive [Fig. 3(a)]. Since nothing new is added by the θ_m^6 term, the θ_m^6 term may then be neglected in $\mathcal{F}$. From Eq. (4.14) we have at equilibrium either $\theta_m=0$ or $\theta_m=\sqrt{C/B}$. For $I<I_{\rm Fr}$, the only real root is at $\theta_m=0$ since C is negative. For $I>I_{\rm Fr}$, $C>0$ and the minimum of $\mathcal{F}$ is at $\theta_m=\sqrt{C/B}$. Thus $I_{\rm Fr}$ is the threshold intensity and the transition is second order since the maximum deformation angle $\theta_m=[(\sqrt{I/I_{\rm Fr}}-1)/B]^{1/2}$ changes continuously from zero with increasing intensity above the threshold intensity.

We now consider the case where B is negative [Fig. 3(b)]. The positively valued G restrains $\mathcal{F}$ from going to minus infinity. The equilibrium condition Eq. (4.14) shows that θ_m is either zero or given by Eq. (4.11). Using

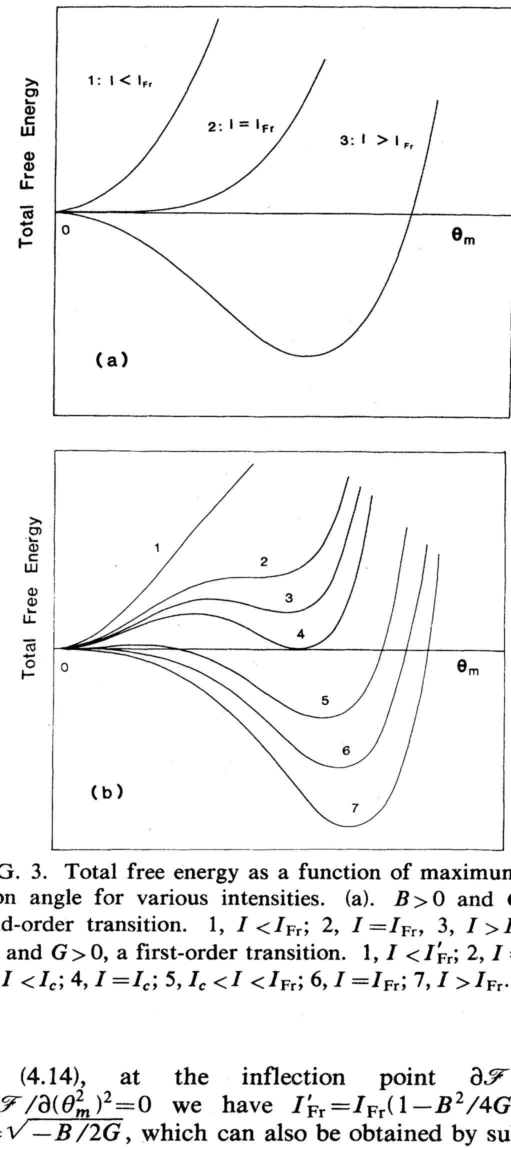

FIG. 3. Total free energy as a function of maximum deformation angle for various intensities. (a). $B>0$ and $G>0$, a second-order transition. 1, $I<I_{\rm Fr}$; 2, $I=I_{\rm Fr}$, 3, $I>I_{\rm Fr}$. (b). $B<0$ and $G>0$, a first-order transition. 1, $I<I'_{\rm Fr}$; 2, $I=I'_{\rm Fr}$; 3, $I'_{\rm Fr}<I<I_c$; 4, $I=I_c$; 5, $I_c<I<I_{\rm Fr}$; 6, $I=I_{\rm Fr}$; 7, $I>I_{\rm Fr}$.

Eq. (4.14), at the inflection point $\partial\mathcal{F}/\partial(\theta_m^2)=\partial^2\mathcal{F}/\partial(\theta_m^2)^2=0$ we have $I'_{\rm Fr}=I_{\rm Fr}(1-B^2/4G)^2$ and $\theta_m=\sqrt{-B/2G}$, which can also be obtained by substituting Eq. (4.11) into Eq. (4.10). For $I<I'_{\rm Fr}$, $\theta_m=0$ is the only real solution for the equilibrium state. From Fig. 3(b) we see that for the rising transition, the state $\theta_m\neq 0$ is unattainable at $I'_{\rm Fr}<I<I_{\rm Fr}$ because of the presence of the potential barrier. Thus the rising transition occurs at $I=I_{\rm Fr}$ and the state changes discontinuously from $\theta_m=0$ to $\theta_m=\sqrt{-B/G}$ and then changes continuously as I increases. For the falling transition, the state changes from $I\geq I_{\rm Fr}$ and $\theta_m\geq\sqrt{-B/G}$ so that the state will assume its local minimum described by Eq. (4.11) until the falling threshold $I'_{\rm Fr}$ which is a point of inflection of $\mathcal{F}$ is reached. Only after $I<I'_{\rm Fr}$ will the state change discontinuously from $\theta_m=\sqrt{-B/2G}$ to $\theta_m=0$. Therefore, the transition with negative B is first order accompanied by hysteresis with $I_{\rm Fr}$ as the rising threshold intensity and $I'_{\rm Fr}$ as the falling threshold intensity. It should be noticed that if the potential barrier is small, then thermal fluctuation may cause the rising transition to occur at $I_c<I<I_{\rm Fr}$ or the falling transition to disappear at $I'_{\rm Fr}<I<I_c$ where $I_c=I_{\rm Fr}(1-3B^2/16G)^2$ is the critical intensity at which

$\mathscr{F}=\partial\mathscr{F}/\partial(\theta_m^2)=0$ and $\theta_m=\sqrt{-3B/4G}>0$.[29] The thermodynamic description of the first-order transition agrees with our previous prediction using the solution of the maximum deformation angle [Eq. (4.5)] but disagrees with the Zel'dovich prediction as discussed earlier.

C. Experimental observation

The natures and the threshold intensities of the optically induced Freedericksz transition are listed in Table III for some known NLC's: E7, 5CB (also referred to as K15, PCB, and 4-cyano-4'pentylbiphenyl), 8CB (also referred to as K24, OCB, OCBP, and 4-*n*-octyl-4'cyanobiphenyl), MBBA [N-(*p*-methoxybenzylidene-*p*-butylaniline)], and PAA (*p*-azoxyanisole). The threshold intensities are calculated for a cell of 250-μm thickness. For those NLC's listed in the Table III, we found that $-0.08<B<0.11$ and $0.05<G<0.07$ for approach I, whereas $-0.28<B<-0.02$ and $0.06<G<0.10$ for approach II. The values of B and G differ quite significantly from those obtained from the singe elastic constant and small dielectric anisotropy approximation in which $B=\frac{1}{4}$ and $G=\frac{11}{192}$. The transition is always second order in the small dielectric and elastic anisotropies approximation.

Experimentally, the deformation can be observed by measuring the induced phase shift ϕ of the extraordinary ray component of a normally incident probe beam,

$$\phi=\frac{2\pi}{\lambda_p}\int_0^d\left[\frac{n_{\mathrm{op}}n_{\mathrm{ep}}}{(n_{\mathrm{ep}}^2\cos^2\theta+n_{\mathrm{op}}^2\sin^2\theta)^{1/2}}-n_{\mathrm{op}}\right]dz\ , \tag{4.15}$$

where λ_p is the wavelength of the probe beam and n_{op} and n_{ep} are, respectively, the ordinary and the extraordinary refractive indices of the nematic medium at the optical wavelength λ_p. By Eq. (4.5), the induced phase shift is given by

$$\phi=\frac{4\pi n_{\mathrm{op}}}{\lambda_p}\left[\frac{ck_{33}}{2n_0 I}\right]^{1/2}\sin\theta_m\left[\frac{1-\beta\sin^2\theta_m}{1-\alpha\sin^2\theta_m}\right]^{1/4}\times\int_{\psi_0}^{\pi/2}\left[\frac{1-k\sin^2\theta_m\sin^2\psi}{1-\left[\dfrac{1-\beta\sin^2\theta_m}{1-\alpha\sin^2\theta_m}\,\dfrac{1-\alpha\sin^2\theta_m\sin^2\psi}{1-\beta\sin^2\theta_m\sin^2\psi}\right]^{1/2}}\times\frac{1}{(1-\sin^2\theta_m\sin^2\psi)(1-u_p\sin^2\theta_m\sin^2\psi)}\right]^{1/2}\cos\psi\,d\psi-\frac{2\pi n_{\mathrm{op}}d}{\lambda_p}\ , \tag{4.16}$$

TABLE III. A comparison of the orders and the threshold intensities of the optically induced Freedericksz transition in approaches I and II.

NLC	Temp. (°C)	k_{11} (10^{-7} dyn)	k_{33} (10^{-7} dyn)	λ (Å)	n_0	n_e	Approach I Order	I_{Fr}	I'_{Fr}	Approach II Order	I_{Fr}	I'_{Fr}
E7[a]	30	10.1	16.20	5893	1.524	1.732	Second	223.1		First	288.1	265.4
5Cb[b]	26	7.20	8.52	5890	1.533	1.703	Second	133.5		First	166.2	162.4
5CB[c]	26	7.20	8.52	5890	1.540	1.719	Second	133.4		First	166.0	165.3
5CB[d]	26	5.20	7.17	5890	1.533	1.703	Second	116.8		First	144.2	141.9
8CB[e]	34	4	7	6328	1.516	1.665	Second	127.9		First	154.3	149.8
8CB[f]	34	4	7	6328	1.521	1.670	Second	127.9		First	154.3	149.8
MBBA[g]	22	6.95	8.99	6328	1.544	1.758	Second	120.6		First	156.4	150.0
PAA[h]	110	9.26	18.10	4800	1.595	1.995	First	149.0	142.8	First	233.1	150.3
PAA	120	7.80	13.60	4800	1.600	1.967	First	119.0	117.0	First	179.9	127.9
PAA	125	6.94	11.90	4800	1.605	1.949	First	108.9	107.9	First	160.8	119.6
PAA	130	5.67	9.05	4800	1.611	1.928	First	88.2	88.1	First	126.3	101.0

[a]Reference 30.
[b]Reference 31.
[c]Reference 32.
[d]Reference 33.
[e]Reference 34.
[f]Reference 35.
[g]Reference 36.
[h]Reference 37.

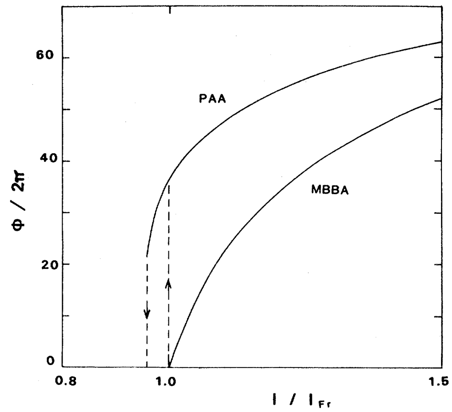

FIG. 4. Induced phase shift of the extraordinary ray component of a normally incident probe beam as a function of the reduced pump beam intensity I/I_{Fr} for MBBA and PAA by approach I for a cell 250-μm thick. Data for the elastic constants and refractive indices for the pump beam are the same as those used in Fig. 1. Rigid anchoring conditions at the two surfaces are assumed. For the probe beam, we put $\lambda_p = 5145$ Å, $n_{op} = 1.562$, $n_{ep} = 1.802$ for MBBA; $\lambda_p = 6438$ Å, $n_{op} = 1.550$, $n_{ep} = 1.839$ for PAA. MBBA shows a second-order transition, whereas PAA shows a first-order transition.

where $\psi_0 = 0$ and $u_p = 1-(n_{op}/n_{ep})^2$. The induced phase shift as a function of the orientating beam intensity is shown in Fig. 4.

Optically induced Freedericksz transitions have been observed in 5CB,[20,44] 8CB,[14,45] MBBA,[12,13,44,45] and a NLC comprised of 60% of mixture "*A*" and 40% of 4-cyano-phenyl-4-*n*-hepanyl-benzoate.[46] However, like the elastic constants and refractive indices, the experimental results by different researchers disagree greatly.[47] For example, for MBAA Csillag *et al.*[45] reported that the measured threshold was 45 ± 10 mW for a 150-μm-thick cell at a spot size of 65 μm and temperature 21.5 °C, i.e., $I_{Fr} = 1070\pm240$ W/cm^2. However, Khoo[12,13,44] reported that for a MBBA cell of 100-μm thickness, $I_{Fr}\sim 1000$ W/cm^2 at 20 °C. Since the threshold intensity is inversely proportional to the cell thickness (see Table I), we have, for a MBBA cell with thickness of 250 μm at room temperature, $I_{Fr}\sim 385$ W/cm^2 according to the Csillag *et al.* result, but $I_{Fr}\sim 160$ W/cm^2 according to the Khoo result. As a result it is difficult to compare the two approaches in the basis of the present experimental results. However, we see that in approach I the Freedericksz transitions for 5CB, 8CB, and MBBA are predicted to be second order, but in approach II the transitions are predicted to be first order, which conflicts with the experimental observations made by Csillag *et al.*,[14,45] Durbin *et al.*,[20] and Khoo.[12,13,44]

Since the first observation made by Freedericksz[52] in 1927, all the observed Freedericksz transitions are second order. In 1974 Deuling and Helfrich[18,19] predicted that for a NLC of strong conductive anisotropy and a dielectric anisotropy of opposite sign the dc electric-field-induced Freedericksz transitions in a twisted NLC cell and a conducting nematic cell can be first order. But the criterion for the existence of a first-order electric-field-induced Freedericksz transition is quite unattainable by the known NLC's. However, as shown in Table III, for PAA, the optically induced Freedericksz transition can be of first order; being of first order is more favored as the temperature decreases from the isotropic to nematic transition temperature, since k_{11}/k_{33} and $\epsilon_\perp/\epsilon_\parallel$ become larger at lower temperature. Clearly, the thermal fluctuation of the director would cause the hysteresis to disappear if the lower threshold intensity I'_{Fr} is too close to the threshold intensity I_{Fr}.[28] From Table III we see that for PAA with an incident orientating beam wavelength of 4800 Å at 110 °C $I'_{Fr}\sim 0.958 I_{Fr}$ and the hysteresis will be stable to thermal fluctuations. Therefore, it is convincing that a first-order Freedericksz transition could be observed experimentally for the first time in the near future.

V. FURTHER DISCUSSION

A. Dynamic response

We now consider the time dependence of the optically induced Freedericksz transition using approach I. For the total free energy density defined by Eq. (2.9), we include a dissipative term $\eta(\partial\hat{n}/\partial t)^2/2$ which will contribute a viscous torque opposing any rapid change of the director, where η is the viscosity of the NLC. The dynamic behavior is described by the resulting Euler equation

$$(1-k\sin^2\theta)\frac{\partial^2\theta}{\partial z^2} - k\sin\theta\cos\theta\left[\frac{\partial\theta}{\partial z}\right]^2 + \frac{In_0u}{ck_{33}}\frac{\sin\theta\cos\theta}{(1-u\sin^2\theta)^{3/2}} = \frac{\eta}{k_{33}}\frac{\partial\theta}{\partial t}\ . \quad (5.1)$$

In the following, we consider second-order transition and neglecting the role of backflow.[53] We look for an approximate solution of the form

$$\sin\theta(z,t) = \sin\theta_m(t)\sin\psi(z,t)\ , \quad (5.2)$$

where $\theta_m(t)$ is the amplitude of the distortion. This form of solution has been considered by Deuling[19] in the study of the dynamics of a dc field-induced Freedericksz transition. By assuming a weak time dependence on $\psi(z,t)$ which contains the spatial distribution of the deformation angle, we have $\partial\theta/\partial t = (\dot{q}/q)\tan\theta$ and the solution of Eq. (5.1) is

$$z = \int_0^\psi \left[\frac{2In_0}{ck_{33}}\left[\frac{1}{(1-uq^2)^{1/2}} - \frac{1}{(1-uq^2\sin^2\psi)^{1/2}}\right] - \frac{\eta\dot{q}}{k_{33}q}\ln\frac{1-q^2\sin^2\psi}{1-q^2}\right]^{-1/2} \times\left[\frac{1-kq^2\sin^2\psi}{1-q^2\sin^2\psi}\right]^{1/2}\cos\psi\, d\psi\ , \quad (5.3)$$

where $q(t) = \sin\theta(t)$ and $\dot{q} = \partial q/\partial t$. In obtaining Eq. (5.3), we have made use of the condition that at all times $\partial\theta/\partial z = 0$ at $z = d/2$. q can be determined by evaluating

Eq. (5.3) at $z=d/2$. In the static case, $\dot{q}=0$ and Eq. (5.3) reduces to the exact solution that is given by Eq. (4.5). The description of the dynamic behavior is by no means complete since, in general, it is still difficult to determine q from Eq. (5.3).

Let us first consider the case where the optical field is abruptly increased from zero to $I \gtrsim I_{\mathrm{Fr}}$. Since even in the final state, described by Eq. (4.6), θ_m will still be small, we may expand the dynamic solution Eq. (5.3) in powers of q and obtain that

$$\dot{q}=q(a-bq^2)+O(q^5)\ , \tag{5.4}$$

where

$$a=k_{33}(\pi/d)^2(I/I_{\mathrm{Fr}}-1)/\eta=n_0u(I-I_{\mathrm{Fr}})/c\eta\ ,$$

$b=2Bk_{33}(\pi/d)^2/\eta$, and $B=(1-k-9u/4)/4$. Equation (5.4) is of the same form for the dc case derived by Pieranski *et al.*[54] Since the constant term is missing, we need a small fluctuation q_i at $t=0$ to get the distortion started. Equation (5.4) can be solved to give

$$\theta(z,t)=\left[\frac{1}{1+q_r^2e^{-2at}}\right]^{1/2}q_f\sin(\pi z/d)\ , \tag{5.5}$$

where $q_f^2=a/b=(I/I_{\mathrm{Fr}}-1)/2B$ and $q_r^2=q_f^2/q_i^2-1$. At $t\approx 0$, Eq. (5.5) describes the exponential growth of a small fluctuation with a time constant $1/a$, which agrees with that obtained by Zel'dovich[15] and by Durbin[20,56]:

$$q(t\gtrsim 0)\approx q_i\exp(at)\ . \tag{5.6}$$

For $t\gg 1$, the deformation amplitude reaches its final value q_f exponentially with a smaller time constant $1/2a$:

$$q(t\gg 1)\approx(1-\tfrac{1}{2}q_r^2e^{-2at})q_f\ . \tag{5.7}$$

Typically, the time constant $1/a$ for a cell 250-μm thick is about $70/(I/I_{\mathrm{Fr}}-1)$ sec.

We now consider the case where the optical field is switched off from $I>I_{\mathrm{Fr}}$ at time $t=0$. The amplitude of the deformation angle can be determined from Eq. (5.3) with $I=0$. For $I\gtrsim I_{\mathrm{Fr}}$, by denoting the initial deformation state by $\theta(z,t=0)=q_i\sin\psi(z)$, q can again be described by Eq. (5.4) with $a=-k_{33}(\pi/d)^2/\eta=-g$, $b=ah$, and $h=\frac{1}{4}+k/2$. The amplitude of the deformation then decreases exponentially:

$$q(t)=\left[\frac{1}{1-hq_i^2(1-e^{-2gt})}\right]^{1/2}q_ie^{-gt}\ . \tag{5.8}$$

The initial and the long-time responses to the laser switch-off are both exponential with a relaxation time $1/g$ which is the same as that obtained by Durbin[20]:

$$q(t)\approx Cq_i\exp(-gt)\ , \tag{5.9}$$

where $C\approx 1$ for $t\approx 0$ and $C\approx 1/(1-hq_i^2)^{1/2}$ for $t\gg 1$. Typically, the time constant $1/g$ for a cell 250-μm thick is about 70 sec.

The transient behavior of the molecular reorientation can be used to determine the viscosity, the threshold intensity, and the bend elastic constant as discussed above. Experimental results[20,57] show that the initial responses to the laser switch-on and the long-time response to the laser switch-off both have exponential time dependence.

B. Interfacial interaction

All the above calculations always assume that the orientations of the NLC's at the two interfaces can never be changed. Such an assumption would require a rigid anchoring between the NLC's and the interfaces. However, some experimental findings show that the anchoring strength between the NLC's and the interfaces is finite and typically of the order of $1-10^{-4}$ erg/cm^2.[58] Khoo found that in order to create molecular reorientation in 5CB, it was necessary to soften the surface anchoring condition.[44] We now discuss the effects of the surface interaction on the optically induced reorientation using approach I.

Without loss of generality, we assume that the anchoring potential at the two surfaces is the same and can be expressed as

$$F_s(\theta)=\tfrac{1}{2}\sum_{n=1}^{\infty}C_{2n}P_{2n}(\cos\theta)$$

$$=\tfrac{1}{2}\sum_{n=1}^{\infty}A_{2n}\sin^{2n}\theta\ , \tag{5.10}$$

where P_{2n} is the Legendre polynomial of order $2n$ and A_{2n} represents the anchoring strength.[59,60] The total free energy density can be written as

$$F=\tfrac{1}{2}k_{33}(1-k\sin^2\theta)\left[\frac{d\theta}{dz}\right]^2-\frac{I}{c}n_p$$

$$+\tfrac{1}{2}[\delta(z)+\delta(d-z)]\sum_{n=1}^{\infty}A_{2n}\sin^{2n}\theta\ . \tag{5.11}$$

The variation of the total free energy leads to three equations, the solutions of which describe the equilibrium orientation of NLC throughout the cell. A bulk equation is given by Eq. (2.9) with $\alpha=0$ and $\beta=u$, and two boundary equations for the two interfaces are

$$k_{33}(1-k\sin^2\theta_0)\frac{d\theta}{dz}=\pm\cos\theta_0\sum_{n=1}^{\infty}nA_{2n}\sin^{2n-1}\theta_0\ , \tag{5.12}$$

where $\theta_0=\theta(z=0)=\theta(z=d)$ and the upper sign for $z=0$ and lower sign for $z=d$.

With the boundary condition that $\theta(z=0)=\theta_0$ and $\theta(z)=\theta(d-z)$, the orientation of the director at $0\le z\le d/2$ is given by Eq. (4.5) with the lower limit in the integration $\psi_0=\sin^{-1}(\sin\theta_0/\sin\theta_m)$. Evaluating Eq. (4.2) at $z=0$ to eliminate $d\theta/z$, we obtain the following equation for determining the surface tilt angle:

$$\cos\theta_0 \sum_{n=1}^{\infty} nA_{2n}\sin^{2n-1}\theta_0=\{(2n_0 Ik_{33}/c)(1-k\sin^2\theta_0)\times[(1-u\sin^2\theta_0)^{1/2}-(1-u\sin^2\theta_m)^{1/2}]/[(1-u\sin^2\theta_0)(1-u\sin^2\theta_m)]^{1/2}\}^{1/2} . \quad (5.13)$$

The induced phase shift of the extraordinary ray component of a normally incident probe beam is given by Eq. (4.16) with $\psi_0=\sin^{-1}(\sin\theta_0/\sin\theta_m)$.

Evidently, the threshold intensity will be lower as the anchoring strength decreases. At higher intensities the effects of finite anchoring will be larger, and there exists a saturation intensity I_s above which all NLC's will orient parallel to the surfaces (the x axis). By computing the solution for the tilt angle up to and including terms $\sim\theta^2$, we find that the new rising threshold intensity $\tilde{I}_{\mathrm{Fr}}$ for finite anchoring can be determined from

$$\cot\left[\frac{\pi}{2}\left[\frac{\tilde{I}_{\mathrm{Fr}}}{I_{\mathrm{Fr}}}\right]^{1/2}\right]=\frac{\pi k_{33}}{dA_2}\left[\frac{\tilde{I}_{\mathrm{Fr}}}{I_{\mathrm{Fr}}}\right] , \quad (5.14)$$

and that the tilt angle just above the threshold intensity in the second-order transition is given by

$$\theta(z)\simeq\theta_m \sin\left\{\frac{\pi}{d}\left[\frac{\tilde{I}_{\mathrm{Fr}}}{I_{\mathrm{Fr}}}\right]^{1/2} z+\frac{\pi}{2}\left[1-\left[\frac{\tilde{I}_{\mathrm{Fr}}}{I_{\mathrm{Fr}}}\right]^{1/2}\right]\right\}+O(\theta_m^3) , \quad (5.15)$$

where

$$\theta_m^2=[(I/I_{\mathrm{Fr}})^{1/2}-(\tilde{I}_{\mathrm{Fr}}/I_{\mathrm{Fr}})^{1/2}]/\tilde{B} ,$$

$$\tilde{B}=\{(1-k-9u/4)\pi(\tilde{I}_{\mathrm{Fr}}/I_{\mathrm{Fr}})^{1/2}+(1-k-3u/4)\sin[\pi(\tilde{I}_{\mathrm{Fr}}/I_{\mathrm{Fr}})^{1/2}]\}/4\pi ,$$

and $I_{\mathrm{Fr}}=ck_{33}(\epsilon_{\parallel}/n_0\epsilon_a)(\pi/d)^2$ is the threshold intensity for rigid anchoring. Consequently, the criterion for the existence of the first-order transition at the rising threshold is given by

$$(1-k-9u/4)\pi(\tilde{I}_{\mathrm{Fr}}/I_{\mathrm{Fr}})^{1/2}+(1-k-3u/4)\sin[\pi(\tilde{I}_{\mathrm{Fr}}/I_{\mathrm{Fr}})^{1/2}]<0 . \quad (5.16)$$

The less than sign ($<$) in Eq. (5.16) is replaced by an equal sign for the case of zero anchoring. As the anchoring strength decreases from infinity to zero, the criterion Eq. (4.9) relaxes completely so that a first-order transition is always possible regardless of the other physical parameters of the NLC, and the threshold intensity changes from finite value to zero.

The function $I(\theta_m)$ which gives the maintenance intensity I at a given θ_m is a single-valued function of θ_m but can assume the same value at different θ_m. Consequently, the function $\theta_m(I)$ which gives the maximum deformation angle at a given intensity is a multivalued function of I. The saturation intensity as well as the nature of the transition at a given intensity can be determined by examining the value of $I(\theta_m)$ as a function of the maximum deformation angle. Clearly the saturation intensity I_s is the maximum value of $I(\theta_m)$ for $0\le\theta_m\le\pi/2$. If $\theta_s\equiv\theta_m(I=I_s)$ is less than $\pi/2$, then at $I=I_s$, the state changes discontinuously from $\theta_m=\theta_s$ to the parallel state [$\theta(z)=\pi/2$ for all z] through a first-order transition. Once the parallel state is attained, the parallel-state-maintenance intensity I_m which is the intensity above which the state remains in the parallel state is given by

$$\coth\left[\frac{\pi}{2}\left[\frac{n_e^3 k_{33} I_m}{n_0^3 \kappa_{11} I_{\mathrm{Fr}}}\right]^{1/2}\right]=\frac{\pi k_{11}}{dM}\left[\frac{n_e^3 k_{33} I_m}{n_0^3 k_{11} I_{\mathrm{Fr}}}\right]^{1/2} , \quad (5.17)$$

where $M=\sum_{n=1}^{\infty} nA_{2n}$. We see that $\tilde{I}_{\mathrm{Fr}}/I_{\mathrm{Fr}}$ depends only on $k_{33}/A_2 d$ as in the dc case.[58–62] However, I_m/I_{Fr} depends not only on k_{11}/k_{33} and M, but also on $n_e^3 k_{33}/n_0^3 k_{11}$. This dependence differs from that in the magnetic field where one can show that the ratio of the parallel-state-maintenance field to the threshold field depends only on k_{11}/k_{33} and M. If the function $I(\theta_m)$ has extremal values at some intermediate angles between 0 and $\pi/2$, then first-order transitions accompanied by hysteresis loops could occur at those angles. *The criteria for the existence of a first-order transition is given as follows.*

(1) If $dI/d(\theta_m^2)<0$ at $\theta_m=0$, i.e., the criterion (5.6) is satisfied, then at the rising threshold intensity $\tilde{I}_{\mathrm{Fr}}$ the rising transition is first order and the state will assume a deformation with $\theta_m=\theta_m(\tilde{I}_{\mathrm{Fr}})>0$. The falling threshold intensity is the minimum value of $I(\theta_m)$ in the range $0\le\theta_m\le\theta_m(I=I_f)$ where the falling transition occurs at $I=I_f$.

(2) If $dI/d\theta_m\le0$ at $0<\theta_i\le\theta_m\le\theta_f\le\pi/2$, then the transition is first order for states with θ_m in the range $\theta_i\le\theta_m\le\theta_f$. In the rising transition, the transition is first order at $I=I_i\equiv I(\theta_i)$ and the state changes from $\theta_m=\theta_i$ to $\theta_m=\theta_m(I=I_i)>\theta_f$. In the falling transition, the first-order transition occurs at $I=I_f\equiv I(\theta_f)$ and the state changes from $\theta_m=\theta_f$ to $\theta_m=\theta_m(I=I_f)<\theta_f$.

(3) If $I_m<I_s$, then the rising transition at $I=I_s$ is first order and the state changes from $\theta_m=\theta_m(I=I_s)<\pi/2$ to the parallel state. Once the parallel state is attained, the parallel state remains for $I>I_m$, and as I drops below I_m, the state changes to $\theta_m=\theta_m(I=I_m)<\pi/2$ through a first-order transition. If $I_m<\tilde{I}_{\mathrm{Fr}}$, then once the parallel state is attained I_m becomes the falling threshold intensity and the state changes back to the homeotropic state as $I<I_m$ through a strongly first-order transition.

We notice that even in a simple case like $F_s=A_2\sin^2\theta+A_4\sin^4\theta$, the rising and falling transitions can be first order at some intermediate angle. We consider a 20-μm thick MBBA cell as an example with the material constants that were used in Fig. 1 and $A_2=-3.5A_4=2$ merg/cm^2. We obtain $\tilde{I}_{\mathrm{Fr}}=0.50I_{\mathrm{Fr}}$ and

$I_s = 0.59 I_{Fr}$ at which the state changes discontinuously from $\theta_m = 70°$ to $\theta_m = \pi/2$. That is, at $I = I_s$, the state changes to the parallel state through a first-order transition resulting from lowering the electromagnetic energy and the elastic energy into their minimum values. Moreover, Eq. (5.17) shows that the parallel-state-maintenance intensity $I_m = 0.41 I_{Fr}$ which is less than the rising threshold intensity. Consequently, I_m is also the falling threshold intensity if the parallel state is attained and an intensity I with $I_m < I < \tilde{I}_{Fr} < I_s$ is enough to maintain the parallel state. As $I < I_m$, the state changes discontinuously from the parallel state back to the homeotropic state through a strongly first-order transition.

Recently, Yang and Rosenblatt reported an interfacial potential of $F_s = (11.7 \sin^2\theta + 7.8 \sin^4\theta)$ merg/cm^2 for a MBBA homeotropic cell.[63] The effects of this interfacial potential on the maximum deformation angle are shown in Fig. 1 for two different thicknesses. We see that the effects of the interfacial interaction become more important as the cell thickness decreases. Using the dependence of θ_m on the $I/\tilde{I}_{Fr}$ and the cell thickness, there are three simple experimental methods to manifest the effects of finite anchoring on the transition.

(1) By measuring the threshold intensity $\tilde{I}_{Fr}$ for the same NLC with different cell thickness: Rigid anchoring conditions would show that the threshold intensity is inversely proportional to the square of the cell thickness (Table I).

(2) By measuring the maximum deformation angle versus $I/\tilde{I}_{Fr}$ for different thickness: Rigid anchoring conditions predict that θ_m depends only on $I/\tilde{I}_{Fr}$ and is independent on the cell thickness.

(3) By measuring the threshold intensity and θ_m versus the intensity for the same NLC and thickness but with different homeotropic surface treatment: The results should be independent of the surface treatment if the anchorings are rigid.

These three rigid anchoring predictions will not be true for finite anchoring conditions.

At present, the surface interaction remains one of the least understood areas of the liquid crystal's physics. Clearly the as yet unexplored and unobserved first-order transition induced by surface interactions could have important practical applications.

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$$\theta_{\max} = \left[2 \frac{E_{\text{op}}^2 - E_F^2}{E_F^2} \right]^{1/2} ;$$

and (d) Eq. (9) should read

$$\delta\epsilon = \frac{\epsilon_\perp \Delta\epsilon}{\epsilon_{\parallel}} \left[\frac{E_{\text{op}}^2}{E_F^2} - 1 \right] \sin^2 \left[\frac{\pi Z}{d} \right] .$$

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