# Optically induced molecular reorientation in a smectic-C liquid crystal

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The Euler equations for describing the director in the optically induced molecular reorientation of a smectic-C liquid crystal in an external magnetic field are presented. The optical alignment effect is shown to be localized and not to produce point defects. Analytic expressions are given explicitly in the small-distortion regime, which, generally, have the form of zeroth-order Bessel functions. Threshold behavior exists if the polarization of the optical beam is normal to the magnetic field. For a magnetic field of the order of 1 kG, the threshold power varies from 3 to 120 mW for a typical smectic-C sample with a laser spot size of about 1 to 100  $\mu$ m, but is independent of the thickness of the sample. For polarizations oblique to the magnetic field, there is no threshold and the amplitude of rotation of the director depends on spot size, laser power, and the angle between laser polarization and external magnetic field. The transient response of molecular reorientation to the laser switch on is shown to have exponential time dependence for a normally polarized incident beam with an incident intensity greater than the threshold intensity. The response time of such a reorientation is of the order of milliseconds, depending on incident laser intensity. We propose that, experimentally, the molecular reorientation can be quantitatively measured by the reflectivity or transmissivity of a normally incident probe beam. Optical reflectivity and transmissivity from a typical smectic-C film are also calculated.

## I. INTRODUCTION

Recently the nonlinear optics of liquid crystals has received a great deal of attention.<sup>1</sup> Shelton and Shen initiated the study of the normal and umklapp optical thirdharmonic generation in cholesteric liquid crystals (CLC).<sup>1-3</sup> The orientational optical nonlinearity of CLC was recently considered by Tabiryan and Zel'dovich<sup>4</sup> in 1981 and by Winful<sup>5</sup> in 1982. Tabiryan and Zel'dovich showed that when a light wave propagates along the helical axis, self-focusing should not occur for circularly polarized light, but for linearly polarized light the nonlinear dielectric constant  $\epsilon_2$  is about  $8 \times 10^{-8}$  cm<sup>3</sup>/erg. The elliptic-function solutions in the Bragg regime have been obtained by Winful and the results show that optically induced changes in the pitch of the cholesteric helix lead to a bistable reflection even in the absence of external reflectors. In the nematic-liquid-crystal (NLC) phase, the optically induced Freedericksz transition has received even more attention. The effect of the optically induced molecular reorientation in the NLC was explained qualitatively by Zolot'ko et al. in 1980.<sup>6</sup> A quantitative theory was later constructed by Zel'dovich, Tabiryan, and Chilingaryan using the geometrical-optics approximation.<sup>7</sup> In 1981, Durbin, Arakelian, and Shen reported the first observation of the optically induced Freedericksz transition in nematic 5CB (4-cyano-4'-pentylbiphenyl) and showed that the results were in quantitative agreement with their theoretical predictions using the infinite-plane-wave approximation.<sup>8</sup> In 1981 Khoo also presented an approximate solution and made a quantitative experimental verification of the associated nonlinear optical processes.9 Recently the exact solution describing the orientation of the NLC molecule was obtained by Ong.<sup>10</sup> These studies show that for certain NLC's with large dielectric and elastic anisotropies, the transition can be first order accompanied by hysteresis, and that the optical nonlinearity of NLC's is larger by eight to ten orders of magnitudes than that of carbon disulfide (CS<sub>2</sub>). The term "gigantic optical nonlinearity" (GON) has been coined by Zel'dovich for such large nonlinearities.

Contrary to cholesterics and nematics, not much work has been done regarding the effects of external fields on smectics. The possibility of a Freedericksz transition in a smectic liquid crystal by an external dc magnetic field has been considered by Helfrich,<sup>11</sup> Rapini,<sup>12</sup> Hurault,<sup>13</sup> and Meirovitch *et al.*<sup>14,15</sup> The results show that all transitions requiring a distortion of the layers are probably not observable because the distortion is a very weak function of the field. Such a transition has therefore been called a "ghost" by Rapini and has not been observed experimentally. However, those magnetic-field-induced transitions which involve the rotation of the director about the normal to the smectic-C-liquid-crystal (SmC) layers should be observable, as discussed by Rapini<sup>12</sup> and by Meirovitch et al.<sup>14</sup> Indeed, magnetic fields have been used to align the azimuthal angle of the SmC experimentally.<sup>14,15</sup> The effects of weak anchoring between the smectics and the surfaces on the dc-field-induced Freedericksz transition have also been studied by Meirovitch et al.<sup>14</sup>

In contrast, using a linearly polarized light source with the electric field directed in the plane of the layer at an angle to the initial orientation, it is possible to reorient the azimuthal component of the director of the SmC molecules. This transition involves only a rotation of the director about the normal to the layers and does not in-

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volve any distortion in the layer. Indeed, the optically induced molecular reorientation in the SmC was recently directly observed by Lippel and Young using a linearly polarized laser beam incident on a freely suspended film of a SmC.<sup>17</sup> They reported that large reorientations have been observed with an incident optical power of less than 50 mW and have presented a simple theory explaining the reorientation effect. The theoretical study of the orientational optical nonlinearity of SmC induced by an optical field in the presence of an external orienting dc magnetic field has been considered by Tabiryan and Zel'dovich who showed that the effective nonlinear dielectric constant  $\epsilon_2$ is  $\sim 0.2$  cm<sup>3</sup>/erg for the self-focusing light and  $\chi_{\rm eff} \sim 0.6 \times 10^{-6}$  cm<sup>3</sup>/erg for the four-wave interaction.<sup>18</sup> These nonlinear constants are, respectively, nine and five orders larger than those of  $CS_2$ . However, the solution to the spatial orientation of the director of the SmC has not been found.

It is the purpose of this paper to present the Euler equations for describing the director in the optically induced molecular reorientation in a SmC sample. The sample is assumed initially oriented by a homogeneous dc magnetic field in the SmC layer so that without the optical field, the azimuthal angle of the SmC is well aligned in the magnetic field direction. A polarized light beam is then normally incident on the sample. If the polarization is at an angle to the magnetic field, the azimuthal angle of the director will vary under the action of the optical field. We discuss the general properties of the solution for the director and give analytic expressions explicitly in the small-distortion linearized regime. The results show that the optical alignment effect is localized and does not produce point defects. Generally, the solutions in the small-distortion regime have the form of zeroth-order Bessel functions. Using the continuity condition at the boundary imposed by the spot size of the optical field, the amplitudes of the deformations can be determined. In regions far away from the optical field, the azimuthal angle approaches its asymptotic orientation exponentially  $\sim e^{-qr}/\sqrt{qr}$  where q is the inverse magnetic field coherence length. If the polarization is normal to the orienting magnetic field, there exists a characteristic threshold intensity below which no molecular reorientation can be induced. The threshold power depends on the applied magnetic field and the laser spot size, but is independent of the sample thickness. For a magnetic field of the order of 1 kG, the threshold power varies from 3 to 120 mW for a typical SmC sample with a laser spot size of about 1 to 100  $\mu$ m in radius. We also discuss the dynamics of the transition. The transient response of molecular reorientation to the laser switch on is shown to have exponential time dependence for a normally polarized incident beam with an incident intensity greater than the threshold intensity. We propose that, experimentally, the molecular reorientation can be quantitatively measured by the reflectivity or transmissivity of a normally incident probe beam. The reflected and transmitted power of the probe beam covering a known area are derived.

In the following sections, we first discuss the free energy density and the Euler equations (Sec. II). A section on the solution describing the orientation of the director in the small-distortion regime follows (Sec. III), including a general discussion of the solutions at the origin and at infinity. Finally, proposed methods to observe the optical nonlinearity effects experimentally are discussed in Sec. IV.

## II. FREE ENERGY DENSITY AND EULER EQUATIONS

The elastic theory of SmC has been considered by the Orsay group<sup>19</sup> and by Rapini.<sup>12</sup> Both approaches use the Oseen description of smectic A, neglecting all changes in internal parameters such as density, interlayer distance, and tilt angle. The Orsay group used the Lagrangian description for the elastic strains with a vector  $\vec{\Omega}(\vec{r})$  describing the local rotation of the director. In order to be consistent with the elastic theory for NLC's, we shall use the Eulerian description developed by Rapini based on the director.

We introduce two unit vectors  $\hat{k}$  and  $\hat{n}$  to describe the SmC structure.  $\hat{k}$  is normal to the layers and  $\hat{n}$  is along the "long axis" of the molecules as shown in Fig. 1. In



FIG. 1. Assumed structure of a smectic-C liquid crystal. (a) A homogeneous dc magnetic field  $H_0$  is directed along the y direction so that the unperturbed state of the SmC sample has  $\hat{k}$ along the z axis and  $\hat{n} = \hat{n}_0$  in the y-z plane making an angle  $\theta_0$ with the z axis. (b) An orienting optical beam is linearly polarized in the x-y plane with the electric field directed at an angle  $\phi_0$  to the y axis and is normally incident on the SmC sample. In equilibrium, the director  $\hat{n}$  of the SmC is oriented at an azimulthal angle  $\phi$  with the y axis with the polar angle  $\theta_0$  being fixed.

the following discussion, an orienting uniform dc magnetic field  $\vec{H}_0 = (0, H_0, 0)$  directed along the y direction is applied to the sample so that in the absence of an optical field, the preferred azimuthal direction is along the y axis. The unperturbed state then has  $\hat{k}$  along the z axis, and  $\hat{n} = \hat{n}_0$  in the z-y plane making an angle  $\theta_0$  with  $\hat{k}$ . It is convenient to introduce a vector  $\hat{n}_{\perp} \equiv \hat{n} - (\hat{n} \cdot \hat{k})\hat{k}$  which lies in the plane normal to  $\hat{k}$  and which satisfies  $\hat{n}_{\perp}^2 = \text{const.}$ 

When a light beam linearly polarized in the x-y plane with the elastic field directed at an angle  $\phi_0$  to the y axis is normally incident on the SmC layer, there is an additional term in the total free energy density of the system due to the optical field. For the discussion of the optical-fieldinduced Freedericksz transition, the total free energy density  $F(\text{erg/cm}^3)$  can be written as

$$F = \frac{1}{2}\alpha_{11}(\operatorname{div}\hat{n}_{\perp})^{2} + \frac{1}{2}\alpha_{22}(\hat{n}_{\perp}\cdot\operatorname{curl}\hat{n}_{\perp})^{2} + \frac{1}{2}\alpha_{33}(\hat{k}\cdot\operatorname{curl}\hat{n}_{\perp})^{2} - \alpha_{23}'(\hat{n}_{\perp}\cdot\operatorname{curl}\hat{n}_{\perp})(\hat{k}\cdot\operatorname{curl}\hat{n}_{\perp}) - \frac{1}{8\pi}(\vec{E}\cdot\vec{D} + \vec{B}\cdot\vec{H}) - \frac{1}{2}\chi_{a}(\hat{n}\cdot\vec{H}_{0})^{2},$$
(2.1)

(2.3)

where  $\chi_a$  is the magnetic susceptibility anisotropy,  $\alpha'_{ij} = \alpha_{ij} \sin^2 \theta_0$ , and  $\alpha_{ij}$  are the smectic curvature elastic moduli.<sup>12</sup> The smectic curvature elastic moduli have the same dimensions and the same magnitudes as the Frank elastic constants for NLC.<sup>16,20</sup> The first three terms of Eq. (2.1) are analogous to splay, twist, and bend in a NLC. These types of deformations were first discussed by Saupe.<sup>21</sup> The term  $-(\vec{E}\cdot\vec{D}+\vec{B}\cdot\vec{H})/8\pi$  is the electromagnetic energy density of the light beam and the last term is the dc magnetic energy density. The Euler equations for the director  $\hat{n}(\vec{r})$  have the form

$$\frac{\delta F}{\delta n_i} - \frac{\partial}{\partial x_k} \frac{\delta F}{\delta(\partial n_i / \partial x_k)} = \lambda n_i , \qquad (2.2)$$

where  $\lambda(\vec{r})$  is an undetermined Lagrange multiplier which ensures that the condition  $|\hat{n}| = 1$  is satisfied. The orientation of the SmC sample is then completely described by the solution to Eq. (2.2) subject to initial and boundary conditions.

From the results of Rapini<sup>12</sup> on the dc-field-induced Freedericksz transition, which show that one needs an extremely high field to vary the polar angle of the SmC sample, it is reasonable for us to assume that the polar angle  $\theta_0$  remains fixed. We denote the angle between the director and the y axis by  $\phi$  [Fig. 1(b)], then the director is given by

$$\hat{n} = (\sin\theta_0 \sin\phi, \sin\theta_0 \cos\phi, \cos\theta_0) ,$$

$$\hat{n}_1 = (\sin\theta_0 \sin\phi, \sin\theta_0 \cos\phi, 0)$$
.

and

The elastic deformation energy density  $F_d$  is then given by

$$F_{d} = \frac{1}{2} \sin^{2}\theta_{0}(\alpha_{11}\cos^{2}\phi + \alpha_{33}\sin^{2}\phi) \left[\frac{\partial\phi}{\partial x}\right]^{2}$$
$$+ \frac{1}{2}\sin^{2}\theta_{0}(\alpha_{11}\sin^{2}\phi + \alpha_{33}\cos^{2}\phi) \left[\frac{\partial\phi}{\partial y}\right]^{2}$$
$$+ \frac{1}{2}\sin^{2}\theta_{0}\sin(2\phi)(\alpha_{33} - \alpha_{11})\frac{\partial\phi}{\partial x}\frac{\partial\phi}{\partial y} . \qquad (2.4)$$

Experimentally, the exact values of the elastic constants of SmC have not yet been measured but are of the same order of magnitudes as NLC's,<sup>16,20</sup> and since the equations that arise from unequal elastic constants are very complicated,<sup>22</sup> we shall use a single-elastic-constant approximation,  $\alpha_{11}=\alpha_{33}=\alpha$ , to simplify the discussion. Then the deformation energy is reduced to the form

$$F_d = \frac{1}{2}\alpha \sin^2 \theta_0 (\operatorname{grad} \phi)^2 . \tag{2.5}$$

For the optical field, the electric and magnetic energy densities are equal<sup>23</sup> so that the total electromagnetic energy density can be written as  $F_{\text{opt}} = -\vec{E}\cdot\vec{D}/4\pi$ . It has been shown that SmC is nearly uniaxial with the optical axis along  $\hat{n}$ .<sup>16,24</sup> Using the uniaxial dielectric tensor<sup>16</sup>

$$\epsilon_{ii} = \epsilon_1 \delta_{ii} + \epsilon_a n_i n_i \tag{2.6}$$

for the SmC,  $\hat{\epsilon}$  can be written as

$$\widehat{\boldsymbol{\epsilon}} = \begin{cases} \boldsymbol{\epsilon}_{\perp} + \boldsymbol{\epsilon}_{a} \sin^{2}\theta_{0} \sin^{2}\phi \quad \boldsymbol{\epsilon}_{a} \sin^{2}\theta_{0} \sin\phi \cos\phi \quad \boldsymbol{\epsilon}_{a} \sin\theta_{0} \cos\theta_{0} \sin\phi \\ \boldsymbol{\epsilon}_{a} \sin^{2}\theta_{0} \sin\phi \cos\phi \quad \boldsymbol{\epsilon}_{\perp} + \boldsymbol{\epsilon}_{a} \sin^{2}\theta_{0} \cos\phi \quad \boldsymbol{\epsilon}_{a} \sin\theta_{0} \cos\theta_{0} \cos\phi \\ \boldsymbol{\epsilon}_{a} \sin\theta_{0} \cos\theta_{0} \sin\phi \quad \boldsymbol{\epsilon}_{a} \sin\theta_{0} \cos\theta_{0} \cos\phi \quad \boldsymbol{\epsilon}_{\perp} + \boldsymbol{\epsilon}_{a} \cos^{2}\theta_{0} \end{cases}$$

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(2.7)

where  $\epsilon_a = \epsilon_{||} - \epsilon_{\perp}$ , and  $\epsilon_{\perp} (=n_0^2)$  and  $\epsilon_{||} (=n_e^2)$  are the dielectric constants perpendicular and parallel to the local director, respectively, at the incident optical-field frequency. Thus, the total electromagnetic energy density of the optical field  $F_{opt}$  can be written as

$$F_{\rm opt} = -\frac{1}{8\pi} [\epsilon_{\perp} | \vec{E} |^2 + \epsilon_a \sin^2 \theta_0 | \vec{E} |^2 \cos^2(\phi_0 - \phi)] . \quad (2.8)$$

Similarly, the magnetic energy density of the dc magnetic field can be written as

$$F_{H} = -\frac{1}{2}\chi_{a}\sin^{2}\theta_{0}H_{0}^{2}\cos^{2}\phi . \qquad (2.9)$$

Consequently, the total free energy density of the SmC can be written as

$$F = \frac{1}{2} \alpha \sin^2 \theta_0 (\operatorname{grad} \phi)^2$$
$$- \frac{1}{8\pi} [\epsilon_\perp | \vec{\mathbf{E}} |^2 + \epsilon_a | \vec{\mathbf{E}} |^2 \sin^2 \theta_0 \cos^2(\phi_0 - \phi)]$$
$$- \frac{1}{2} \chi_a H_0^2 \sin^2 \theta_0 \cos^2 \phi . \qquad (2.10)$$

In general, the fields produced by a monochromatic wave incident upon a slab of SmC consists of four partial waves.<sup>25</sup> The fields as well as the polarization of the opti-

cal beam in the SmC clearly depends on the orientation of the SmC. The complete determination of the electromagnetic fields in the SmC is complicated and will be left for further investigation. To simplify the discussion, we shall fix the amplitude and polarization of the electric field. Then the term  $-\epsilon_1 |\vec{E}|^2 / 8\pi$  in the optical energy makes no contribution to the Euler equation. By the cylindrical symmetry of the problem, the relevant total free energy of the sample can be written as

$$\mathcal{F} = d \sin^2 \theta_0 \int_0^\infty \widetilde{F} dr$$

with the final planar total free energy density per area in the x-y plane  $\tilde{F}$  (erg/cm<sup>2</sup>) given by

$$\widetilde{F} = \begin{cases} \frac{1}{2} \left[ \alpha \left[ \frac{d\phi}{dr} \right]^2 + \frac{\epsilon_a |\vec{\mathbf{E}}|^2}{4\pi} \sin^2(\phi - \phi_0) + \chi_a H_0^2 \sin^2\phi \right] r, & r \le r_0 \\ \frac{1}{2} \left[ \alpha \left[ \frac{d\phi}{dr} \right]^2 + \chi_a H_0^2 \sin^2\phi \right] r, & r > r_0 \end{cases}$$

$$(2.11)$$

where d is the thickness of the sample and  $r_0$  is the cutoff radius defined by the profile of the intensity of light beam *I*:

$$I = \begin{cases} \text{const, } r \le r_0 \\ 0, r > r_0 \end{cases}.$$
 (2.12)

The resulting Euler equations take the forms

$$\frac{\partial^2 \phi}{\partial r^2} + \frac{1}{r} \frac{\partial \phi}{\partial r} - \frac{1}{2} [u^2 \cos(2\phi_0) + q^2] \sin(2\phi) + \frac{1}{2} u^2 \sin(2\phi_0) \cos(2\phi) = 0 \text{ for } r \le r_0 \quad (2.13a)$$

and

$$\frac{\partial^2 \phi}{\partial r^2} + \frac{1}{r} \frac{\partial \phi}{\partial r} - \frac{1}{2} q^2 \sin(2\phi) = 0 \text{ for } r > r_0 \qquad (2.13b)$$

where  $u \equiv |\vec{E}| \sqrt{\epsilon_a/4\pi\alpha}$  and  $q \equiv H_0 \sqrt{\chi_a/\alpha}$  are the inverse optical and magnetic coherence lengths. Since the intensity of the incident field in related to the electric field through  $I = cn_0 |\vec{\mathbf{E}}|^2 / 8\pi$ , we have  $u^2 = 2\epsilon_a I / cn_0 \alpha$ . For a typical SmC liquid crystal,  $\chi_a \sim 10^{-7}$  in cgs units,  $\alpha \sim 0.5 \times 10^{-6}$  dyn, and  $\epsilon_a \sim 0.6$ , we have  $q_2^2 \sim 0.2 H_0^2$  and  $u^2 \sim 0.5I$ , where I is expressed in mW/cm<sup>2</sup>. For a spot size of 100  $\mu$ m in radius and a magnetic field of about 1 kG,  $q \sim 450$  cm<sup>-1</sup> and  $qr_0 \sim 4.5$ ,  $u \sim 400$  cm<sup>-1</sup> for an optical field of about 100 mW in total power  $P = \pi r_0^2 I$ . In the following, we consider solutions which minimize the total free energy. Consequently,  $\phi$  must be a continuous function of r and must tend to zero (i.e., aligned in the magnetic field direction) as  $r \rightarrow \infty$ . At the boundary  $r=r_0$ , the solution and its first derivative must be Since  $\cos[2(2\pi - \phi_0)] = \cos(\pm 2\phi_0)$ continuous. and  $\sin[2(2\pi - \phi_0)] = \sin(-2\phi_0) = -\sin(2\phi_0)$ , the orienting effects of an optical field with polarization  $\phi_0$  is the same as that of an optical field with polarization  $-\phi_0$  or  $2\pi - \phi_0$ ,

except that  $\phi$  changes sign, i.e., if  $\phi_0 \rightarrow -\phi_0$  or  $\phi_0 \rightarrow 2\pi - \phi_0$ , then  $\phi(r) \rightarrow -\phi(r)$ .

Equations (2.13) are nonlinear in  $\phi$  and general solutions cannot be found except by numerical means.<sup>22</sup> However, by considering small distortions, the equations can be linearized and solved. Physically, there are now two competing alignment fields: the external magnetic field and the optical polarization field. It is these two fields together with the elastic energy that determines the spatial orientation of the SmC director.

## III. GENERAL PROPERTIES AND SMALL-DISTORTION-REGIME SOLUTION

## A. General properties of the solution at the origin and at infinity

We first formulate the general conditions for which the solution must satisfy. From the form of Eq. (2.13a), we can establish the general behavior of  $\phi$  at the origin. Equation (2.13a) is exactly satisfied by the constant deformation angle determined from the following equation

$$\tan(2\phi) = u^2 \sin(2\phi_0) / [u^2 \cos(2\phi_0) + q^2]$$
(3.1)

and so we may have  $\partial \phi / \partial r = 0$  at r = 0. Since  $\phi$  is bounded, we deduce that

$$\lim_{r \to 0} \left| r \frac{\partial^2 \phi}{\partial r^2} + \frac{\partial \phi}{\partial r} \right| = 0.$$
 (3.2)

Suppose that in the neighborhood of r=0 there exists a series

$$\frac{\partial \phi}{\partial r} = \sum_{n=0}^{\infty} C_n r^n .$$
(3.3)

Then

$$\lim_{r \to 0} \sum_{n=0}^{\infty} C_n r^n = -\lim_{r \to 0} \sum_{n=1}^{\infty} n C_n r^n , \qquad (3.4)$$

from which follows that  $C_0=0$ , so that the first derivative of the solution with respect to r must vanish at the origin:

$$\frac{\partial \phi}{\partial r} = 0 \text{ at } r = 0.$$
 (3.5)

Thus, there is no point defect at the origin. Consequently, the exponential functions  $\exp(ar)$ , the normal and modified Bessel functions of the second kind,  $Y_n(ar)$  and  $K_n(ar)$ , cannot be the solutions of the deformation angle for  $r < r_0$ , where a is a characteristic wave vector and n is any integer.

We now consider the asymptotic behavior of the deformation angle. The constribution to  $\mathcal{F}$  for  $r > r_0$  is

$$\mathscr{F}_{>} = \frac{1}{2}\alpha d \sin^2\theta_0 \int_{r_0}^{\infty} \left[ \left( \frac{\partial \phi}{\partial r} \right)^2 + q^2 \sin^2\phi \right] r \, dr \; . \; (3.6)$$

Therefore, the solution must tend to  $e^{-qr}/\sqrt{qr}$  so that the integral converges at infinity. Since the solution approaches zero for large r, the optical alignment effect is localized.

#### B. Linearized regime

We now consider the equilibrium orientation state in the linearized regime which can be described by linearizing Eqs. (2.13a) and (2.13b):

$$\frac{\partial^2 \phi}{\partial r^2} + \frac{1}{r} \frac{\partial \phi}{\partial r} - g^2 \phi + \frac{1}{2} u^2 \sin(2\phi_0) = 0 \quad \text{for} \quad r \le r_0 \qquad (3.7a)$$

and

$$\frac{\partial^2 \phi}{\partial r^2} + \frac{1}{r} \frac{\partial \phi}{\partial r} - q^2 \phi = 0 \quad \text{for} \quad r > r_0 , \qquad (3.7b)$$

where  $g^2 \equiv u^2 \cos(2\phi_0) + q^2$ .  $g^2$  is always positive for q > u. However, if  $q \le u$ ,  $g^2$  can be positive, zero, or negative depending on the polarization angle  $\phi_0$ . We let  $\phi_c$  be the smaller positively valued angle satisfying  $\phi_c = \frac{1}{2}\cos^{-1}(-q^2/u^2)$ . Then  $0 < \phi_c \le \pi/2$  and  $g^2 > 0$  if  $n\pi - \phi_c < |\phi_0| < n\pi + \phi_c$ , g = 0 if  $|\phi_0| = n\pi \pm \phi_c$ , and  $g^2 < 0$  if  $n\pi + \phi_c < |\phi_0| < (n+1)\pi - \phi_c$ , where  $n = 0, 1, 2, \ldots$ . In the following discussion, we always set  $g = |g^2|^{1/2}$ .

For  $r > r_0$ ,  $\phi$  satisfies the modified Bessel equation for which the solutions are the zeroth-order modified Bessel functions of the first and second kinds,  $I_0(qr)$  and  $K_0(qr)$ . Since as  $r \to \infty$ , the solution must tend to  $e^{-qr}/\sqrt{qr}$ , the physically acceptable solution is the zeroth-order modified Bessel function  $K_0(qr)$  of the second kind:

$$\phi = C_{>}^{s} K_{0}(qr) \text{ for } r > r_{0}$$
 (3.8)

The amplitude of the deformation angle for  $r > r_0$ ,  $C_>^s$ , will be determined later by matching the solution for  $r \le r_0$  at  $r = r_0$ .

For  $r \leq r_0$ , the nature of the solution depends on the azimuthal polarization angle  $\phi_0$ . We shall consider the following two cases separately.

Case 1. The incident wave is polarized at an oblique

angle to the dc magnetic field:  $\phi_0 \neq \pi/2$ .

Case 2. The incident wave is polarized normal to the dc magnetic field:  $\phi_0 = \pi/2$ .

#### C. Oblique polarization

For oblique polarization  $(\phi_0 \neq \pi/2)$ , the differential equation is inhomogeneous for  $r \leq r_0$  with a particular solution given by

$$\phi = C_0^s = \frac{1}{2} \tan^{-1} [u^2 \sin(2\phi_0)/g^2] . \qquad (3.9)$$

Notice that  $C_0^s$  is positive for  $g^2 > 0$ , negative for  $g^2 < 0$ ,  $\pi/4$  for g=0. The solution to the homogeneous part can be modified Bessel functions or normal Bessel functions of order zero, depending on the sign of  $g^2$  being positive or negative. Since the deformation angle is finite at r=0, the solution for the homogeneous part is  $I_0(gr)$  for  $g^2 > 0$  and  $J_0(gr)$  for  $g^2 < 0$ . Together with the particular solution, by imposing the boundary condition that the solutions and their first derivatives must be continuous at the boundary  $r=r_0$ , the amplitudes of the deformation angles can also be determined. For  $g^2 > 0$ , the solution which is finite at r=0 and which satisfies the boundary condition at  $r=r_0$  can be expressed in terms of the zeroth-order modified Bessel function of the first kind:

$$\phi = C_{<}^{s} I_{0}(gr) + C_{0}^{s} \quad \text{for} \quad r \le r_{0} \tag{3.10a}$$

with

$$C_{<}^{s} = \frac{qK_{1}(qr_{0})}{qI_{0}(gr_{0})K_{1}(qr_{0}) + gI_{1}(gr_{0})K_{0}(qr_{0})}C_{0}^{s}$$
(3.10b)

and

$$C_{>}^{s} = \frac{gI_{1}(gr_{0})}{qI_{0}(gr_{0})K_{1}(qr_{0}) + gI_{1}(gr_{0})K_{0}(qr_{0})}C_{0}^{s}$$
(3.10c)

for Eq. (3.8). The solution for  $g^2 < 0$  is the zeroth-order Bessel function of the first kind and has the form

$$\phi = C_{<}^{s} J_{0}(gr) + C_{0}^{s} \quad \text{for} \quad r \le r_{0} \tag{3.11a}$$

with

$$C_{<}^{s} = -\frac{qK_{1}(qr_{0})}{qJ_{0}(gr_{0})K_{1}(qr_{0}) - gJ_{1}(gr_{0})K_{0}(qr_{0})}C_{0}^{s} \qquad (3.11b)$$

and

$$C_{>}^{s} = \frac{gJ_{1}(gr_{0})}{qJ_{0}(gr_{0})K_{1}(qr_{0}) - gJ_{1}(gr_{0})K_{0}(qr_{0})}C_{0}^{s} \quad (3.11c)$$

for Eq. (3.8).<sup>26</sup> Since  $\sin[2(2\pi-\phi_0)] = \sin(-2\phi_0)$ =  $-\sin(2\phi_0)$  we have, as  $\phi_0 \rightarrow -\phi_0$  or  $\phi_0 \rightarrow 2\pi - \phi_0$ , then  $C_{<}^s \rightarrow -C_{<}^s$  and  $\phi(r) \rightarrow -\phi(r)$ , which show that the orienting effects of an optical field with polarization  $\phi_0$  is the same as that of an optical field with polarization  $-\phi_0$  or  $2\pi - \phi_0$ , except that  $\phi$  changes sign  $[\phi(r) \rightarrow -\phi(r)]$  as shown earlier in Sec. II.

For the special case where g=0, i.e.,  $\cos(2\phi_0) = -q^2/u^2$ , the linearized equation for  $r \le r_0$  takes the form

$$\frac{1}{r}\frac{\partial}{\partial r}\left[r\frac{\partial\phi}{\partial r}\right] + \frac{1}{2}u^2\sin(2\phi_0) = 0. \qquad (3.12)$$

Again, by using the boundary conditions at r=0 and  $r=r_0$ , the complete solution for g=0 is

$$\phi = \frac{1}{8}u^{2}\sin(2\phi_{0})\left[r_{0}^{2} - r^{2} + \frac{2r_{0}K_{0}(qr_{0})}{qK_{1}(qr_{0})}\right] \text{ for } r \leq r_{0}$$
(3.13a)

with

$$C_{>}^{s} = r_{0}u^{2}\sin(2\phi_{0})/[4qK_{1}(qr_{0})]$$
 (3.13b)

for Eq. (3.8).

The spatial orientation of the azimuthal angle of the director as a function of the optical-field intensity is shown in Fig. 2. For  $qr_0=5$  and  $\phi_0=40^\circ$ , substantial alignment is already present for u = q/2 or incident total power of 30 mW. Almost complete alignment at the center of the spot can be achieved with u = 3q or  $P \sim 1.1$ W. (For  $r_0 \sim 100 \ \mu m$ ,  $qr_0 \sim 5$  corresponds to a magnetic field of 1.1 kG.) Figure 3 shows the spatial orientation of the azimuthal angle of the director as a function of the radius of the laser illumination spot for  $q = 500 \text{ cm}^{-1}$ ,  $ur_0=2.5$ , and  $\phi_0=40^\circ$ . With magnetic field and incident optical power fixed, further increases the alignment can be achieved by using a smaller laser spot at low laser power. The alignment at the center of the spot can increase dramatically by a reduction of the illumination-spot radius from 100 to 10  $\mu$ m. The dependence of the deformation on the polarization of the optical field is shown in Fig. 4 for a fixed magnetic field and optical beam power. The results indicate that there is a polarization angle  $\phi_0$ which gives the maximum alignment effect.

### D. Normal polarization

For a normally polarized incident light beam  $(\phi_0 = \pi/2)$ , there exists a threshold intensity below which no molecular reorientation can be induced. To investigate the



FIG. 3. Spatial orientation of the azimuthal angle of the director as a function of the radius of laser illumination spot for  $q = 500 \text{ cm}^{-1}$ ,  $ur_0 = 2.5$ , and  $\phi_0 = 40^\circ$ .

threshold behavior, we again linearize Eq. (2.13a) at small distortion. The resulting equation takes the form of the zeroth-order Bessel equation

$$\frac{\partial^2 \phi}{\partial r^2} + \frac{1}{r} \frac{\partial \phi}{\partial r} + (u^2 - q^2)\phi = 0 \quad \text{for} \quad r \le r_0 \;. \tag{3.14}$$

We let  $w^2 \equiv |u^2 - q^2|$ . We first consider the case where u < q. Then the possible solutions are 0,  $K_0(wr)$ , and  $I_0(wr)$ . Neither  $K_0(wr)$  nor  $I_0(wr)$  can be the solution because the first derivative of  $K_0(wr)$  diverges at the origin and  $I_0(wr)$  does not satisfy the condition of continuity of the logarithmic derivative to a  $K_0(qr)$  function at the boundary  $r = r_0$ . Therefore, the solution zero is the only acceptable solution for u < q.

When  $u \ge q$ , the solutions of Eq. (3.14) are 0,  $J_0(wr)$ , and  $Y_0(wr)$ .  $Y_0(wr)$  is not an acceptable solution since it diverges at r=0. Consequently, the only possible nonzero solution is the zeroth-order Bessel function of the first kind  $J_0(wr)$ . The continuity of the logarithmic derivative of the solutions at the boundary  $r=r_0$  yields the following equation for determining the threshold intensity<sup>27,28</sup>:



FIG. 2. Spatial orientation of the azimuthal angle of the director as a function of the reduced optical-field intensity  $u = [2\epsilon_a I/cn_0\alpha]^{1/2}$  for  $qr_0 = 5$  and  $\phi_0 = 40^\circ$ .



FIG. 4. Spatial orientation of the azimuthal angle of the director as a function of the polarization of the optical field  $\phi_0$  for  $ur_0 = qr_0 = 5$ .

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$$\frac{w_{\rm th}J_1(w_{\rm th}r_0)}{J_0(w_{\rm th}r_0)} = \frac{qK_1(qr_0)}{K_0(qr_0)} .$$
(3.15)

In the case  $qr_0 \ll 1$  we obtain, from Eq. (3.15),

$$w_{\rm th}r_0 \sim -2/[0.5772 + \ln(qr_0/2)]$$
. (3.16)

For  $qr_0 \sim 1$  we have

$$w_{\rm th} r_0 \sim 1.4$$
 (3.17)

For  $qr_0 > \pi$ ,

$$w_{\rm th}r_0 \sim J_{01}[1 - J_{01}K_0(qr_0)/qr_0K_1(qr_0)],$$
 (3.18)

where  $J_{01} \approx 2.405$  is the first zero of the Bessel function  $J_0$ . In the limit of  $qr_0 \gg 1$ , Eq. (3.18) shows that  $w_{th}r_0 \sim J_{01}$ . The threshold intensity is then given by

$$I_{\rm th} = cn_0 \alpha u_{\rm th}^2 / 2\epsilon_a , \qquad (3.19)$$

where  $u_{th}^2 = w_{th}^2 + q^2$ . The threshold power of the light beam is

$$P_{\rm th} = \pi r_0^2 I_{\rm th} = \pi c n_0 \alpha u_{\rm th}^2 r_0^2 / 2\epsilon_a$$

For  $qr_0 \sim 5$ ,  $u_{th}r_0 \sim 5.4$  and  $P_{th}$  is of the order of 145 mW. In the limit  $qr_0 \ll 1$ ,  $w_{th}r_0$  is much greater than  $qr_0$  so that  $u_{th} \sim w_{th} \sim -2/r_0[0.5772 + \ln(qr_0/2)]$ . Thus the threshold optical energy is essentially independent of the external magnetic field and works against the elastic deformation energy. For  $qr_0 \gg 1$ ,  $w_{th}r_0 \sim J_{01} \ll qr_0$  so that  $u_{th} \sim q$  which shows that  $|\vec{\mathbf{E}}_{th}| \sim 2\sqrt{\chi_a/\epsilon_a}H_0$  and the threshold optical field is essentially competing against the magnetic field. The reduced threshold power as a function of the spot size and reduced magnetic field strength  $qr_0$  is shown in Fig. 5.

The amplitude of the azimuthal angle in the region above the threshold can not be determined from the linear approximation, but can be determined with allowance for



FIG. 5. Threshold power as a function of the spot size and reduced magnetic field strength  $qr_0 = \sqrt{\chi_a/\alpha}H_0r_0$ . Threshold power  $P_{\rm th}$  is related to  $(u_{\rm th}r_0)^2$  by  $P_{\rm th} = \pi c n_0 \alpha (u_{\rm th}r_0)^2/2\epsilon_a$ . For a typical SmC sample,  $P \sim 5(ur_0)^2$  and the thresold power for  $P_{\rm th} = 5(u_{\rm th}r_0)^2$  is shown in the figure where the power is expressed in mW. Insert shows the variation of the function  $w_{\rm th}r_0 = (u_{\rm th}^2 - q^2)^{1/2}r_0$  with respect to  $\log_{10}(qr_0)$ .

the nonlinear terms in the Euler equation. By expanding Eq. (2.13a) up to and including terms  $\sim \phi^3$  we obtain

$$\frac{\partial^2 \phi}{\partial r^2} + \frac{1}{r} \frac{\partial \phi}{\partial r} + w^2 (1 - \frac{2}{3} \phi^2) \phi = 0 \text{ for } r \le r_0 . \quad (3.20)$$

We look for an approximate solution of the form

$$\phi = C_{<}^{s} J_{0}(w_{\rm th}r) , \qquad (3.21)$$

where  $C_{<}^{s} = \phi(r=0)$  is the deformation angle at the origin. By putting Eq. (3.21) into Eq. (3.20) and evaluating at r=0 we obtain

$$C_{<}^{s} = [3(1 - w_{\rm th}^{2}/w^{2})/2]^{1/2}$$
 (3.22)

Therefore, when the incident intensity is above threshold, the approximate solutions are given by Eqs. (3.8) and (3.21) with the respective amplitudes given by Eq. (3.22) and

$$C_{>}^{s} = C_{<}^{s} J_{0}(w_{\rm th}r_{0}) / K_{0}(qr_{0})$$
(3.23)

outside the illuminated region.

#### E. Dynamic response

We now consider the time dependence of the molecular reorientation. For the total free energy density defined by Eq. (2.1) we include a dissipative term  $\eta(\partial \hat{n}/\partial t)/2$  which will contribute a viscous torque opposing any rapid change of the director, where  $\eta$  is the viscosity of the SmC. The dynamic behavior is described by the resulting Euler equations of the forms

$$\frac{\partial^2 \phi}{\partial r^2} + \frac{1}{r} \frac{\partial \phi}{\partial r} - \frac{1}{2} g^2 \sin(2\phi) + \frac{1}{2} u^2 \sin(2\phi_0) \cos(2\phi)$$
$$= h^2 \frac{\partial \phi}{\partial t} \quad \text{for } r \le r_0 \qquad (3.24a)$$

and

$$\frac{\partial^2 \phi}{\partial r^2} + \frac{1}{r} \frac{\partial \phi}{\partial r} - \frac{1}{2} q^2 \sin(2\phi) = h^2 \frac{\partial \phi}{\partial t} \quad \text{for } r > r_0 \qquad (3.24b)$$

where  $h^2 \equiv \eta / \alpha$ .

Although backflow effects are completely ignored in the dynamic equations (3.24a) and (3.24b), these equations are still complicated even in the linearized regime. In the following, we consider only the case of normal polarization in the small-distortion regime which can be described by

$$\frac{\partial^2 \phi}{\partial r^2} + \frac{1}{r} \frac{\partial \phi}{\partial r} + w^2 (1 - \frac{2}{3} \phi^2) \phi = h^2 \frac{\partial \phi}{\partial t} \quad \text{for } r \le r_0 . \quad (3.25)$$

By assuming a weak time dependence of the spatial distribution of the deformation angle, we look for an approximate solution for  $I \ge I_{\text{th}}$  of the form

$$\phi(r,t) = \begin{cases} C_{<}^{d}(t)J_{0}(w_{\text{th}}r), & r \leq r_{0} \\ C_{>}^{d}(t)K_{0}(qr), & r > r_{0} \end{cases}$$
(3.26)

where  $C_{<}^{d}$  and  $C_{>}^{d}$  are the amplitudes of the distortion for  $r \leq r_{0}$  and  $r > r_{0}$ , respectively. Putting Eq. (3.26) into Eq. (3.25) and evaluating at r=0, the amplitude for the distortion within the optical-field illuminating region is described by

$$\frac{\partial}{\partial t}C^d_{<} = C^d_{<}\left[a - b(C^d_{<})^2\right], \qquad (3.27)$$

where  $a \equiv (w^2 - w_{\text{th}}^2)/h^2$  and  $b \equiv 2w^2/3h^2$ . Equation (3.27) is of the same form for the optically induced Freedericksz transition in a NLC.<sup>10</sup> Since there is no constant term on the right-hand side of the equation,  $C_{<}^d = 0$  can be a solution. Therefore, we need a small fluctuation  $C_i$  at t=0 to get the distortion started. Equation (3.27) can then be solved to give

$$C_{<}^{d} = \left[\frac{1}{1+C_{r}^{2}e^{-2at}}\right]^{1/2}C_{<}^{s} , \qquad (3.28)$$

where  $C_r = [(C_{\leq}^s / C_i)^2 - 1]^{1/2}$  and

$$C_{<}^{s} = \sqrt{a/b} = [3(1 - w_{\text{th}}^{2}/w^{2})/2]^{1/2}$$
$$= \lim_{t \to \infty} C_{<}^{d}$$

which is the static solution [Eq. (3.22)]. By matching the solution at  $r=r_0$ , we have

$$C_{>}^{d}(t) = C_{<}^{d}(t)J_{0}(w_{\rm th}r_{0})/K_{0}(qr_{0}) . \qquad (3.29)$$

At  $t \sim 0$ , Eq. (3.28) describes the exponential growth of a small fluctuation  $C_i$  with a time constant 1/a:

$$C^{d}_{<}(t \sim 0) \sim C_{i} e^{at}$$
 (3.30)

For  $t \gg 1$ , the deformation amplitude  $C_{<}^{d}$  reaches its final value  $C_{<}^{s}$  exponentially with a smaller time constant 1/2a:

$$C^{d}_{<}(t \gg 1) \sim (1 - \frac{1}{2}C^{2}_{r}e^{-2at})C^{s}_{<}$$
 (3.31)

Note that the time constant 1/a is proportional to  $(w^2 - w_{\rm th}^2)^{-1}$  and is dependent on the incident laser power. For a 10- $\mu$ m-radius laser beam incident on a sample in an external 1-kG magnetic field, the threshold power is ~8 mW. Using  $\eta \sim 1$  cp and  $\alpha \sim 0.5 \times 10^{-6}$  dyn for a typical SmC sample, the time constant is ~16 msec for incident power twice threshold, decreasing to 2 msec for a power ten times threshold. Hence, in general, the response time is of the order of milliseconds, which is faster than typical response time<sup>7,8,10</sup> in the nematic case.

## IV. OPTICAL REFLECTIVITY AND TRANSMISSIVITY

Experimentally, the molecular reorientation can be qualitatively measured by the reflectivity or transmissivity of a normally incident probe beam. In the following, we consider a probe beam incident normally onto the SmC polarized along *OP* which is selected by a polarizer making an angle  $\phi_p$  with the y axis (Fig. 6). On entering the SmC, each ray is divided into two rays with different effective refractive indices, and with their electric displacement vectors  $D_p$  and  $D_v$  vibrating in two mutually orthogonal directions at right angles to the SmC normal.  $D_p$  lies in the plane containing the optical axis of the SmC. The ray vibrating along  $D_p$  acts like the extraordinary ray having an effective refractive index

$$n_p = n_{op} n_{ep} / (n_{op}^2 \sin^2 \theta_0 + n_{ep}^2 \cos^2 \theta_0)^{1/2}$$
,



FIG. 6. Construction of the vibration components transmitted by a polarizer and analyzer for the reflectivity and transmissivity measurements. The director of the SmC is oriented at an azimuthal angle  $\phi$  with the y axis. A probe beam is incident normally onto the SmC with polarization selected by a polarizer which is denoted by *OP* making an angle  $\phi_p$  with the y axis. On entering the SmC, each ray is divided into two rays with different effective refractive indices, and with their electric displacement vectors  $D_p$  and  $D_v$  vibrating in two mutually orthogonal directions at right angles to the SmC normal.  $D_p$  lies in the plane containing the optical axis of the SmC. The ray vibrating along  $D_p$  acts like the extraordinary ray. The ray vibrating along  $D_v$  acts like the ordinary ray.  $\vec{E}_0$  is the electric field of the probe beam.  $\Psi = \phi_p - \phi$  is the angle that the polarizer makes with the plane containing the optical axis  $OD_p$ . The measured quantity can be either the reflectivity or the transmissivity. The direction of vibration of the measured field is selected by an analyzer which is represented by OA making an angle  $\chi$  with the polarizer.

where  $n_{op}$  and  $n_{ep}$  are the ordinary and the extraordinary refractive indices at the probe-beam wavelength  $\lambda_p$ . The ray vibrating along  $D_v$  acts like the ordinary ray having an effective refractive index  $n_v = n_{op}$ . The direction of vibration of the measured field is selected by an analyzer which is denoted by OA and the measured quantity can be either the reflectivity or the transmissivity.

In the following, we denote by subscripts p and v quantities referring to the ray vibrating along  $D_p$  and  $D_v$ . We let  $R_j$  and  $T_j$  be the reflection and transmission coefficients of the ray vibrating along  $D_j$  (j=p or v). Since the azimuthal angle of the director depends only on r, the SmC medium can be considered as an anisotropic dielectric medium with an refractive index depending on r but is independent of z. Then the refractive index N of the system is

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$$N = \begin{cases} 1, & z < 0 \\ n_j(r), & 0 \le z \le d \\ 1, & z > d \end{cases}$$
(4.1)

The amplitudes of the reflected (r) and transmitted (t) waves at the interfaces z=0 (subscript 1) and z=d (subscript 2) are, respectively,<sup>29</sup>

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(4.2)

(4.3)

$$(r_1)_j = (n_j - 1)/(1 + n_j),$$
  
 $(r_2)_j = (1 - n_j)/(1 + n_j) = -(r_1)_j,$ 

and

$$(t_1)_j = 2/(1+n_j)$$
,  
 $(t_2)_j = 2n_j/(1+n_j) = n_j(t_1)_j$ .

By taking into account multiple reflections at the two interfaces z=0 and z=d, the reflection and transmission coefficients are

$$R_{j} = \frac{(r_{1})_{j} + (r_{2})_{j} e^{i2\beta_{j}}}{1 + (r_{1})_{j} (r_{2})_{j} e^{i2\beta_{j}}}$$

and

$$T_{j} = \frac{(t_{1})_{j}(t_{2})_{j}e^{i\beta_{j}}}{1 + (r_{1})_{j}(r_{2})_{j}e^{i2\beta_{j}}},$$

where  $\beta_j \equiv 2\pi n_j d / \lambda_p$ . With the use of Eq. (4.2),  $R_j$  is reduced to

$$R_j = |R_j| e^{i\delta'_j} \tag{4.4a}$$

with

$$|R_{j}| = \frac{(n_{j}^{2} - 1)\sin\beta_{j}}{[4n_{j}^{2} + (n_{j}^{2} - 1)^{2}\sin^{2}\beta_{j}]^{1/2}}$$
(4.4b)

and

$$\tan \delta_j^r = -\frac{2n_j}{1+n_j^2} \cot \beta_j . \qquad (4.4c)$$

Consequently, the phase difference between the two reflected rays  $\delta' = \delta'_p - \delta'_v$  is given by

$$\tan \delta^{r} = 2 \frac{n_{v}(1+n_{p}^{2})\tan\beta_{p}-n_{p}(1+n_{v}^{2})\tan\beta_{v}}{4n_{p}n_{v}+(1+n_{p}^{2})(1+n_{v}^{2})\tan\beta_{p}\tan\beta_{v}} .$$
(4.5)

Similarly, the transmission coefficient can be reduced to

$$T_j = |T_j| e^{i\delta_j^t} \tag{4.6a}$$

with

$$T_j \mid = \frac{2n_j}{[4n_j^2 + (n_j^2 - 1)^2 \sin^2 \beta_j]^{1/2}}$$
(4.6b)

and

$$\tan \delta_j^t = \frac{1+n_j^2}{2n_j} \tan \beta_j = \tan \left[ \frac{\pi}{2} + \delta_j^r \right] \,. \tag{4.6c}$$

The phase difference between the two transmitted rays  $\delta^t = \delta_p^t - \delta_v^t$  is the same as  $\delta^r$ , i.e.,  $\delta^r = \delta^t \equiv \delta$ . By Eqs. (4.4)–(4.6), the reflection and transmission coefficients satisfy  $|R_j|^2 + |T_j|^2 = 1$  which is in agreement with the law of conservation of energy.

Since the analyzer transmits only the components parallel to OA, the resultant fields of two rays after passing through the analyzer are given by

$$E_{op} = E_p Q_p \cos(\Psi - \chi) e^{i\delta_j}$$
  
=  $E_0 Q_p \cos\Psi \cos(\Psi - \chi) e^{i\delta_j}$   
(4.7)

and

$$E_{ov} = E_v Q_v \sin(\Psi - \chi) e^{i\delta_j}$$
  
=  $E_0 Q_v \sin\Psi \sin(\Psi - \chi) e^{i\delta_j}$ ,

where  $\Psi \equiv \phi_p - \phi$  is the angle that the polarizer makes with  $OD_p$ ,  $\chi$  is the angle between the analyzer and the polarizer,  $Q_j = |R_j|$  and  $\delta_j = \delta'_j$  for the reflectivity measurement,  $Q_j = |T_j|$  and  $\delta_j = \delta'_j$  for the transmissivity measurement, and  $E_0$  is the electric field of the probe beam. The two rays  $E_{op}$  and  $E_{ov}$  are superposed at the observation point with the field given by  $\vec{E}_{op} = \vec{E}_{op} + \vec{E}_{ov}$ :

$$I_{ob} = \frac{c}{4\pi} \langle \vec{E}_{ob}^2 \rangle$$
  
=  $I_0 \left[ [Q_p \cos \chi + (Q_v - Q_p) \sin \Psi \sin(\Psi - \chi)]^2 - Q_p Q_v \sin(2\Psi) \sin[2(\Psi - \chi)] \sin^2 \frac{\delta}{2} \right], \quad (4.8)$ 

where  $I_0 = c \langle \vec{E}_0^2 \rangle / 4\pi$  is the probe-beam intensity. Equation (4.8) is the general expression for the reflected and transmitted intensities for the probe beam at a point where the local orientation of the director is known. In our calculations, the molecular orientation is cylindrically symmetric. Hence, the total reflected or transmitted power of the probe beam covering an area of radius  $r_{\rm ob}$  is given by

$$P_t = 2\pi \int_0^{\gamma_{\rm ob}} I_{\rm ob}(r) r \, dr \; . \tag{4.9}$$

(4.10)

Consequently, the reflectivity  $(\mathcal{R})$  and transmissivity  $(\mathcal{F})$  are given by  $P_t/P_0$  where  $P_0 = \pi r_{ob}^2 I_0$  is the power of the probe beam:

$$\mathscr{R} = \frac{2}{r_{ob}^2} \int_0^{r_{ob}} \left[ \left[ \left| R_p \right| \cos \chi + \left( \left| R_v \right| - \left| R_p \right| \right) \sin \Psi \sin(\Psi - \chi) \right]^2 - \left| R_p \right| \left| R_v \right| \sin(2\Psi) \sin 2 \left[ \left( \Psi - \chi \right) \right] \sin^2 \frac{\delta}{2} \right] r \, dr$$

and

$$\mathcal{F} = \frac{2}{r_{ob}^2} \int_0^{r_{ob}} \left[ \left[ |T_p| \cos \chi + (|T_v| - |T_p|) \sin \Psi \sin(\Psi - \chi) \right]^2 - |T_p| |T_v| \sin(2\Psi) \sin[2(\Psi - \chi)] \sin^2 \frac{\delta}{2} \right] r \, dr \; .$$

We now consider two special cases corresponding to polarized and depolarized reflection and transmission, respectively.

Case 1. Polarizer and analyzer are parallel. Then  $\chi = 0$  and

$$I_{ob} = I_0 \left[ [Q_p + (Q_v - Q_p)\sin^2 \Psi]^2 - Q_p Q_v \sin^2 2\Psi \sin^2 \frac{\delta}{2} \right] .$$
(4.11)

*Case 2.* Polarizer and analyzer are perpendicular. Then  $\chi = \pi/2$  and

$$I_{\rm ob} = I_0 \sin^2 2\psi \left[ \frac{1}{4} (Q_v - Q_p)^2 - Q_p Q_v \sin^2 \frac{\delta}{2} \right]. \quad (4.12)$$

Figure 7 shows the depolarized reflectivity and transmissivity for a 3000-Å-thick SmC film as a function of reduced reorientating intensity  $u/q = [2\epsilon_a I/cn_0\chi_a H^2]^{1/2}$ . The probe beam has a wavelength of 6328 Å, polarized along the direction of the unperturbed director (y axis) and has the same spot size as the orienting beam which is polarized at 40° to the y axis (same case as presented in Fig. 2). The depolarized intensities increase strongly and approach limiting values as the power of the orienting laser beam is increased. Therefore, these molecular reorientation effects are measurable and can be quantitatively compared with the calculations presented in this paper.



FIG. 7. Optical reflectivity and transmissivity as a function of the reduced intensity  $u/q = [2\epsilon_a I/cn_0\chi_a H^2]^{1/2}$  for a cell of 0.3  $\mu$ m thick with  $qr_0=5$  and  $\phi_0=40^\circ$ . For the probe beam, we set  $\lambda_p=6328$  Å,  $n_{op}=1.55$ ,  $n_{ep}=1.75$ ,  $\theta_0=30^\circ$ ,  $\chi=90^\circ$ , and  $\phi_p=0^\circ$ .

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