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Optical-Field-Induced Birefringence and Freedericksz Transition in a Nematic Liquid Crystal

S. D. Durbin, S. M. Arakelian,[a] and Y. R. Shen
Department of Physics, University of California, Berkeley, California 94720

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Optical-field–induced birefringence in nematic 4-cyano-4′-pentylbiphenyl was measured with cw pump and probe beams, and the optical-field–induced Freedericksz transition was observed for the first time. The results are in quantitative agreement with the theoretical prediction.

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Laser-induced molecular reorientation is a common cause of optical nonlinearity in a fluid medium. In this respect, liquid crystals are often strongly nonlinear because of their large molecular anisotropy and strong correlation between molecules. The nonlinear optical properties of liquid crystals in the isotropic phase have already been studied quite extensively by a number of researchers in the past decade.[1] This is, however, not true for liquid crystals in the mesophases. Not until recently have limited theoretical and experimental studies of nonlinear optical properties of mesophases appeared in the literature.[2-4] Actually, as a result of the collective behavior of molecules, the optical nonlinearity of a mesophase can be extraordinarily large.[3] Crudely speaking, in reorienting the molecules, an optical field is equivalent to a dc field, with the optical dielectric anisotropy replacing the dc dielectric anisotropy. (A more detailed description of the optical-field–induced reorientation is outlined below.) It is known that a dc field of ~ 1 esu is sufficient to induce a significant reorientation of the molecules in a mesophase. The same is expected with an optical field of comparable strength or ~ 100 W/cm^2 in intensity. Such a field can easily be obtained by focusing a cw laser beam. In fact, a cw laser beam is best for observing the molecular reorientation effect because the response of the collective motion of the molecules is usually very slow. Two groups have recently published the observation of optical-field–induced effects in the nematic mesophase.[4] We should like to report here the first quantitative measurements of the very strong optical-field–induced reorientation of molecules in the nematic liquid crystal 4-cyano-4′-pentylbiphenyl (5CB). The results are shown to be in excellent agreement with the theory. In particular, the optical-field–induced Freedericksz transition is observed as predicted by the theory below.

In the mesophase, the average direction of molecular orientation is given by the director $\hat{n}$. For a p-polarized incident beam, the theory of field-induced molecular reorientation follows the usual derivation starting from the free-energy density[5]

$$F = \tfrac{1}{2}[K_{11}(\nabla\cdot\hat{n})^2 + K_{22}(\hat{n}\cdot\nabla\times\hat{n})^2 + K_{33}(\hat{n}\times\nabla\times\hat{n})^2 - Sn_r/c] \tag{1}$$

for a nematic liquid crystal, where K_{ii} are the Frank elastic constants. The total electromagnetic energy density $\vec{E}\cdot\vec{D}/4\pi$ is here written as Sn_r/c, where S is the magnitude of the Poynting vector, c/n_r is the ray velocity with $n_r = \{(\epsilon_\parallel + \epsilon_\perp)/\epsilon_\parallel\epsilon_\perp - [\epsilon_\perp + \Delta\epsilon(\hat{n}\cdot\hat{k})^2]^{-1}\}^{-1/2}$, $\epsilon_\perp$ and $\epsilon_\parallel$ are the optical dielectric constants parallel and perpendicular to the director $\hat{n}$, respectively, $\Delta\epsilon = \epsilon_\parallel - \epsilon_\perp$, and $\hat{k}$ is the propagation direction. Assume the sample geometry in Fig. 1 with $\hat{n}$ lying in the $\hat{x}$-$\hat{z}$ plane and being a function of z only. If $\theta(z)$ is the angle between $\hat{n}$ and $\hat{z}$ at z, then $n_x = \sin\theta(z)$ and $n_z = \cos\theta(z)$. The free-energy density becomes a function of $\theta(z)$. In the infinite-plane-wave approximation, S is a constant throughout the medium.[6] Then, minimization of the free energy $\mathfrak{F} = \int F\,d^3r$ leads to the Euler equation which can be integrated to give

$$\frac{d\theta}{dz} = \pm\left(\frac{G(\theta) - G(\theta_m)}{H(\theta)}\right)^{1/2}, \tag{2}$$

where $G(\theta) = -Sn_r(\theta)/cK_{33}$, $H(\theta) = \cos^2\theta + (K_{11}/K_{33})\sin^2\theta$, and θ_m is the value of θ at the center of the sample cell (chosen as $z = 0$). The solution of Eq. (2) with the proper boundary conditions yields $\theta(z)$ which describes the molecular reorientation by the field.

In this paper, we are interested in the case where the initial molecular alignment is homeotropic. We expect $\theta(z) = \theta(-z)$. With the boundary condition $\theta(\pm d/2) = 0$, integration of Eq. (2) yields for $-d/2 \leqslant z \leqslant 0$

$$z + \frac{d}{2} = \int_0^\theta \left[\frac{G(\theta') - G(\theta_m)}{H(\theta')}\right]^{1/2} d\theta'. \tag{3}$$

The value of θ_m can be obtained by setting $\theta=\theta_m$ at $z=0$ in the above equation. A normally incident probe beam with a wavelength λ_p traversing the medium should have its extraordinary component experience an induced phase shift

$$\varphi=\frac{2\pi}{\lambda_p}\int_{-d/2}^{d/2}\left[\frac{n_0 n_e}{(n_e^2\cos^2\theta+n_0^2\sin^2\theta)^{1/2}}-n_0\right]dz, \tag{4}$$

where n_0 and n_e are respectively the ordinary and maximum extraordinary refractive indices of the nematic medium. Equations (3) and (4) have analytical solutions only in especially simple cases, but they can in general be solved numerically to give $\theta(z)$ and φ as a function of the pump field intensity $I=S=cn_0|\mathscr{E}|^2/8\pi$ and the angle α. For $\alpha=0$, there is a threshold intensity $I_{\text{th}}=(c/n_0)(\epsilon_{\parallel}^2/\epsilon_{\perp})(K_{33}/\Delta\epsilon)(\pi^2/d^2)$, below which no molecular reorientation can be induced. This is analogous to the dc-field-induced Freedericksz transition. Unlike the dc case, however, the optical-field-induced Freedericksz transition cannot occur for initially planar molecular alignment. This is a consequence of Mauguin's "adiabatic theorem" which states that a light beam with ordinary or extraordinary polarization rotates its polarization to follow a slow distortion of the director.[7]

The dynamic behavior of the optical-field-induced Freedericksz transition is also analogous to the dc case.[8] The initial response of the induced molecular reorientation to the laser switch-on and the long-time response to the laser switch-off are both exponential with relaxation times τ_{on} and τ_{off}, respectively.

$$\tau_{\text{on}}=\frac{c}{n_0}\frac{\epsilon_{\parallel}^2}{\epsilon_{\perp}}\frac{\gamma_1^*}{\Delta\epsilon}(I-I_{\text{th}})^{-1}, \qquad \tau_{\text{off}}=\gamma_1^* d^2/K_{33}\pi^2, \tag{5}$$

where γ_1^* is a Leslie viscosity coefficient corrected for the backflow effect. Measurements of τ_{on} and τ_{off} as a function of I allow us to determine γ_1^*, I_{th}, and K_{33} if the other parameters are known.

In our experiment, we used a 250-μm-thick sample of 5CB sandwiched between glass slides treated with dimethyl-n-octadecyl-3-aminopropyltrimethoxysilyl-chloride to give homeotropic molecular alignment. The sample was situated in an oven with a temperature stability of ± 0.05 K. A cw Ar^+ laser was used as the pump beam to induce the molecular reorientation. At high Ar^+ laser intensity ($\gtrsim 350$ W/cm^2) local heating of the sample by ~ 2 K was noticed. Therefore, the sample cell was placed horizontally to minimize the convection flow that may affect the measurements. A He-Ne laser was used to probe the optical birefringence φ resulting from the induced molecular reorientation. To make sure that the transverse variation of the pump beam intensity has little effect on the result, the probe beam at the sample should have a radius less than $2\pi/|\nabla\varphi|$. This was achieved by focusing the He-Ne beam tightly and using a diaphragm to limit the probe region to $\lesssim 6$ μm as compared to the Ar^+ laser spot of ~ 800 μm at the sample. The measured φ were collected and analyzed by a microcomputer.

To observe the optical-field-induced Freedericksz transition, the angle of incidence of the pump beam was set at ~ 0°. The observed birefringence versus pump intensity at $T_{NI}-T=9.2$ K is shown in Fig. 1. It exhibits a threshold intensity at $I_{\text{th}}\simeq 155$ W/cm^2. According to Eq. (5), I_{th} can also be obtained from the measurements of τ_{on}^{-1} vs I, which exhibits a critical slowing down behavior as I approaches I_{th}. Our results are shown in Fig. 2. The linear fit to the data also yields $I_{\text{th}}=155$ W/cm^2, in very good agreement with the direct observation. It also gives a value for the viscosity coefficient $\gamma_1^*=0.89$ P, using $n_0=1.54$ and $n_e=1.73$ for 5CB. The measured value of I_{th} allows us to find $K_{33}=0.8\times10^{-6}$ dyn. Then, substituting the values of γ_1^* and K_{33} in Eq. (5), we obtain $\tau_{\text{off}}\simeq 70$ sec, which is in good agreement with our directly measured value of $\tau_{\text{off}}=63$

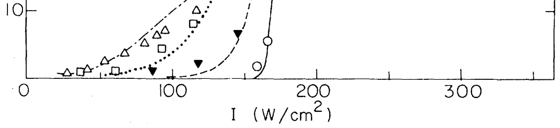

FIG. 1. Experimental points and theoretical curves for the induced birefringence at different angles α: circles and solid curve, $\alpha=0°$; solid triangles and dashed curve, $\alpha=3°$; squares and dotted curve, $\alpha=11°$; open triangles and dot-dashed curve, $\alpha=30°$. Inset shows experimental geometry.

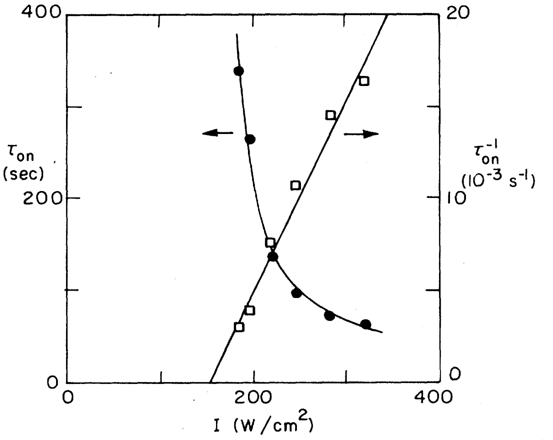

FIG. 2. Turn on time constant for $\alpha = 0°$; the straight line for τ_{on}^{-1} is least-squares fitted and determines the curve for τ_{on}.

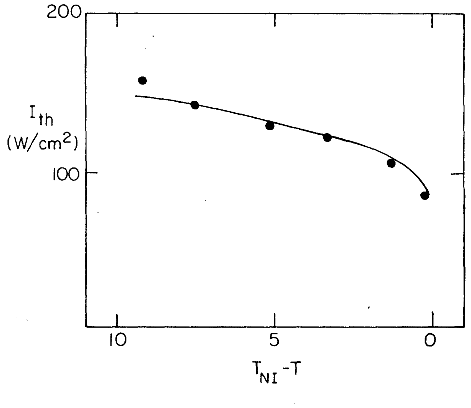

FIG. 3. Temperature dependence of threshold intensity at $\alpha = 0°$. The normalized curve shows the temperature variation of the order parameter $\langle P_2 \rangle$, computed from data in Ref. 11.

sec. We notice that our values of γ_1^* and K_{33} are fairly close to those found in the literature (Ref. 10), $\gamma_1^* = 0.67$ P and $K_{33} = 0.85 \times 10^{-6}$ dyn. The discrepancy presumably arises from the fact that the optical field had a transverse dimension of only ~ 800 μm.

With the pump beam incident at an angle α and the probe beam at normal incidence, the observed birefringence as a function of pump intensity, shown also in Fig. 1 for several values of α, exhibits no threshold behavior. Theoretical curves for these cases can be calculated from Eqs. (3) and (4) using our value of K_{33} and the values of K_{11}/K_{33}, n_o, and n_e from the literature.[9,10] In the actual experiment, the pump beam suffered a scattering loss in traversing the medium, and the probe beam experienced a small transverse variation of the pump intensity and hence the induced birefringence when α is finite. It is difficult to include these effects rigorously in the theoretical calculation. In obtaining the theoretical curves in Fig. 1, we simply assumed that the scattering loss would decrease the pump intensity uniformly through the sample, and the transverse variation of the induced birefringence seen by the probe beam could be accounted for by a crude averaging (which amounts to a reduction of φ by ~ 5% at $\alpha = 30°$). As shown in Fig. 1, the experimental data are in fair quantitative agreement with the theoretical curves, considering that no adjustable parameter was used in the calculation. The discrepancy may be due to the crude approximation used to take into account the scattering loss and the probe-beam averaging, but it is also because the theory assumes a free-energy density with no transverse variation.

We also measured, for the Freedericksz transition, I_{th} as a function of temperature, as shown in Fig. 3. We found that, more or less, I_{th} is proportional to the order parameter S. This is expected from the expression of I_{th} since $K_{33} \propto S^2$ and $\Delta\epsilon \propto S$.

From our results, we have estimated that for a pump intensity of 100 W/cm^2 at $\alpha = 30°$, the maximum reorientation angle of $\hat{n}$ is $\theta_m \sim 30°$, which corresponds to $\Delta n \sim 0.04$. This is compared to $\Delta n \sim 2 \times 10^{-11}$ in CS$_2$ at the same pump intensity. We therefore expect that nonlinear optical propagation effects in a nematic liquid crystal are easily observable.[4] We did, in fact, observe strong wavefront distortion of the pump-laser beam in our experiment. The nonlinearity can be further enhanced with a proper dc electric or magnetic bias field on the medium.[3,4] Novel nonlinear optical experiments arising from such large nonlinearities can be anticipated.

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[a] On leave from Erevan State University, Erevan, Armenia, USSR.

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[6] Because of the spatially varying anisotropy, the optical field varies in traversing the medium, but the magnitude of the Poynting vector remains constant assuming negligible loss. Not only the electricial energy, but also the magnetic energy, of the optical field participate in reorientation of the molecules. This is different from the dc case where the applied electric field is usually a constant and is solely responsible for the molecular reorientation.

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