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Transient laser-induced molecular reorientation and laser heating in a nematic liquid crystal

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Transient molecular reorientation induced in a liquid crystal by a giant laser pulse was observed for the first time in a nematic substance 4′-*n*-pentyl-4-cyanobiphenyl (5CB). Even though the response time of molecular reorientation was several orders of magnitude longer than the laser pulsewidth, the effect on the refractive indices was still large enough to be easily detectable by a Mach-Zehnder interferometer. Transient laser heating of the medium also affected the refractive indices, but with proper geometric and polarization arrangement, it could be decoupled from molecular reorientation and measured separately. Theoretical calculations using the Ericksen-Leslie theory for molecular reorientation in liquid crystals and the heat-diffusion theory for the laser heating effect were able to explain the experimental observations quantitatively.

I. INTRODUCTION

Liquid crystals are generally characterized by the strong correlation between molecules, which respond cooperatively to external perturbations. That strong molecular reorientation (or director reorientation) can be easily induced by a static electric or magnetic field is a well-known phenomenon.[1] The same effect induced by optical fields was, however, only studied recently.[2–6] Unusually large nonlinear optical effects based on the optical-field-induced molecular reorientation have been observed in nematic liquid-crystal films under the illumination of one or more cw laser beams.[3,4,6,7] In these cases, both the static and dynamical properties of this field-induced molecular motion are found to obey the Ericksen-Leslie continuum theory,[8] which describes the collective molecular reorientation by the rotation of a "director" (average molecular orientation).

The superiority of using lasers for material studies often lies in its spatial and temporal flexibilities, that is, the material can be selectively excited and probed in space and time. These qualities may allow us to elucidate fundamental material properties not accessible to conventional techniques. The location, dimension, direction, and duration of the material excitation can be readily controlled through adjustment of the beam spot, direction, polarization, and pulse width of the exciting laser field. The flexibilities can be further enhanced when two or more light waves are used to induce excitations. Such a technique, however, has not yet been fully explored in liquid-crystal research. Although the recent studies of optical-field-induced molecular reorientation in nematic liquid-crystal films have demonstrated the ability of the technique to resolve spatial variation of excitations, corresponding transient phenomena induced by pulsed optical fields have not yet been reported in the literature.[9] Because of the possibility of using lasers to induce excitations on a very short time scale, such studies could provide rare opportunities to test the applicability of the continuum theory in the extreme cases.

We report here the first observation of transient molecular reorientation induced in nematic 4′-*n*-pentyl-4-cyanobiphenyl (5CB) thin films by a 6-nsec laser pulse. Other than inducing the molecular reorientation, the intense laser pulse also heated the sample and thermally induced transient birefringence change. On the relatively short-time scale, this thermal effect was quite appreciable, and hence could prevent clean observation of the molecular reorientation. In our experiment, therefore, a static magnetic field was also applied to induce a stationary molecular reorientation in the nematic films: First, to enhance the transient optical-field-induced molecular reorientation, and second, to provide a condition for temporal discrimination against the thermal effect. The laser-induced changes in the 5CB sample were then probed by a weak cw laser beam with the help of a Mach-Zehnder interferometer. The detailed experimental arrangement is described in Sec. II.

The experimental results are given in Sec. III. We found that the magnitude of the transient molecular reorientation was proportional to the total energy of the laser pulse, and the relaxation time of such excitation was 9 orders of magnitude longer than the duration of the exciting laser pulse. The laser-induced thermal birefringence was also present. Its time dependence could be well described by the sum of two exponentials. The theory of these laser-induced effects is presented in Sec. IV. It is seen that the Ericksen-Leslie continuum theory, taking into account the finite beam size, agrees well with the experiment, and a simple heat-diffusion model can explain the temporal behavior of the thermal effect. More general discussion of the problem is given in Sec. V.

II. EXPERIMENTAL ARRANGEMENT

The 5CB used in this experiment was purchased from British Drug Houses, Inc. Its nematic phase occurs between 22.5 and 35.2 °C. Single-domain, homeotropic films were formed between two glass substrates which had been treated with *n*,*n*-dimethyl-*n*-octadecyl-3-aminopropyltrimethoxysilyl chloride (DMOAP).[10] The sample temperature was stabilized at 24.4±0.02 °C. Two samples, 130 and 190 μm thick, were studied. The thickness was controlled by Mylar spacers and its actual value was deduced from the observed threshold magnetic field (H_c) for Fréedericksz transition.[1]

The experimental arrangement is shown in Fig. 1(a). A 6-nsec laser pulse at 1.06 μm from a Q-switched Nd:YAG laser was focused with nearly normal incidence to an e^{-2} diameter of 500 μm on the sample to induce transient molecular reorientation in the sample. The resulting change of refractive indices was probed by a cw He-Ne laser beam at 632.8 nm, which was normally incident and closely overlapped with the pump laser beam at the sample. Spatial filtering was used at the output to ensure that only the central portion of the cylindrical region illuminated by the pump pulse was probed. The sample was placed in one arm of a Mach-Zehnder interferometer.[11] The induced refractive-index change in the sample resulted in a phase shift in the output interferogram which could be accurately measured. By displacing one mirror with a piezoelectric transducer, we could adjust the quiescent optical path difference between the two arms of the interferometer to an odd multiple of $\lambda/4$ to achieve the best signal-to-noise ratio. Vibrational and acoustic isolations were needed to ensure stability of the interferometer. The accuracy of our phase-shift measurements was better than 5×10^{-3} rad, corresponding to an average refractive-index change of $\lesssim 4\times10^{-6}$ in the samples. This is the best method we could think of that allows us to measure separately the refractive-index changes resulting from the laser-induced molecular reorientation and the laser-induced thermal effect mentioned earlier. The measurements were carried out in the following way.

A dc magnetic field H was applied along the direction parallel to the plane surfaces of the sample. With H larger than the critical field H_c for the Freedericksz transition, it could induce a molecular reorientation in the $\hat{H}$-$\hat{z}$ plane with a spatial variation $\theta_0(z)$ [see Fig. 1(b)]. A linearly polarized pump beam could, in addition, modify the refractive indices of the sample through both molecular reorientation and induced thermal effect; the latter was the result of laser heating of the medium. However, if the pump beam was polarized perpendicular to the $\hat{H}$-$\hat{z}$ plane, the molecular reorientation by the pump beam was negligibly small; only laser heating contributed to the refractive-index change, which could be probed by the probe beam polarized either parallel or perpendicular to the $\hat{H}$-$\hat{z}$ plane.

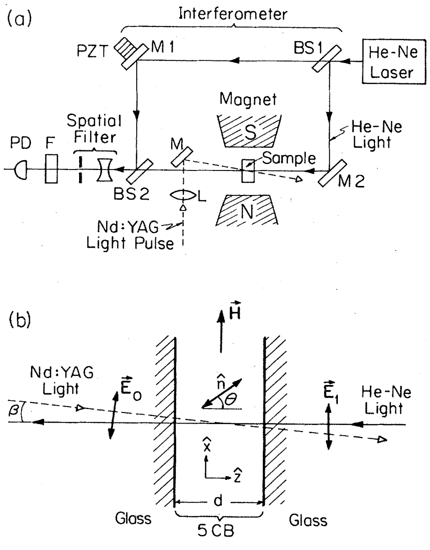

FIG. 1. (a) Experimental arrangement. BS1, BS2—50%-50% beam splitters; M, $M1$, $M2$—mirrors; L—lens; F—narrow-band filter (632.8 nm); PD—fast photodiode. (b) Experimental geometry. $\vec{\mathrm{H}}=H\hat{x}$, $\vec{\mathrm{E}}_1 \| \vec{\mathrm{H}}$. $\beta\simeq1°$.

If the pump beam was polarized in the $\hat{H}$-$\hat{z}$ plane, even a relatively weak pump intensity could induce a molecular reorientation in the $\hat{H}$-$\hat{z}$ plane, in addition to the laser heating effect. The probe beam polarized perpendicular to $\hat{H}$-$\hat{z}$ again monitored only the thermally induced refractive-index change, but when polarized parallel to $\hat{H}$, it should feel both the thermal effect and the molecular reorientation effect. The refractive-index changes due to both effects were actually fairly small; we can therefore write the laser-induced phase shift experienced by the probe beam in traversing the sample cell of thickness d as

$$\begin{aligned}
&\Delta\Phi(t)=\Delta\Phi_{\mathrm{MR}}+\Delta\Phi_{\mathrm{th}}\\
&\Delta\Phi_{\mathrm{MR}}=\frac{2\pi}{\lambda}\int_{-d/2}^{d/2}\left[\frac{\partial n_{\mathrm{eff}}}{\partial\theta}\right]_{\theta_0}\Delta\theta(z,t)dz \qquad (1)\\
&\Delta\Phi_{\mathrm{th}}=\frac{2\pi}{\lambda}\int_{-d/2}^{d/2}\left[\frac{\partial n_{\mathrm{eff}}}{\partial T}\right]_{\theta_0}\Delta T(z,t)dz\ ,
\end{aligned}$$

where $\Delta\Phi_{\mathrm{MR}}$ and $\Delta\Phi_{\mathrm{th}}$ are the induced phase shifts due to molecular reorientation $\Delta\theta$ and temperature rise ΔT, respectively, and $n_{\mathrm{eff}}(z,t)$ is the refractive index seen by the probe beam at position z and time t.

In the above case, the temperature rise across the sample should be uniform initially. We then have, at $t\sim0$,

$$\Delta\Phi_{\mathrm{th}}(t\sim0)\simeq\frac{2\pi}{\lambda}\Delta T_0\int_{-d/2}^{d/2}\left[\frac{\partial n_{\mathrm{eff}}}{\partial T}\right]_{\theta_0}dz\ . \qquad (2)$$

The effective refractive index n_{eff} can be written as[12]

$$n_{\mathrm{eff}}=\left[\frac{\cos^2\theta}{n_0^2}+\frac{\sin^2\theta}{n_e^2}\right]^{-1/2}, \qquad (3)$$

where n_0 and n_e are ordinary and extraordinary refractive indices, respectively, and hence

$$\frac{\partial n_{\mathrm{eff}}}{\partial T}=\frac{\partial n_{\mathrm{eff}}}{\partial n_0}\frac{\partial n_0}{\partial T}+\frac{\partial n_{\mathrm{eff}}}{\partial n_e}\frac{\partial n_e}{\partial T}\ . \qquad (4)$$

Since for 5CB, $\partial n_0/\partial T>0$ and $\partial n_e/\partial T<0$ (also, $\partial n_{\mathrm{eff}}/\partial n_0>0$ and $\partial n_{\mathrm{eff}}/\partial n_e>0$),[13] it was possible to choose a certain θ_0 such that $\Delta\Phi_{\mathrm{th}}(t\sim0)=0$. This then allowed us to obtain $\Delta\Phi_{\mathrm{MR}}(t\sim0)$ directly from the measured $\Delta\Phi(t\sim0)$.

As time went on, heat diffusion in the cell became significant, and the temperature across the cell was no longer uniform. The result was $\Delta\Phi_{\mathrm{th}}(t>0)\neq0$. However, in

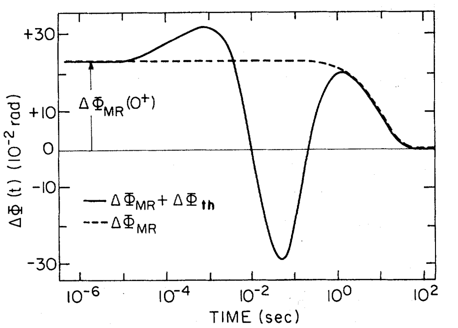

FIG. 2. Phase shift of the probe beam induced by a 27-mJ pulsed laser excitation at $t=0$. The dashed curve represents the molecular reorientation contribution obtained by subtracting the thermal contribution from the total phase shift (solid curve). The sample is a nematic 5CB film of 190 μm thickness.

liquid crystals, the relaxation time of the thermal effect is generally much shorter than the orientational relaxation time. Therefore, $\Delta\Phi_{MR}(t)$ could be obtained simply by subtracting $\Delta\Phi_{th}(t)$ from $\Delta\Phi(t)$.

III. EXPERIMENTAL RESULTS

Figure 2 displays a typical trace of the total phase shift, $\Delta\Phi(t)=\Delta\Phi_{MR}(t)+\Delta\Phi_{th}(t)$, on a logarithmic time scale detected with both the pump beam and the probe beam polarized in the $\hat{H}$-$\hat{z}$ plane. During the first 10 μsec after the pump laser excitation (which is essentially a δ function at $t=0$ on the graph), the thermal signal $\Delta\Phi_{th}$ is practically zero with our discrimination technique. A purely thermal signal $\Delta\Phi_{th}(t)$ was obtained using a pump pulse with polarization perpendicular to the $\hat{H}$-$\hat{z}$ plane. A typical trace is shown in Fig. 3. The molecular reorientation part, $\Delta\Phi_{MR}(t)$, of the total signal could then be singled out, as indicated by the dashed curve in Fig. 2.

From the measurements we found that after the pump pulse was over, $\Delta\Phi_{MR}(t)$ decayed exponentially with a time constant 9.2±0.9 sec for the 130-μm sample, or 23±4.8 sec for the 190-μm scale. The magnitude of $\Delta\Phi_{MR}$ at $t=0^+$ immediately after the pump pulse excitation was linearly proportional to the pump pulse energy, as shown in Fig. 4. The proportional constant was 4.84×10^{-3} rad/mJ for the 130-μm sample, or 7.88×10^{-3} rad/mJ for the 190-μm sample.

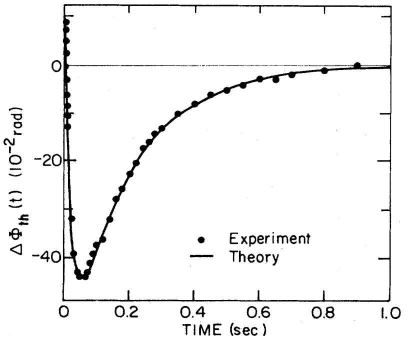

FIG. 3. An example of the observed phase shift as a function of time induced by pulsed laser heating in a 5CB film. The data were obtained for a 27-mJ laser pulse propagating in a sample 190 μm thick under the condition $\vec{E}_0 \perp \vec{H}$. The solid curve is described by the expression $\Delta\Phi_{th}=-ae^{-t/t_0}+be^{-t/t_1}$ with $t_0=190$ ms, $t_1=19$ ms, $a=0.67$, and $b=0.83$.

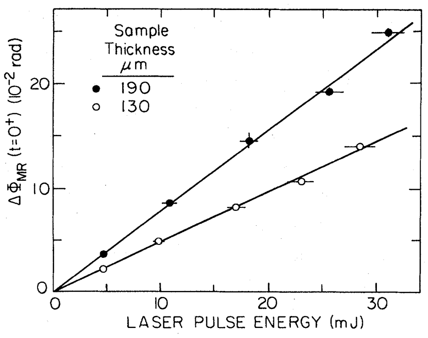

FIG. 4. Initial phase shift associated with molecular reorientation in 5CB films induced by pulsed laser excitation as a function of pulse energy.

The thermal signal $\Delta\Phi_{th}(t)$, in general, could be fit empirically by a function of the form $-a\exp(-t/t_0)+b\exp(-t/t_1)$ with $a,b>0$. An example is given in Fig. 3 where the solid curve is the empirical fit. While the coefficients a and b depend on the pump pulse energy, the time constants t_0 and t_1 are only functions of the sample thickness and characteristics of 5CB. We found for the 130-μm sample, $t_0=135$ msec and $t_1=13.5$ msec, and for the 190-μm sample, $t_0=190$ msec and $t_1=19$ msec. In both cases, the ratio of the two time constants was $t_0/t_1=10$.

IV. THEORY AND COMPARISON WITH EXPERIMENT

We neglect in the following theoretical derivation the coupling between laser heating and molecular reorientation. In general, with a finite pump beam traversing through a liquid-crystal cell, the induced temperature gradient in the medium can lead to a fluid flow which in turn orients the molecules through the flow-alignment mechanism.[14] In our case, because of the pumping geometry and the relatively large beam size, the thermally induced flow alignment is not significant. We can therefore treat the laser-induced molecular reorientation and the laser-induced thermal effect separately as two independent parts contributing to the observed phase shift $\Delta\Phi$.

A. Transient optical-field induced molecular reorientation

We use here the Ericksen-Leslie continuum theory[8] to describe the effect. The rotational motion of the director (i.e., molecular reorientation) is driven by the pump laser pulse, but it is also coupled with the translation motion (flow) of the fluid through viscosity. Thus, with a finite pump beam, a rigorous theoretical calculation would require the solution of a set of coupled three-dimensional nonlinear partial differential equations for the angle of director rotation θ and the fluid velocity $\vec{v}$. The mathematical complexity, however, can be greatly simplified through appropriate approximations.

If the laser-beam diameter is much larger than the sample thickness, and if only the core region is probed, then the pump beam can be approximated as an infinite plane wave. The rotation of the director can then be described by a one-dimensional equation of motion following the theory of Ericksen and Leslie. For the system in Fig. 1(b), the equation takes the form[15]

$$\rho_1\frac{\partial^2\theta}{\partial t^2}+\gamma_1\frac{\partial\theta}{\partial t}=[K_{33}+(K_{11}-K_{33})\sin^2\theta]\frac{\partial^2\theta}{\partial z^2}+\tfrac{1}{2}(K_{11}-K_{33})\sin(2\theta)\left[\frac{\partial\theta}{\partial z}\right]^2+\Gamma_H+\Gamma_{\text{opt}}+\tfrac{1}{2}[\gamma_1-\gamma_2\cos(2\theta)]\frac{\partial v_x}{\partial z}\ . \tag{5}$$

Here, $\theta(z,t)$ defines the orientation of the director $\hat{n}=(\cos\theta, 0, \sin\theta)$, and $v_x(z,t)$ is the x component of the flow velocity $\vec{v}=(v_x, 0, 0)$; ρ_1 is the "moment of inertia" per unit volume, which will be taken as zero in our calculation because its effect can be significant only during a subnanosecond pulse excitation; K_{11} and K_{33} are the Frank elastic constants, and γ_1 and γ_2 are the Leslie viscosity coefficients; finally, Γ_H and Γ_{opt} are the torques on the medium produced by the static magnetic field $\vec{\mathrm{H}}$ and the pulsed optical field $\vec{\mathrm{E}}$, respectively:[1]

$$\begin{aligned}\Gamma_H&=\Delta\chi(\hat{n}\cdot\vec{\mathrm{H}})\,|\hat{n}\times\vec{\mathrm{H}}|=\tfrac{1}{2}\Delta\chi H^2\sin(2\theta)\ ,\\ \Gamma_{\text{opt}}&=\frac{\Delta\epsilon}{4\pi}\langle(\hat{n}\cdot\vec{\mathrm{E}})\,|\hat{n}\times\vec{\mathrm{E}}|\,\rangle\ ,\end{aligned} \tag{6}$$

where $\Delta\chi=\chi_{\parallel}-\chi_{\perp}$ and $\Delta\epsilon=\epsilon_{\parallel}-\epsilon_{\perp}$, with $\chi_{\parallel}$ and $\chi_{\perp}$ being the diamagnetic susceptibilities parallel and perpendicular to the director, respectively, and $\epsilon_{\parallel}$ and $\epsilon_{\perp}$ being the corresponding optical dielectric constants.

The pump field at the input end is represented approximately by

$$\vec{\mathrm{E}}_0=\hat{x}\tfrac{1}{2}\mathscr{E}_0(t)\exp(ik_0z-i\omega t)+\text{c.c.} \tag{7}$$

Inside the liquid crystal, the optical axis of the medium, which is along $\hat{n}$, varies with z because of molecular reorientation. Accordingly, the $\hat{z}$ component of the $\vec{\mathrm{E}}$ field becomes nonvanishing in order to satisfy the requirement $\hat{z}\cdot\overleftrightarrow{\epsilon}\cdot\vec{\mathrm{E}}=0$; we have[12]

$$E_z/E_x=-\tfrac{1}{2}\Delta\epsilon\sin(2\theta)/(\epsilon_{\parallel}-\Delta\epsilon\sin^2\theta) \tag{8}$$

with $|E_x|=\mathscr{E}_0$. Absorption and scattering loss in the medium are negligible in our case.

The appropriate boundary conditions for Eq. (5) are $\theta(z=\pm d/2)=0$ and $v_x(z=\pm d/2)=0$, assuming strong anchoring of the homeotropically aligned molecules at the windows. If the optical-field-induced molecular reorientation is much weaker than the dc magnetic-field-induced reorientation, θ and v_x in Eq. (5) can be written approximately in the form

$$\begin{aligned}\theta(z,t)&=\theta_0(z)+\Delta\theta(z,t)\ ,\\ v_x(z,t)&=v_m(t)[\sin(qz)-2z/d]\ ,\end{aligned} \tag{9}$$

with

$$\theta_0(z)=\theta_m\cos(qz)\ ,$$

$$\Delta\theta(z,t)=\phi_m(t)\cos(qz)\ ,$$

where $q=\pi/d$ and $|\phi_m|\ll|\theta_m|$. The initial conditions for $\phi_m(t)$ and $v_m(t)$ are $\phi_m(0)=v_m(0)=0$. The dc field-induced director reorientation θ_0 can be determined from the static equation

$$[K_{33}+(K_{11}-K_{33})\sin^2\theta_0]\frac{\partial^2\theta_0}{\partial z^2}+\tfrac{1}{2}(K_{11}-K_{33})\sin(2\theta_0)\left[\frac{\partial\theta_0}{\partial z}\right]^2+\tfrac{1}{2}\Delta\chi H^2\sin(2\theta_0)=0\ , \tag{10}$$

which shows $\theta_0\neq0$ only if H is larger than the critical field $H_c=q(K_{33}/\Delta\chi)^{1/2}$ for the Fréedericksz transition.[1] With the known values[16] of $\Delta\chi$ and K's for 5CB, we find $\theta_m=1.14$ rad in the $d=130$-μm sample with $H=900$ Oe, and $\theta_m=1.12$ rad in the $d=190$-μm sample with $H=605$ Oe.

Equation (5) can now be transformed into an equation for the optical-field-induced director orientation $\phi_m(t)$. Since $|\phi_m|\ll|\theta_m|$, the equation can be linearized by keeping only terms up to the first order in ϕ_m and v_m. In reducing the equation further, the quantity Γ_{opt} is expanded into a power series of $\Delta\epsilon/\epsilon_{\parallel}$ ($=0.187$ for 5CB at 24.4 °C) and terms of orders higher than $(\Delta\epsilon/\epsilon_{\parallel})^2$ are neglected. The coefficients of the resultant equation are further expanded into power series of θ_m; with $\theta_m\sim1.1$ rad, these series all converge rapidly. Finally, by multiplying the equation by $\cos qz$ and integrating from $z=-d/2$ to $d/2$, it reduces to the form

$$\gamma_1\frac{\partial\phi_m}{\partial t}+\gamma_1\tau^{-1}\phi_m=f_{\text{opt}}+\gamma q v_m\ , \tag{11}$$

where

$$\begin{aligned}\gamma_1\tau^{-1}(t)&=q^2[K_{33}+(K_{11}-K_{33})G_1]+\Delta\chi H^2G_2+\frac{\Delta\epsilon}{8\pi}(\mathscr{E}_0(t))^2G_3\ ,\\ f_{\text{opt}}(t)&=\frac{\Delta\epsilon}{8\pi}(\mathscr{E}_0(t))^2G_4\ ,\\ \gamma&=\frac{1}{2}\left[1-\frac{8}{\pi^2}\right](\gamma_1-\gamma_2)+\gamma_2G_5\ .\end{aligned} \tag{12}$$

The G's are defined as

$$G_1=\sum_{n=0}^{\infty}(-1)^n\frac{2n+3}{4(n+1)}\frac{P_n}{(2n+1)!}(2\theta_m)^{2n+2}\ ,$$

$$G_2=\sum_{n=0}^{\infty}(-1)^{n+1}\frac{2P_n}{(2n)!}(2\theta_m)^{2n}\ ,$$

$$G_3=\sum_{n=0}^{\infty}(-1)^{n+1}\frac{2P_nQ_n}{(2n)!}(2\theta_m)^{2n}\,, \tag{13}$$

$$G_4=\sum_{n=0}^{\infty}(-1)^{n}\frac{P_nQ_n}{(2n+1)!}(2\theta_m)^{2n+1}\,,$$

$$G_5=\sum_{n=0}^{\infty}(-1)^{n+1}\left[P_n-\frac{2}{\pi^2}\frac{P_n^{-1}}{(n+1)}\right]\frac{(2\theta_m)^{2n}}{(2n)!}\,,$$

where

$$P_n=\frac{1}{2}\times\frac{3}{4}\times\frac{5}{6}\times\cdots\times\frac{2n+1}{2n+2}\,,$$

$$Q_n=1-2^{2n}(\Delta\epsilon/\epsilon_{\parallel})-(1+2^{2n+3}-3^{2n+2})(\Delta\epsilon/\epsilon_{\parallel})^2/16\,.$$

In Eq. (11) the γ term arises from the flow-orientation coupling, the magnitude of which depends on the static molecular orientation through the coefficient G_5. To find ϕ_m we need to know v_m. In general, the solution can only be obtained by solving Eq. (11) coupled with the equations of motion for the fluid flow. Physically, we expect the effect of the flow-orientation coupling to be as follows. First, the pump pulse rotates the director, and through the flow-orientation coupling, induces a flow. Then, even after the pulse is over, the inertia of the flow should drag the director reorientation further before it turns around and relaxes. Such behavior was, however, not observed in our experiment. It therefore suggests that in our case, the flow-orientation coupling may be rather weak. Indeed, with the previously mentioned values of θ_m in G_5, and using $\gamma_1=0.85$ P and $\gamma_2=-0.93$ P (Ref. 17) for 5CB (at 24.4 °C), we find $\gamma\simeq10^{-3}$ P for both the 130-μm and the 190-μm samples. This indicates a negligibly small flow-orientation coupling which agrees with the experimental observation. Thus, we can safely neglect the γ term in Eq. (11) and reduce it to the simpler form

$$\left[\frac{\partial}{\partial t}+\frac{1}{\tau}\right]\phi_m=f_{\text{opt}}/\gamma_1\,. \tag{14}$$

The response time τ given by Eq. (12) is a function of the pump intensity, but in our case, τ (of the order of 10–100 μsec during the pump pulse) is always much longer than the pump pulse width. The solution of Eq. (14) is then trivial. Immediately after the pump pulse is over, the optical-field-induced reorientation is simply

$$\phi_m(t=0^+)\cong\int_{-\infty}^{\infty}(f_{\text{opt}}/\gamma_1)dt\,, \tag{15}$$

and from Eqs. (1), (3), and (9), we have the corresponding phase shift

$$\Delta\Phi_{\text{MR}}(t=0^+)=\left[\frac{2\pi}{\lambda}\int_{-d/2}^{d/2}\left[\frac{\partial n_{\text{eff}}}{\partial\theta}\right]_{\theta_0}\cos(qz)dz\right]\times\int_{-\infty}^{\infty}(f_{\text{opt}}/\gamma_1)dt=AU_0\,, \tag{16}$$

where $U_0=\pi W^2(c/8\pi)\int_{-\infty}^{\infty}\mathscr{E}^2dt$ is the incoming pump pulse energy in air, W is the e^{-2} beam radius, and A is the proportional constant given by

$$A=\left[\frac{2\pi}{\lambda}\int_{-d/2}^{d/2}\left[\frac{\partial n_{\text{eff}}}{\partial\theta}\right]_{\theta_0}\cos(qz)dz\right]\times\frac{G_4\Delta\epsilon}{\pi W^2c\epsilon^{1/2}\gamma_1}\left[\frac{4\epsilon^{1/2}}{(1+\epsilon^{1/2})^2}\right] \tag{17}$$

with $4\epsilon^{1/2}/(1+\epsilon^{1/2})^2$ being the transmission coefficient at the air-sample boundary. The above result shows explicitly that $\Delta\Phi_{\text{MR}}(t=0^+)$ is directly proportional to the pump pulse energy, in agreement with the experimental observation depicted in Fig. 4. Using the measured value of $W=250\ \mu$m and the known quantities[13,17] for 5CB, we obtain from Eq. (17) $A=5.0\times10^{-3}$ rad/mJ for the 130-μm sample and $A=7.6\times10^{-3}$ rad/mJ for the 190-μm sample. These values are in excellent agreement with those deduced from the experimental data in Fig. 4. Note that if $\theta_m=0$, then $f_{\text{opt}}=0$ and $\phi_m(0^+)=0$, and we would see no optical-field-induced reorientation effect. This is why the application of a dc magnetic field to induce a finite θ_m is necessary in our experiment.

According to Eq. (14), after the pump pulse is over, ϕ_m should decay exponentially to zero,

$$\phi_m(t)=\phi_m(0^+)\exp(-t/\tau_0)$$

with a time constant $\tau_0=\tau\ (\mathscr{E}_0^2=0)$. The exponential decay is indeed what was observed, as shown in Fig. 2. The values of τ_0 calculated from Eq. (12) are 13 sec for the 130-μm sample and 30 sec for the 190-μm sample. Both, however, are significantly larger than the experimental values given in Sec. III.

A better prediction for τ_0 can be obtained if the finite beam size is taken into account. The equation of motion for ϕ_m should now include a term proportional to the second derivative of ϕ_m in the transverse direction.[8] For $t>0^+$ (after the pump pulse is over), it has the form

$$\gamma_1\frac{\partial\phi_m}{\partial t}+(\gamma_1/\tau_0)\phi_m=K\nabla_\rho^2\phi_m\,, \tag{18}$$

where $\nabla_\rho^2=\partial^2/\partial\rho^2+(1/\rho)(\partial/\partial\rho)$, ρ is the polar coordinate in the transverse plane, and for simplicity we have assumed $K_{11}=K_{22}=K_{33}=K$ for the $\nabla_\rho^2\phi_m$ term. The general solution of Eq. (18) with the simplified boundary condition $\phi_m(\rho\geq W)=0$ should have the expression

$$\phi_m(\rho,t)=\sum_{n=1}^{\infty}a_nJ_0(S_n\rho/W)\exp(-t/\tau_n) \tag{19}$$

with

$$\tau_n=[\tau_0^{-1}+(K/\gamma_1)(S_n/W)^2]^{-1}\,, \tag{20}$$

where J_0 is the Bessel function of order zero, S_n is the nth root of J_0, and a_n's are coefficients. We assume that the $\exp(-t/\tau_1)$ term dominates and the solution takes the simple form

$$\phi_m(\rho,t)=\begin{cases}\phi_m(0,0^+)J_0(S_1\rho/W)\exp(-t/\tau_1) & \text{for } \rho\leq W\\ 0 \ \text{ for } \rho>W\end{cases} \tag{21}$$

with $S_1=2.405$. The relaxation time τ_1 is now given by Eq. (20). As expected, the relaxation time decreases with the beam size. Taking $K\simeq K_{33}=0.8\times10^{-6}$ dyn,[16]

$W=250$ μm and the value of τ_0 obtained earlier, we find $\tau_1=12$ sec for the 130-μm sample, and $\tau_1=24$ sec for the 190-μm sample, in fair agreement with the measured values.

B. Laser-induced thermal effect

Residual absorption at the pump laser frequency can lead to heating of the liquid-crystal medium. Although the absorption in 5CB at 1.06 μm is very weak, it can still result in a detectable temperature change. Right after the pump pulse is over, the temperature rise ΔT has a distribution nearly constant along z, but proportional to the pulse energy distribution transversely. Then, through heat diffusion, the temperature variation should relax back to the uniform distribution at equilibrium. At any given instant, a weighted integration of the temperature change $\Delta T(\rho,z)$ along z is reflected in the phase shift $\Delta\Phi_{\text{th}}$ described in Eq. (1).

A general analysis of the variation of $\Delta T(\rho,z)$ with time is quite complicated. However, since the pump beam diameter was much larger than the sample thickness in our experiment, we can use the approximation of neglecting heat diffusion in the transverse plane. The temperature change ΔT now obeys the one-dimensional heat-diffusion equation

$$\frac{\partial}{\partial t}\Delta T=\alpha\frac{\partial^2}{\partial z^2}\Delta T \ . \tag{22}$$

We have assumed here that the thermal diffusivity α is independent of z, that is, we have neglected the dependence of α on the director orientation in the medium. With the initial condition $\Delta T(t=0^+)=\Delta T_0$ throughout the sample and the boundary condition $\Delta T(z=\pm d/2,\ t)=0$, we readily find the solution of Eq. (22) as[18]

$$\Delta T(z,t)=\frac{4\Delta T_0}{\pi}\sum_{n=0}^{\infty}\frac{(-1)^n}{2n+1}\cos[(2n+1)qz]e^{-t/t_n} \ , \tag{23}$$

where $t_n=1/\alpha(2n+1)^2q^2$ and $q=\pi/d$. The corresponding phase shift $\Delta\Phi_{\text{th}}$ due to ΔT can then be obtained from Eq. (1):

$$\Delta\Phi_{\text{th}}(t)=\frac{8\Delta T_0}{\lambda}\sum_{n=0}^{\infty}\frac{(-1)^n}{2n+1}e^{-t/t_n}\times\int_{-d/2}^{d/2}\left[\frac{\partial n_{\text{eff}}}{\partial T}\right]_{\theta_0}\times\cos[(2n+1)qz]dz \tag{24}$$

with $\theta_0=\theta_m\cos qz$.

As we mentioned in Sec. II, it is possible to choose a value of θ_m, by adjusting the magnetic field H, to make $\Delta\Phi_{\text{th}}(0^+)=0$. In principle, this value of θ_m can be estimated from Eq. (2) using Eq. (3) and is independent of the sample thickness. However, the large uncertainty in the known ratio $(\partial n_e/\partial T)/(\partial n_0/\partial T)$ allows only a rather crude estimate. Our experimental result yielded $\theta_m\simeq1.1$ (corresponding to $H/H_c\simeq1.35$) for the two samples of different thickness. Even with $\Delta\Phi(0^+)=0$, Eq. (24) shows that at later times, $\Delta\Phi_{\text{th}}(t)$ would still become finite. For $\Delta\Phi_{\text{th}}(t)$ in the relatively long-time regime, we need to keep only the two terms with the longest relaxation times in Eq. (24). By expanding $(\partial n_{\text{eff}}/\partial T)_{\theta_0}$ into a series up to the θ_0^2 term and evaluating the integral in Eq. (24), we find

$$\Delta\Phi_{\text{th}}(t)\cong\text{const}\times[-\exp(-t/t_0)+\tfrac{3}{5}\exp(-t/t_1)] \ . \tag{25}$$

The result here shows that $t_0=9t_1=d^2/\pi^2\alpha$, and the ratio of the amplitudes of the two terms is $-\frac{3}{5}$, independent of the pump pulse energy and the sample thickness.

Our experimental data on $\Delta\Phi_{\text{th}}(t)$ can indeed be fit approximately by the linear combination of two exponentials with proper signs on their amplitudes. The ratio of the two relaxation times was found to be $t_0/t_1\simeq10$ for the two samples of different thickness, in fair agreement with the theoretical prediction. By using $t_0=d^2/\pi^2\alpha$, the value of α deduced from the measured t_0 is, however, different for the two samples of different thickness. It is 1.26×10^{-4} cm^2/sec for the 130-μm sample, or 1.93×10^{-4} cm^2/sec for the 190-μm sample. These values of α are in the correct range of thermal diffusivities of liquid crystals. The dependence of α on sample thickness can be qualitatively attributed to the effect of transverse heat diffusion in the sample cell which we have neglected in our analysis. In a thicker cell, the transverse heat diffusion is more important, and therefore, the effective one-dimensional thermal diffusivity α should be larger. The values of α deduced above roughly scale with the sample thickness. A more serious discrepancy between theory and experiment is in the relative amplitude of the two exponentials of $\Delta\Phi_{\text{th}}(t)$. The theoretical value is $-\frac{3}{5}$, while the experimental value is -1.2 in Fig. 3. This discrepancy presumably also arises from the approximations we have made in the analysis.

V. DISCUSSION

We have studied in this work transient refractive-index change in a nematic liquid crystal 5CB arising from the pulsed-laser-induced molecular reorientation and thermal effect. The same mechanisms actually also lead to a change in optical birefringence. This is because the ordinary and extraordinary refractive indices of the medium have different functional dependences on the director orientation angle and on the temperature. In the steady state, the induced refractive-index change from molecular reorientation in 5CB is larger than that from the thermal effect by about 1 order of magnitude.[7] The former, however, has a much slower response time than the latter. Consequently, when pulsed laser excitation is used, the thermal effect can appear dominating the molecular reorientation effect. The two effects often can be separated by measurements with different pump polarizations; this is because the thermal effect is generally independent of the pump polarization.

The change in refractive index induced in a nematic liquid crystal by a steady-state optical field is known to be extremely large, with $\Delta n\sim0.05$ for a pump field of ~100 W/cm^2.[5] However, because of the very slow time response of the correlated molecular orientational motion, the amount of Δn that can be induced by a $\sim$10-nsec laser pulse through molecular reorientation is rather small. It is $\sim(T_p/\tau)$ times smaller than the steady-state Δn, assuming equal cw and pulsed beam intensity, where T_p is the pulse width and τ is the response time of molecular reorientation ($\tau \sim 10$ sec for 100-W/cm^2 pump intensity). Thus, to attain an average Δn of 10^{-4} in a 100-μm cell (corresponding to a phase shift of $\Delta\Phi_{\rm MR} \sim 0.1$ rad) by a 10-nsec pulsed laser excitation, we would need a laser intensity of $\sim 2\times 10^8$ W/cm^2. This is indeed roughly the same order of magnitude as the one we had to use in our experiment. Since $\Delta\Phi_{\rm MR} \sim 0.1$ rad is readily detectable, we can then use the time variation of $\Delta\Phi_{\rm MR}$ to study the dynamics of molecular reorientation. We found from our measurement that it obeys the continuum theory of Ericksen and Leslie very well.

The Ericksen-Leslie theory has been widely used in discussing the dynamic phenomena of the nematic state. However, the inertia term in the equation is always neglected. While this approximation should be valid for long-pulse excitation, it may break down if the exciting pulse is sufficiently short. In our experiment with 6-nsec exciting laser pulses, there was no evidence of the inertia term being important. This sets a limit on the magnitude of the moment of inertia ρ_1 in Eq. (5) to be less than $\sim 10^{-8}$ g cm^{-1}. If we assume that ρ_1 corresponds to the moment of inertia of a bunch of oriented molecules within a volume of the size of the correlation length ($\sim$100 Å) of the liquid crystal, then we expect $\rho_1 \sim 10^{-12}$ g cm^{-1}. The effect of such a "moment of inertia" on molecular reorientation could be measured by picosecond laser pulses.

In our measurements, the situation was greatly simplified by using a geometric arrangement such that the flow-orientation coupling was negligible. As a result, the effect of fluid flow could be disregarded, and the molecular reorientation part and the thermal part could be decoupled. In general, however, the flow-orientation coupling can be important, and it can lead to interesting but complicated dynamic phenomena in liquid crystals. For example, laser heating resulting in a strong local temperature gradient can give rise to a convection flow in the fluid, which then orients the molecules. The back reaction of the molecular reorientation should in turn affect the flow. That laser heating can be well localized and switched on or off very rapidly should allow us to excite and investigate certain dynamic phenomena in a fluid that cannot be studied with other techniques.

More generally, acoustic waves can also be generated in a liquid crystal by pulsed laser excitation. They arise from both the direct electrostrictive coupling between the laser field and the acoustic waves and the indirect excitation via thermal expansion caused by laser heating. Both mechanisms rely on the existence of an intensity gradient in the laser-beam profile. For a larger beam cross section, the acoustic waves generated are less intense. In our experiment, the pump beam size was sufficiently large so that the effect of acoustic-wave generation was negligible. It is, however, possible to generate acoustic waves in a liquid crystal more effectively by crossing two laser pulses at an angle and inducing a transient density grating in the sample. The resulting acoustic waves and their properties can be probed and studied by a third laser pulse.

Generally speaking, excitation of a medium by short laser pulses can be used to study dynamic properties of the medium over a very wide time range. Here, we have shown that nanosecond-pulse excitation can yield information about the dynamics of molecular reorientation on the $\sim$10-sec time scale, and thermal effect on the 10–100-msec time scale. The power of this technique lies in the fact that a single δ-function-like laser pulse may induce a number of fundamental excitation modes of vastly different time constants. Consider, for example, molecular reorientation coupled with flow induced by a picosecond laser pulse in a liquid crystal. It can be shown that, aside from the thermal effect, the transient behavior will manifest itself with three characteristic time constants:

$$\tau_i \simeq \rho_1/\gamma_1 \;,$$
$$\tau_f \sim \rho/\bar{\gamma} q^2 \;,$$
$$\tau_s \sim \bar{\gamma}/\bar{K} q^2 \;,$$

where ρ_1, γ_1, and q have the meaning defined earlier, and $\bar{\gamma}$ and $\bar{K}$ are the effective viscosity coefficient and elastic constant, respectively, and ρ is the density of the fluid. The time constants τ_f and τ_s are associated with the so-called "fast" and "slow" modes[8] in a liquid crystal and dominated by the damping of flow and the relaxation of molecular reorientation, respectively. The molecular reorientation observed in our experiment corresponds to the slow mode. The time constant τ_i, which is independent of pumping power and geometry, is associated with a cooperative librational mode. If we take $\rho_1 \sim 10^{-12}$ g cm^{-1}, $\bar{\gamma} \sim 1$ P, $\bar{K} \sim 10^{-6}$ dyn, $\rho \simeq 1$ g cm^{-3}, and $q^{-2} \sim 10^{-5}$ cm^2 (for $d \sim 100\ \mu$m), then $\tau_i \sim 1$ psec, $\tau_f \sim 10\ \mu$sec, and $\tau_s \sim 10$ sec. These three modes excited by the same picosecond laser pulse will therefore have very different time responses, and can, in principle, be studied by a probe laser in different time regimes.

VI. CONCLUSION

We have presented here the first observation of transient molecular reorientation induced in a liquid crystal by a Q-switched laser pulse. The response time of molecular reorientation in the nematic phase is of the order of 10–100 μsec. Although this is 10^3–10^4 times longer than the duration of the laser pulse, transient molecular reorientation is still strong enough to yield an easily detectable phase shift in the probe beam. Residual absorption and subsequent very rapid radiationless conversion into heat can result in a temperature rise in the medium which decays via heat diffusion with relaxation times in the 10–200 msec range. The temperature rise also induces a refractive-index change in the medium and hence a phase shift in the probe beam. This thermal effect and the molecular reorientation are initiated simultaneously by the pulsed laser excitation. They are in general coupled via fluid flow. In our experiment, an initial orientation of molecules by a dc magnetic field allowed us to decouple the two. Then, by using different pump polarizations, the two effects could be separately measured. Theoretical calculation following the continuum theory of Ericksen and Leslie for liquid crystals showed excellent agreement with the experimental results on molecular reorientation. On the other hand, a simple theory of heat diffusion explained the thermal effect fairly well.

The experiment reported in this work demonstrates the possibility of using a short laser pulse (or pulses) to simultaneously excite various dynamic modes in a medium and subsequently probing the responses of these modes which could vary on vastly different time scales. The technique allows the studies of dynamic properties of several material excitations by a single measurement in the time domain. This is in contrast to the usual spectroscopic probing technique. Aside from being indirect, the latter has the disadvantage of requiring several different analyzing systems in order to study material excitations with very different time responses.

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