Laser-induced thermal, orientational, and density nonlinear optical effects in nematic liquid crystals

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We present a theoretical analysis of transient diffractions in nematic liquid crystals, where the nonlinear optical response is due to laser-induced changes in the temperature, density, and director axis reorientation. Our solutions of the field-induced reorientation equation and the coupled density and thermal conduction equations provide a quantitative theoretical model for the experimentally observed grating diffractions, oscillations, and relaxation phenomena.

Third-order nonlinear optical effects in nematic-liquids-crystal thin films have been studied for several years.[1-6] Three basic physical mechanisms are involved. One is the optically induced molecular reorientation effect, the others are thermal and density effects. The steady-state behavior of these nonlinear effects has been well explored both in the theory aspect and the experimental aspect.[1-6] On the other hand, the transient processes have been explored mainly[5] experimentally. In this paper we present a quantitative analysis of these three mechanisms by solving the field-induced director axis reorientation equation and the coupled hydrodynamic equations for the temperature and density fluctuations.

Figure 1 shows the basic experiment configuration. Two incident laser pulses b_1 and b_2 with proper polarization directions intersect each other at the location of a thin nematic-liquid-crystal film. These two beams induce a refractive-index grating by thermal, density, and reorientation effects. The refractive-index grating will diffract a probe light beam, which is a geometry used in recent studies.[5,6] The input beams could also self-diffract from this index grating, producing s_1 and s_2. We will consider the general situation where all three mechanisms—thermal, density, and reorientation effects—are involved (cf. Refs. 1 and 2), and the incident laser pulses are of duration much shorter than the shortest characteristic relaxation times involved here. In the present context the acoustic decay time constant is the shortest, and is on the order of 100 ns or so (cf. Refs. 5 and 6), i.e., one should use nanosecond laser pulses. For simplicity, the incident lasers are assumed of plane-wave form and linearly polarized. By using purely extraordinary- or ordinary-ray input, and probing the resulting index changes by parallel- or cross-polarized probe beam, and/or monitoring their (the index changes') decay behavior, one could distinguish the relative contribution from these orientational, thermal, and density contributions.

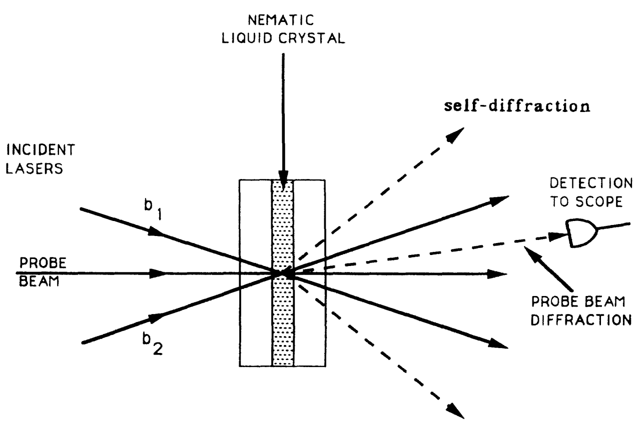

FIG. 1. Schematic diagram of two incident coherent laser beams b_1 and b_2 interacting with a nematic film producing self-diffractions and diffractions of a probe beam.

Inside the liquid crystal, the total optical intensity I_{op} is given by

$$I_{op} = \frac{cn}{2\pi}(|E_1|^2 + |E_2|^2) + \frac{cn}{2\pi}[2|E_1 E_2|\cos(\mathbf{k}_1 - \mathbf{k}_2)\cdot\mathbf{r}] , \quad (1)$$

where $|E_1|, |E_2|$ are the amplitudes, and $\mathbf{k}_1$ and $\mathbf{k}_2$ are the wave vectors of the pump and probe beam, respectively.

In general, the optically induced reorientation $\Delta\theta$, the density fluctuation $\Delta\rho$, and the temperature fluctuation ΔT, are functions of the total optical intensity. In accordance with the impressed intensity given in (1), which consists of a spatially static part (in parentheses) and an oscillatory part (in square brackets), these induced changes are also made up of a spatially static and oscillatory component. For the experiment as depicted in Fig. 1, the diffractions from the probe beam (and also the pump beams' self-diffraction) are sensitive only to the oscillatory part of the induced index changes. In the following calculation, therefore, the optical intensity that appears as a source term in the equations describing $\Delta\rho$,

$\Delta\theta$, and ΔT is understood to be the spatially oscillatory component, i.e.,

$$\frac{C}{2\pi}(2E_1E_2\cos\mathbf{q}\cdot\mathbf{r})=I=\frac{cn}{2\pi}E_{\rm op}^2\ , \tag{2}$$

where

$$\mathbf{q}=\mathbf{k}_1-\mathbf{k}_2\ ,\quad |q|=2k\sin\left[\frac{\theta}{2}\right]\ , \tag{3}$$

k is the magnitude of $\mathbf{k}_1$ and $\mathbf{k}_2$ and θ is the wave-mixing angle. For simplicity, we let $|E_1|=|E_2|=E_{\rm op}$.

From standardized dynamic grating theory,[7] the diffraction of the probe beam is proportional to $(\Delta n)^2$, where Δn is the amplitude of the total induced refractive-index changes. The problem under study here is to find Δn of liquid crystal as a function of time, and the relative contributions from these three physical mechanisms.

We now write

$$\Delta n=\Delta n^r+\Delta n^t(T,\rho)\ , \tag{4}$$

where Δn^r is the refractive-index change due to the reorientation effect and $\Delta n^t(T,\rho)$ is that due to the coupled thermal and density fluctuations.

Δn^r follows the Debye relaxation equation, cf. Ref. 3,

$$\left[\frac{\partial}{\partial t}+\frac{1}{\tau_r}\right]\Delta n^r(t)=KE_{\rm op}^2\ , \tag{5}$$

where τ_r is the relaxation time of molecule orientation for the first-order approximation. K is a coupling constant and depends on experiment configuration and can be calculated by the usual theory of optical field-induced director axis reorientation theory.[3]

Take the instant that the two incident lasers reach the liquid-crystal film as $t=0$. Assuming that the laser is of constant intensity, the reorientational refractive-index change is given by

$$\Delta n^r(t)=\frac{1}{\tau_r}\int_0^t KE_{\rm op}^2\exp\left[-\frac{(t-\eta)}{\tau_r}\right]d\eta =KE_{\rm op}^2\tau_r\left[1-\exp\left[\frac{-t}{\tau_r}\right]\right]\ . \tag{6}$$

We note here that if the input laser pulse duration τ_p is short (i.e., $\tau_p \ll \tau_r$), $\Delta n^r(t)\sim KE_{\rm op}^2\tau_p$ (which is the effect reported in Ref. 3). The refractive-index change $\Delta n^t(\rho,T)$ may be expressed in the form

$$\Delta n^t=\left[\frac{\delta n}{\delta T}\right]_\rho\Delta T+\left[\frac{\delta n}{\delta\rho}\right]_T\Delta\rho\ , \tag{7}$$

where T and ρ stand for the temperature and the density of the liquid crystal, respectively. $(\delta n/\delta T)_\rho$ and $(\delta n/\delta\rho)_T$ are regarded approximately as constants for small temperature changes.[8] $\Delta n(\rho,T)$ obeys the coupled hydrodynamic equations.[9]

Following the derivation of Hoffman[10] and Batra, Enns, and Pohl[11] we have, for $\Delta\rho$ and ΔT,

$$-\frac{\partial^2}{\partial t^2}(\Delta\rho)+\frac{v^2}{\gamma}\nabla^2(\Delta\rho)+\frac{v^2\beta_T\rho_0}{\gamma}\nabla^2(\Delta T)+\frac{\eta}{\rho_0}\frac{\partial}{\partial t}\nabla^2(\Delta\rho) =\frac{\gamma^e}{8\pi}\nabla^2(E_{\rm op}^2)\ , \tag{8}$$

$$\rho_0c_p\frac{\partial}{\partial t}(\Delta T)-\lambda_T\nabla^2(\Delta T)=\frac{\alpha nc}{8\pi}E_{\rm op}^2\ , \tag{9}$$

where ρ_0 is the density of the liquid crystal. c_p and c_v are the specific heats, λ_T is the thermal conductivity, η is the viscosity, v is the speed of sound, γ^e is the electrical elastic coefficient, and $\gamma\equiv c_p/c_v\sim 1$.

Substituting $\nabla^2E^2=-2E_{\rm op}^2q^2\cos(\mathbf{q}\cdot\mathbf{r})$, Eqs. (8) and (9) become

$$-\frac{\partial^2}{\partial t^2}(\Delta\rho)+v^2\nabla^2(\Delta\rho)+v^2\beta_T\rho_0\nabla^2(\Delta T)+\frac{\eta}{\rho_0}\frac{\partial}{\partial t}\nabla^2(\Delta\rho) =-\frac{\gamma^e}{4\pi}|E_{\rm op}^2|q^2\cos(\mathbf{q}\cdot\mathbf{r})\ , \tag{8'}$$

$$\rho_0c_p\left[\frac{\partial}{\partial t}\right](\Delta T)-\lambda_T\nabla^2(\Delta T)=\frac{\alpha nc}{4\pi}|E_{\rm op}^2|\cos(\mathbf{q}\cdot\mathbf{r})\ . \tag{9'}$$

We assume that ΔT and $\Delta\rho$ have the same space period as the light interference field, i.e.,

$$\Delta T=W(t)|E_{\rm op}^2|\cos(\mathbf{q}\cdot\mathbf{r})\ , \tag{10}$$

$$\Delta\rho=D(t)|E_{\rm op}^2|\cos(\mathbf{q}\cdot\mathbf{r})\ . \tag{11}$$

Substituting (10) and (11) into (8′) and (9′) yields

$$\rho_0c_p\frac{\partial}{\partial t}W(t)+\lambda_Tq^2W(t)=\frac{\alpha nc}{4\pi}\ , \tag{12}$$

$$-\frac{\partial^2}{\partial t^2}D(t)-v^2q^2D(t)-v^2\beta_T\rho_0q^2W(t)-\frac{\eta}{\rho_0}\frac{\partial}{\partial t}D(t)q^2 =-\frac{\gamma^e}{4\pi}q^2\ . \tag{13}$$

Since $n=n_0+\Delta n^r+\Delta n^t(\rho,T)$ in Eq. (12), it is difficult to find the exact solution. If $\Delta n^r,\Delta n^t(\rho,T)$ are $\ll n_0$, which is usually the case, we can use the iteration method. We define $W^{(0)}(t)$ by taking $n=n_0+\Delta n^r$ and Eq. (12) gives

$$\frac{\partial}{\partial t}W^{(0)}(t)+\Gamma_RW^{(0)}(t)=\frac{\alpha c}{4\pi\rho_0c_p}\left\{n_0+K|E_{\rm op}|^2\tau_r\left[1-\exp\left[-\frac{t}{\tau_r}\right]\right]\right\}\ , \tag{14}$$

where $\Gamma_R=\lambda_Tq^2/\rho_0c_p$. Using the initial condition $W^{(0)}(0)=0$, $W^0(t)$ is solved from Eq. (14) to yield

$$W^{(0)}(t)=\frac{\alpha c}{4\pi\rho_0c_p\Gamma_R}(n_0+K|E_{\rm op}|^2\tau_r)[1-\exp(-\Gamma_Rt)]+\frac{\alpha cKE_{\rm op}^2\tau_r^2}{4\pi\rho_0c_p(\Gamma_R\tau_r-1)}\left[\exp(-\Gamma_Rt)-\exp\left[-\frac{t}{\tau_r}\right]\right]\ . \tag{15}$$

Substitute Eq. (15) into Eq. (13), then

$$\frac{\partial^2}{\partial t^2}D^{(0)}(t)+\Gamma_B\frac{\partial}{\partial t}D^{(0)}(t)+q^2v^2D^{(0)}(t)=\frac{\gamma^e}{4\pi}q^2-v^2\beta_T\rho_0q^2W^{(0)}(t)\ , \quad (16)$$

where $\Gamma_B=\eta q^2/2\rho_0$.

The homogeneous solution of Eq. (16) is

$$D_h^{(0)}(t)=A\exp(-\Gamma_B t)\cos(\Omega t+\Phi)\ , \quad (17)$$

where $\Omega=(q^2v^2-\Gamma_B^2)^{1/2}$, and Φ is a phase factor for the density (acoustic) wave.

Considering the values (in mks units[1,8]) $\rho_0\sim10^3$, $v\sim1540$, $\eta\sim3\times10^{-2}$, $k\sim1.2\times10^7$, $\theta\sim3\times10^{-2}$, and $q=k\theta$, we conclude that $q^2v^2\gg\Gamma_B^2$ and $\Omega=qv$.

Assume that

$$D^{(0)}(t)=D_h^{(0)}(t)+P+S\exp(-\Gamma_R t)+R\exp\left[-\frac{t}{\tau_r}\right],$$

and substitute it into Eq. (16) and note that $\Gamma_B/\Gamma_R=\eta c_p/2\lambda_T\gg1$ ($\eta\sim3\times10^{-2}$, $c_p\sim10^3$, $\lambda_T\sim2\times10^{-2}$). We have

$$P=\frac{\gamma^e}{4\pi v^2}-\frac{\beta_T\alpha c}{4\pi\rho_0c_p\Gamma_R}(n_0+K|E_{op}|^2\tau_r)\ ,$$

$$S=\frac{\beta_T\alpha c}{4\pi c_p\Gamma_R}(n_0+K|E_{op}^2|\tau_r)-\frac{\beta_T\alpha cK\tau_r^2|E_{op}^2|}{4\pi c_p(\Gamma_R\tau_r-1)}\ ,$$

$$R=\frac{\beta_T\alpha cK\tau_r^2|E_{op}^2|}{4\pi c_p(\Gamma_R\tau_r-1)}\ ,$$

with the initial condition $D^{(0)}(0)=0$, $A\cos\Phi+P+S+R=0$, then

$$A=-(P+S+R)=-\frac{\gamma^e}{4\pi v^2}$$

(take $\cos\Phi=1$ for simplicity). Finally,

$$D^{(0)}(t)=\frac{\gamma^e}{4\pi v^2}[1-\exp(-\Gamma_B t)\cos\Omega t]-\frac{\beta_T\alpha c}{4\pi c_p\Gamma_R}(n_0+K|E_{op}|^2\tau_r)[1-\exp(-\Gamma_R t)]-\frac{\beta_T\alpha cK\tau_r^2|E_{op}^2|}{4\pi c_p(\Gamma_R\tau_r-1)}\left[\exp(-\Gamma_R t)-\exp\left[\frac{-t}{\tau_r}\right]\right]. \quad (18)$$

Collecting all the contributions from Δn^r and $\Delta n(\rho,T)$, the total change in refractive index is therefore given by

$$\Delta n=|E_{op}|^2\left\{K\tau_r\left[1-\exp\left[-\frac{t}{\tau_r}\right]\right]+c_1[1-\exp(-\Gamma_R t)]+c_2\left[\exp(-\Gamma_R t)-\exp\left[-\frac{t}{\tau_r}\right]\right]+c_3[1-\exp(-\Gamma_B t)\cos(\Omega t)]\right\}, \quad (19)$$

where

$$c_1=\left[\frac{\delta n}{\delta T}-\beta_T\rho_0\frac{\delta n}{\delta\rho}\right]\frac{\alpha c}{4\pi\rho_0c_p\Gamma_R}(n_0+K|E_{op}|^2\tau_r)\ , \quad (20)$$

$$c_2=\left[\frac{\delta n}{\delta T}-\beta_T\rho_0\frac{\delta n}{\delta\rho}\right]\frac{\alpha cK\tau_r^2|E_{op}|^2}{4\pi\rho_0c_p(\Gamma_R\tau_r-1)}\ , \quad (21)$$

$$c_3=\frac{\delta n}{\delta\rho}\frac{\gamma^e}{4\pi v^2}\ . \quad (22)$$

The first term in the large square brackets on the right-hand side of Eq. (19) is the contribution that comes from the optically induced reorientational effect. The second term with a coefficient c_1 may be termed the temperature effect as it comes from $W^0(t)$, and the last term with a coefficient c_3 is the density effect, with respective relaxation time constants τ_r, Γ_r^{-1}, and Γ_B^{-1}. The second term with a coefficient c_2 arises as a result of the coupling of the reorientation with the temperature-density effect. Notice that

$$c_2/c_1=\frac{KE_{op}^2\tau_r}{n_0}\left[\frac{\Gamma_R}{\Gamma_R-\tau_r^{-1}}\right];$$

since $KE_{op}^2\tau_r$, the maximum magnitude of the optically induced refractive-index change [cf. Eq. (6)], is much smaller than n_0, and also $\Gamma_R\gg\tau_r^{-1}$, in general, the contribution from c_2 is small compared to c_1.

The c_3 term shows that there is a short oscillation process which is due to the density (acoustic) wave. The frequency is $\Omega=qv$ and the damping coefficient $\Gamma_B=\eta q^2/2\rho_0$. Because the value of c_3 is small, it is only under conditions where the incident beam is strong and the time resolving of the detector is high that the oscillation phenomenon may be detected. Experimental observation of these density effects has been reported in Refs. 5 and 6, where Γ_B was measured to be on the order of 10^{-7} sec.

The terms related to $\Gamma_R=\lambda_Tq^2/\rho_0c_p$ are nonlinear thermal grating effect terms. The value of Γ_R is dependent on the grating constant and/or the film thickness. Experimentally, they are measured to be on the order of 50–100 μs (Refs. 5 and 6) or milliseconds (Ref. 4) depending on the characteristic diffusion lengths involved.

τ_r depends on the property of the liquid crystal and the thickness of the sample. Its value typically is on the order of milliseconds to seconds. With the use of nanosecond laser pulses ($\tau_{laser}\ll\tau_r$) [cf. Eq. (6), $\Delta n^r\sim KE_{op}^2\tau_{laser}$], the induced refractive-index change depends only on the optical intensity and its pulse width, and the orientation of the optical field vector with respect to the director axis. Such laser-induced reorientation effects were reported in Ref. 3 with nanosecond laser pulse.

In conclusion, we have derived an analytical expression for the laser-induced refractive-index change associated with the director axis reorientations, the density and temperature changes. The results for the self-diffraction contain general features which are in agreement with experimental observations. They allow us to make a quantitative determination of the relative contributions from all three mechanisms for the specific interaction geometries used in previous experiments. More recently, density and temperature effects have been observed in experiments involving self-diffraction and probe beam diffraction in a nematic liquid crystal that exhibits all three mesophases (smectic, nematic, and isotropic) of liquid crystals.[12] Both transient effects (of the type described here) and permanent grating effects (similar to the permanent grating induced by nanosecond laser pulses reported in Ref. 6) can be induced by a single-shot 66-ps laser pulse [second-harmonic (0.53 μm) output of a Q-switched, mode-locked Nd:YAG laser system,[12] where YAG means yttrium aluminum garnet]. A complete analysis of these effects and estimates of the absolute contributions from the density, temperature, and reorientation effects in these three mesophases of liquid crystal will be presented in a longer article elsewhere.

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