Flow-Alignment and Inertial Effects in Picosecond Laser-Induced Reorientation Phenomena of Nematic Liquid Crystals

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Collective molecular-reorientation phenomena in the nematic liquid crystal 5CB are investigated in dynamic-grating experiments using picosecond excitation pulses. Grating buildup and decay is monitored by diffraction of a cw laser beam. It is shown that molecular reorientation still increases after the excitation pulse but relaxes exponentially later. The observed dynamics can be explained by a flow-alignment theory considering photoelastic stresses leading to additional forces besides the optical torques, which are well known from previous experiments.

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The molecular alignment of a well-prepared and oriented nematic liquid crystal can be distorted easily by external fields, e.g., by static electric or magnetic fields [1], flow fields [2], or optical fields [3]. Light-induced reorientation of the nematic director results in nonlinear optical effects which may be useful for phase conjugation, for photonic switching and processing, or in optically bistable systems. The torque-induced orientational optical nonlinearity has been investigated with low-power cw lasers [4] but also recently with short laser pulses on the nanosecond [5] and picosecond [6] time scales. Time-resolved grating experiments [7,8] show that the dynamical responses of liquid crystals to short intense ps laser pulses are rather complex in general, because of coupling between density, temperature, flow, and reorientation. A first attempt to a quantitative description of the observations has been presented by Khoo *et al.* [8] using an experimental arrangement where *parallel* polarizations of the excitation beams nearly normally incident on a homeotropic liquid crystal did *not allow* laser-induced reorientation in principle.

In the present paper we report investigations with a different geometry which *allows* laser-induced reorientation effects, on one hand, and uses *crossed* excitation beam polarizations to avoid thermal gratings, on the other hand. It is shown that the short excitation pulse is followed by a delayed reorientation process in this case, indicating a large effective inertial moment. This inertial moment is explained in terms of flow-alignment effects which are not driven by density or temperature modulations but by photoelastic stresses.

The experimental wave-mixing arrangement is sketched in Fig. 1. The two 80-ps FWHM excitation pulses (532 nm) are obtained from a frequency-doubled mode-locked Nd-doped yttrium-aluminum-garnet laser with a symmetric beam splitter. The linearly polarized beams are focused to a diameter of 800 μm on a 25-μm film of a homeotropically aligned nematic liquid crystal 5CB (4-*n*-pentyl-4′-cyanobiphenyl) which is temperature controlled at $T=25.0\pm0.1\,^{\circ}\mathrm{C}$, unless noted otherwise. The polarizations of the excitation beams are either parallel or perpendicular to each other, producing intensity or polarization gratings in the sample. The resulting optical-field fringes modulate the optical properties of the birefringent liquid. The center of the induced phase grating (period $\Lambda=30$ μm) is probed by a weak argon cw laser (488 nm) with a spot diameter of 100 μm. The first-order diffracted probe-beam intensity is measured with a photodiode and a fast real-time oscilloscope. The detector response to an 80-ps laser pulse is about 2 ns FWHM. The unperturbed director and the optical fields E_{Ar} and E_1 are in the x-z plane while the second pump field E_2 is chosen parallel or perpendicular to E_1. If $\mathbf{E}_1\|\mathbf{E}_2$ the two beams interfere to give an intensity grating while in the case $\mathbf{E}_1\perp\mathbf{E}_2$ a polarization grating [9] with a much weaker intensity modulation (due to imperfect polarization of the beams) is formed. $\mathbf{E}_1\perp\mathbf{E}_2$ is used for the distinct excitation of polarization-dependent effects like, e.g., reorientation, and to suppress laser-induced thermal gratings at the same time. Although we observed that the linear absorption of 5CB is fairly weak (about $\alpha=0.01\ \mathrm{cm}^{-1}$) at the green excitation wavelength, strong periodic heating occurs in the case $\mathbf{E}_1\|\mathbf{E}_2$ at high fluence due to a three-photon absorption process as will

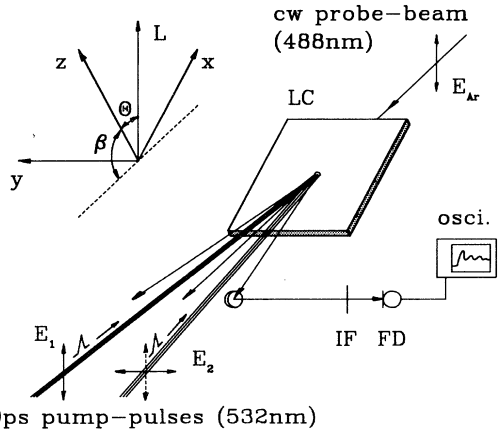

FIG. 1. Experimental wave-mixing arrangement with LC, liquid crystal 5CB; FD, fast photodiode; IF, interference filter (488 nm); E_1 and E_2, optical excitation fields; E_{Ar}, optical probe field; Θ, reorientation angle, $\beta=22.5°$.

be discussed elsewhere. However, using a polarization grating and excitation energies between 0.1 and 0.6 mJ, thermal gratings have not been observed, although this nonlinear absorption is strongly anisotropic. The contribution of laser-induced thermal birefringence changes has been reduced further by choosing the angle between the probe-beam polarization and the nematic director [5]. The angle $\beta=22.5°$ (see Fig. 1) results in small thermal refractive index changes and helps to overcome the "nematic barrier" which occurs in reorientation phenomena at normal incidence [6,7,9].

Figure 2 displays oscilloscope traces of the diffracted probe intensity during the first 500 ns after excitation with an 80-ps pulse for parallel and perpendicular polarizations. In both cases the diffracted signal is still increasing long after the pump pulse. The strong oscillations in the case $\mathbf{E}_1 \| \mathbf{E}_2$ are due to laser-induced standing ultrasound waves [9] and diffraction at the resulting density modulations ("forced Brillouin scattering"). These acoustic gratings can be explained by thermal heating due to the nonlinear absorption process discussed above and are suppressed almost completely by using crossed polarizations of the excitation beams as shown in Fig. 2. This experimental result indicates that thermal gratings do not contribute much to the diffracted signal and can be neglected in the following discussion if the excitation is performed with a polarization grating as mentioned above.

The slow increase of the signal at low energies for polarization-grating excitation is of the type $1-\exp\{-t/\tau_r\}$, with rise times τ_r between 20 and 50 ns, depending on excitation energy as also shown in Fig. 2. Afterwards the diffracted signal decreases exponentially as can be seen in Fig. 3. The evaluated decay times are in the millisecond range, depending on sample temperature. The observed grating decay is a second experimental result confirming that thermal grating contributions are less important for our experimental conditions, because the relaxation of a thermal grating is governed by a heat diffusion time of about 250 μs in the present geometry, which we did not observe with $\mathbf{E}_1 \perp \mathbf{E}_2$ at $\beta=22.5°$, but did, e.g., in the case $\mathbf{E}_1 \| \mathbf{E}_2$ at $\beta=0$.

The observed diffraction dynamics can be described by a flow-alignment theory assuming that laser-induced flow gradients modulate the orientation of the nematic director and the refractive index of the birefringent fluid to form a phase grating. The first-order diffracted intensity for weak thin gratings is proportional to the squared amplitude of the induced phase modulation [10], which is mainly given by the induced refractive index change at a given probe-beam polarization. The thin-grating approximation is valid here because the observed diffraction efficiency only amounts to a few percent or less. The problem is simplified by treating the case where the director rotates only in the x-z plane, which is the main effect as follows. The refractive index changes δn established by molecular reorientation are then given by

$$\delta n = n_e(\Theta+\beta) - n_e(\beta)\,, \tag{1}$$

where $n_e(\vartheta) = n_\perp n_\| (n_\|^2 \cos^2\vartheta + n_\perp^2 \sin^2\vartheta)^{-1/2}$ with $\vartheta=\Theta+\beta$ and $\vartheta=\beta$, respectively. $n_\|$ and $n_\perp$ are the refractive indices parallel and perpendicular to the optical axis. Exact calculation of the reorientation angle due to light-induced flow alignment requires a self-consistent treatment of the molecular-reorientation dynamics together with the hydrodynamics of the anisotropic fluid, which is a complex physical problem in general [11]. The treatment can be simplified here because only one strong flow component is induced in the performed polarization-grating experiments. The director motion can be described by a single reorientation angle Θ in this case which is obtained by a torque balance in addition to the

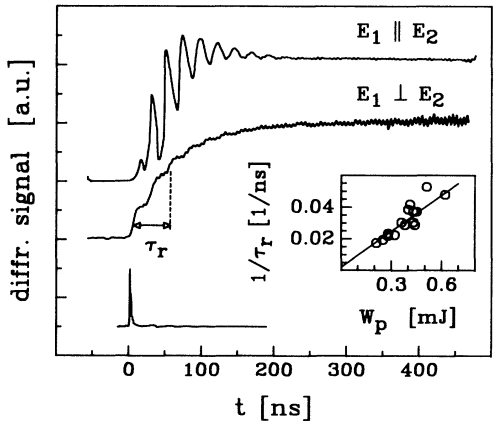

FIG. 2. Diffracted probe-beam intensity vs time after ps grating excitation with parallel or perpendicular pump-beam polarization. Excitation energy is $W_p=0.25$ mJ. The detector response to the excitation pulse is shown for comparison. Inset: The evaluated reciprocal rise times vs pump energy.

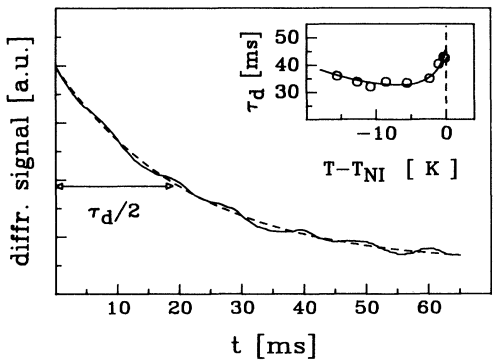

FIG. 3. Grating decay after excitation with 80-ps laser pulse at $T=25\,°C$. The solid line corresponds to experimental oscilloscope trace; the broken line is an exponential fit. Inset: Evaluated decay times vs reduced temperature. $T_{NI}=36.5\,°C$ is the nematic-isotropic phase-transition temperature.

Navier-Stokes equation [12],

$$\mu\frac{\partial^2\Theta}{\partial t^2}+\gamma_1\frac{\partial\Theta}{\partial t}+M_{\rm el}+M_f+M_{\rm op}=0\,, \tag{2a}$$

$$\rho_0\frac{\partial v_x}{\partial t}-\gamma_s\Delta v_x=F_x\,, \tag{2b}$$

where $M_{\rm el}=-K\Delta\Theta$ is the elastic torque, $M_f=-\frac{1}{2}(\gamma_1-\gamma_2\cos2\Theta)\partial v_x/\partial z$ is a flow coupling term, and $M_{\rm op}$ is the optical torque [5]. K is an elastic constant in a one-elastic-constant approximation, γ_1 is the rotational viscosity, γ_s and γ_2 are flow viscosities, $\varepsilon_a=\varepsilon_\parallel-\varepsilon_\perp$ is the optical dielectric anisotropy, ρ_0 is the mass density, and E is the optical field. μ is an inertial moment (density) connected with reorientation phenomena. v_x is the x component of a flow velocity which is excited by an external force F_x.

In laser-induced reorientation experiments where reorientation is due to optical torques resulting from the optical anisotropy of the liquid crystal, the description with Eq. (2a) is sufficient. For our case, however, it turned out that this equation alone gives far too small reorientation rise times of picoseconds or less, because the inertial moment μ is rather small ($\sim10^{-16}$ kg/m). Therefore translational motion of the molecules is also considered by adding Eq. (2b). In liquid crystals this translational motion is coupled to rotation and reorientation of the molecules via anisotropic frictional forces described by the flow coupling term M_f. Following this model, the "inertia" of a laser-induced viscous flow drags the reorientation further, resulting in the observed nanosecond grating buildup times, even when the excitation is over. Although a similar process has already been discussed in principle in Ref. [5], it was not observed in this work. It should also be noted that a double-exponential time dependence of the laser-induced thermal birefringence as reported in these previous experiments will not occur in a grating experiment and therefore cannot explain our results.

For excitation of the transverse flow, shear forces have to be present, which result from photoelastic stresses [12]. However, the elasto-optic coefficients relating these stresses to the light field are not known for liquid crystals. Therefore we consider only the force density $\mathbf{F}$ corresponding to Maxwell stresses [13] in Eq. (2b) as a qualitative approximation:

$$\mathbf{F}=(\mathbf{D}\cdot\nabla)\mathbf{E}^*-\tfrac{1}{2}\nabla(\mathbf{E}\cdot\mathbf{D}^*)\,. \tag{3}$$

$\mathbf{E}$ is the optical field, $\mathbf{D}$ is the displacement vector, and the asterisk denotes the complex conjugate. The complex notation gives the time-averaged real value of $\mathbf{F}$. From Eq. (3) the main force for a polarization grating is given by the x component $F_x=\frac{1}{2}\varepsilon_0\varepsilon_\perp n_\perp q_l E^2\cos(q_l y)$ in our geometry and the excitation of other velocity components is negligible. q_l is the wave number of the induced grating. The inverse flow-alignment process has been estimated to be of less importance in the present experiments and has been omitted from Eq. (2b) so far. Further, a coupling between molecular alignment and laser heating has been neglected.

Neglecting further the second time derivative because of the small inertial moment μ as discussed above, an approximate solution of Eqs. (2) under the boundary conditions $\Theta(z=0)=\Theta(z=d)=0$ and $v_x(z=0)=v_x(z=d)=0$ is found by restriction to the lowest spatial modes supporting each other as

$$\Theta=\Theta_m(t)\sin\{q_l z\}\cos\{q_t y\}\,, \tag{4a}$$

$$v_x=v_m(t)(1-\cos\{q_l z\})\cos\{q_t y\}\,, \tag{4b}$$

where $q_l=2\pi/d$ and $q_t=2\pi/\Lambda$. The time-dependent amplitude $v_m(t)$ can be eliminated from Eqs. (2) in this case to give

$$\tilde\mu\frac{\partial^2\Theta_m}{\partial t^2}+\tilde\gamma\frac{\partial\Theta_m}{\partial t}+\tilde D\Theta_m=\tilde F(t)\,, \tag{5}$$

with

$$\tilde\mu=\frac{2\rho_0\gamma_1}{q_l q_t(\gamma_1-\gamma_2)},\quad \tilde\gamma=\frac{2(q_l^2+q_t^2)(\rho_0K+\gamma_1\gamma_s)}{q_l q_t(\gamma_1-\gamma_2)},$$

$$\tilde D=\frac{2K\gamma_s(q_l^2+q_t^2)^2}{q_l q_t(\gamma_1-\gamma_2)},\quad \tilde F(t)=\frac{F_x}{q}=\frac{1}{2}\varepsilon_0\varepsilon_\perp E^2(t)\,.$$

In evaluating Eq. (5) we have also neglected the optical torque, which gives a small contribution to the expression $F(t)$ on the right-hand side, and we have used the small-angle approximation, $\gamma_1-\gamma_2\cos\{2\Theta\}\approx\gamma_1-\gamma_2$.

According to this simple model the time-dependent amplitude of the reorientation grating is described by the equation of an overdamped oscillator if we use typical material parameters $\gamma_1=0.01$ kg/ms, $\gamma_2=-1.09\gamma_1$ [5], $K=10^{-12}$ N, $\rho_0=1$ g/cm^3, and experimental data $q_l=0.25\ \mu\text{m}^{-1}$, $q_t=0.2\ \mu\text{m}^{-1}$. Note that the second time derivative appearing in Eq. (5) is a consequence of the elimination of the flow and is quite different from the one given in Eq. (2a), which has been omitted here. The apparent parameter $\tilde\mu\approx\rho_0/q_l q_t$ can be interpreted as the inertial moment (density) of the flowing molecules in the excited regions. It can be calculated to be $\tilde\mu=10^{-8}$ kg/m using the given parameters and is much larger than the inertial moment μ given above. If we approximate our pump pulse by a delta peak $E^2(t)=E_0^2\delta(t)$ the solution of Eq. (5) is given by

$$\Theta_m=\frac{\varepsilon_0\varepsilon_\perp n_\perp E_0^2\tau_p}{4\delta'\tilde\mu}(\exp\{-t/\tau_d\}-\exp\{-t/\tau_r\})\,, \tag{6}$$

where $\delta'=(\delta^2-\omega_0^2)^{1/2}$ with $\delta=\tilde\gamma/2\tilde\mu$ and $\omega_0=[K(q_l^2+q_t^2)/\tilde\mu]^{1/2}$. The quantities $\tau_r=(\delta+\delta')^{-1}$ and $\tau_d=(\delta-\delta')^{-1}$ can be interpreted as the rise time and the decay time of the reorientation process. τ_p is the laser pulse duration. Developing the root δ' results in

$$\tau_r=\rho_0/\gamma_s(q_l^2+q_t^2)\,, \tag{7a}$$

$$\tau_d=\gamma_1/K(q_l^2+q_t^2)\,. \tag{7b}$$

As with simple reorientation models the relaxation of the induced grating is given by the viscoelastic behavior of the nematic fluid whereas the observed rise times are governed by the damping of the induced flow and the flow viscosity. If we take $\gamma_1 = 0.015$ kg/ms and $K = 7 \times 10^{-12}$ N, one gets $\tau_d = 36$ ms in good agreement with experimental data (at $T = 25\,^{\circ}\mathrm{C}$). The observed temperature dependence of τ_d as shown in Fig. 3 can be explained by the temperature dependence [2] of γ_1/K, confirming that we have induced reorientation effects by the ultrashort laser pulses. However, using Eqs. (1) and (6) with the material parameters as given above, the calculated grating diffraction efficiency is smaller than observed. Therefore the photoelastic coupling has to be stronger than expected from Maxwell stresses only.

The observed energy dependence of τ_r can be explained assuming a non-Newtonian flow viscosity. In this case the viscosity and thus [cf. Eq. (7a)] the reciprocal rise time contains a term which is proportional to the induced shear rate dv_x/dz, which, on the other hand, is proportional to the excitation energy W_p as can be seen with Eqs. (2b) and (3). As a consequence, a linear dependence of $1/\tau_r$ on W_p can be expected, which is in agreement with the experimental data of Fig. 2. By extrapolating this graph to $W_p \rightarrow 0$ we get with Eq. (7a) $\gamma_s = 0.02$ kg/ms which is in excellent agreement with published data for small shear rates [14].

In conclusion, we have investigated the dynamics of molecular-reorientation phenomena in a transient-grating experiment using picosecond excitation pulses. We have observed an increase of the director deformation for times much longer than the pump-pulse duration, leading to a delayed maximum of the diffracted signal. The reorientation grating relaxes afterwards exponentially with decay times of 30 to 40 ms, which can be explained well by the viscoelastic properties of the distorted liquid crystal. The observed rise times are on the order of 20 to 50 ns, depending on pump energy. These experimental rise times agree with calculations assuming molecular reorientation due to a laser-induced flow which persists a corresponding time after the excitation pulse. The transverse flow is generated by photoelastic stresses in the anisotropic fluid. The theoretical description is based on coupled equations for the reorientation angle and the flow velocity. For our experimental situation the flow velocity can be eliminated, leading to a harmonic oscillator equation for the reorientation angle. This equation contains an inertial term which is much larger than expected from the inertial moments of individual molecules.

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