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Inertial effects in the dynamics of picosecond laser-induced molecular reorientation in nematic liquid crystals

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Abstract

Dynamics of collective molecular reorientation phenomena in a nematic liquid crystal 5CB are investigated in transient grating experiments using picosecond excitation pulses.

Molecular correlations in nematic liquid crystals can lead to a more or less uniform alignment of the molecules, which can be distorted easily by external fields[1]. Optical field-induced collective reorientation[2] of nematics has been investigated with low power cw-lasers[3,4], with short laser pulses on the nanosecond time scale[5,6] and has been demonstrated with picosecond pulses recently[7]. Light- induced reorientation results in nonlinear optical effects which may be useful in applications like phaseconjugation, photonic switching and processing of light or optical bistability.

In the present paper we describe investigations of dynamic collective molecular reorientation phenomena in a nematic liquid crystal by using transient grating techniques and picosecond excitation pulses. For the first time we have observed effects which give evidence that inertial motions of many correlated molecules have to be considered in this case. It is discussed how the experimental results can be explained by a flow alignment theory.

In our experiments we used a wave-mixing arrangement which is schematically shown in Fig. 1a. Two pump-pulses (532 nm) of 80 ps FWHM obtained from a frequency doubled mode-locked Nd:YAG laser with a single-pulse extraction are focussed to an e^{-2}-diameter of 800 μm on a thin film (25 μm) of a homeotropically aligned nematic liquid crystal 5CB (4'-n-pentyl-4-cyanobiphenyl). The two pump beams are linear polarized, their polarizations either parallel or perpendicular to each other, producing an intensity- or polarization grating in the sample. The resulting optical field-fringes modulate the alignment of the molecules and change the optical properties of the birefringent liquid by rotating the alignment-tensor (or the director) and the optical axis. The center of the induced phase-grating (grating period $\Lambda = 30\,\mu m$) is probed by a weak argon cw-laser (488 nm) with a spot diameter of 100 μm. The first order diffracted intensity of the probe-beam is measured with a fast fotodiode and a fast real-time oscilloscope. The oscilloscope traces are recorded and processed with a digitizing video-camera system. The time resolution is limited by a rise-time of 400 ps and a decay-time of less than 4 ns with the PIN-diode used so far. The experimental geometry is depicted in Fig. 1b. The unpertubed 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 $E_1 \parallel E_2$ the two beams interfere to give an intensity-grating and reorientation effects will occur as well as e.g. thermal effects (due to finite absorption and laser-heating) in a periodic structure, leading to diffraction of the probe beam. If however $E_1 \perp E_2$, the two beams do not interfere and intensity dependent effects (e. g. thermal heating) do not appear as a grating and are not detected. But the optical fields still form a polarization-grating[8] and the liquid crystal will respond with a periodic reorientation pattern which can now be detected by diffraction without any additional intensity-dependent effects.

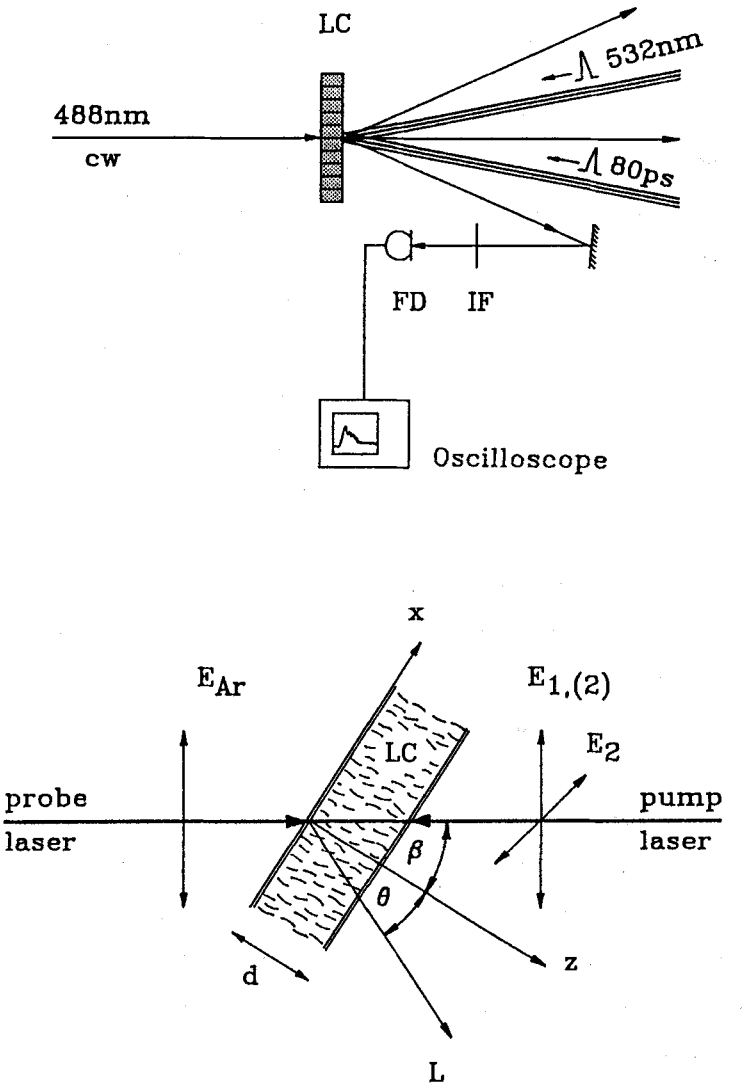

Fig.1: Wavemixing arrangement (a) and experimental geometry (b). LC: Liquid crystal 5CB, FD: Fast photo diode, IF: Interference filter (488nm), E_1 and E_2: Optical excitation fields, E_{AR}: Optical probe field, Θ: Reorientation angle, $\beta = 22.5$ deg.

The direction of the initial alignment has always been $\beta = 22.5$ deg to overcome the "nematic barrier", which will occur in the case of normal incidence[7]. The liquid crystal is further placed in a temperature-controlled stage, stabilized at $T = (25.0 \pm 0.1)\,^{\circ}C$ (unless not noted otherwise).

Fig. 2 displays typical oscilloscope traces of the diffracted probe intensity during the first 500 ns after excitation with a 80 ps pulse for the two types of excitation grating. In both cases the diffracted signal is still increasing long after the pump-pulse (which can be approximated as a δ-peak at $t \equiv 0$ on the graph) leaves the sample, showing strong oscillations in the case $E_1 \parallel E_2$. These oscillations occur due to laser-induced standing ultrasound waves and additional diffraction at the resulting density modulations (forced Brillouin-scattering). In liquid crystals such acoustic gratings are generated by thermal heating or electrostriction and can be supressed by using crossed polarization of the excitation beams as can be seen in the trace with $E_1 \perp E_2$. It should be mentioned however, that forced Brillouin-scattering has also been observed even with a polarization-grating at higher pump energies in these experiments, which is beyond the scope of the present paper and will be discussed elsewhere.

The slow increase of the signal at low energies is given by a function of the type $(1 - \exp\{-t/\tau_r\})$, with rise-times τ_r between 20 and 50 ns, depending on excitation energy as shown in Fig. 3. Much later the diffracted signal decreases exponentially as can be seen in Fig. 4. The evaluated decay-times are in the millisecond range, depending on sample temperature as depicted in Fig. 5.

The observed effects can be described by optical field-induced reorientation of the liquid crystal (for calculation of the optical nonlinearity) in combination with a simple grating diffraction model. The interference field of the

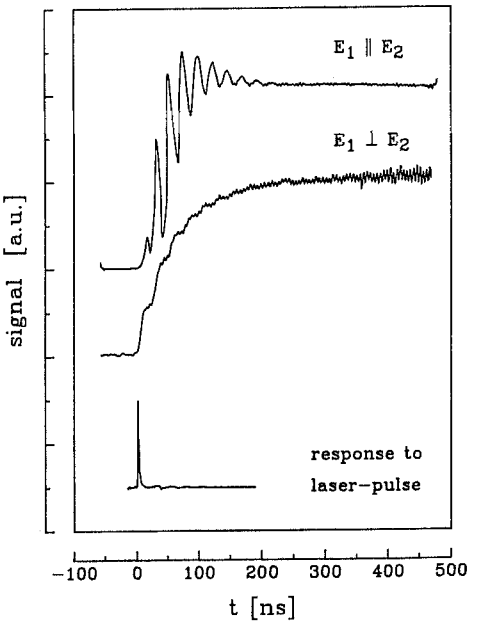

Fig.2: Diffracted probe-beam intensity vs. time after ps-grating excitation with parallel or perpendicular pump-beam polarization. Excitation energy is $W_P = 0.25$ mJ. The direct response of the detector-system is shown for comparision.

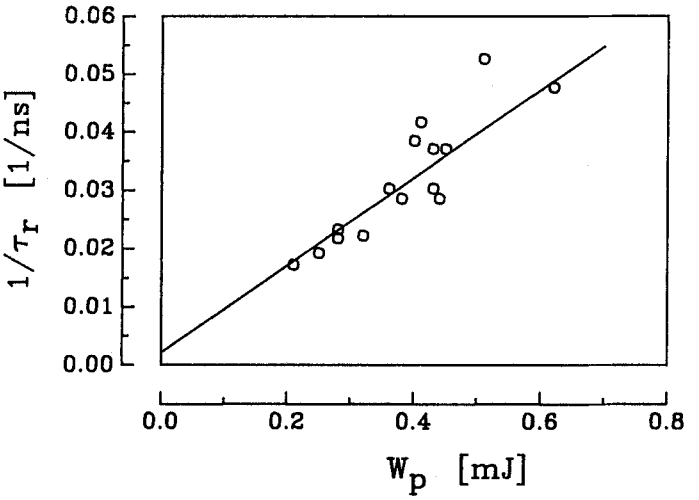

Fig.3: Evaluated reciprocal rise-time vs. pump-energy. Nematic 5CB $d = 25\,\mu m$, $T = 25\,^{\circ}C$, $\beta = 22.5$ deg.

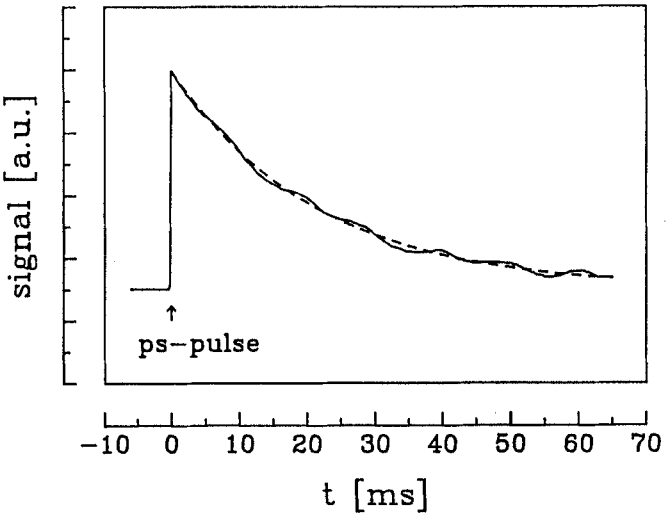

Fig.4: Grating decay after excitation with 80 ps laserpulse. Nematic 5CB d = 25 μm, T = 25 °C, β = 22.5 deg. Full line corresponds to experimental oscilloscope trace, broken line is an exponential fit.

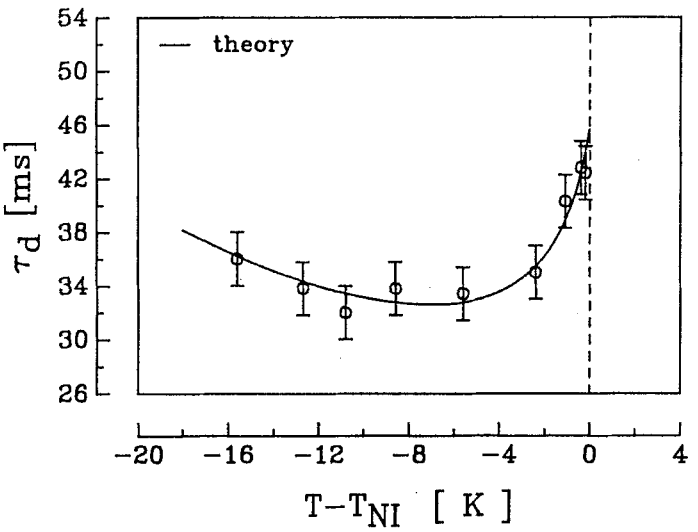

Fig.5: Evaluated decay-time vs. reduced sample temperature. T_{NI} = 36.5 °C is the nematic-isotropic phase transition temperature.

incident laser beams modulates the orientation of the director and the refractive index of the birefringent fluid to form a phase-grating. The first order diffracted probe-beam intensity I_d for weak thin gratings can be written as [9]

$$I_d = I_o \left(\frac{\pi}{\lambda} \int_0^d \delta n(z,t)\, dz\right)^2 \qquad (1)$$

where λ is the light wavelength, d the film thickness, δn the induced refractive index change and I_o the input probe-beam intensity. If we simplify the problem 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 established by molecular reorientation are given by

$$\delta n = n_e(\Theta+\beta) - n_e(\beta) \qquad (2)$$

where $n_e(\vartheta) = n_\perp n_\parallel (n_\parallel^2 \cos^2\vartheta + n_\perp^2 \sin^2\vartheta)^{-1/2}$ with $\vartheta = \Theta+\beta$ and $\vartheta = \beta$ respectively. $n_\parallel$ and $n_\perp$ are the refractive indicees parallel and perpendicular to the optical axis. Field- and flow-induced reorientation of the alignment-tensor [10] for a nematic liquid in the given geometry can be described by a single reorientation angle Θ which is obtained by a torque balance in addition with Navier-Stokes equation[12]

$$\mu \frac{\partial^2\Theta}{\partial t^2} + \gamma_1 \frac{\partial\Theta}{\partial t} + M_{el} + M_f + M_{op} = 0 \qquad (3a)$$

$$\rho_o \frac{\partial v_x}{\partial t} - \gamma_s \Delta v_x = F_x \qquad (3b)$$

where $M_{el} = -K\Delta\Theta$ is the elastic torque, $M_f = -1/2\ (\gamma_1 - \gamma_2 \cos 2\Theta)(\partial v_x/\partial z)$ a flow coupling term and $M_{op} = -1/2\ \varepsilon_o \varepsilon_a E^2 \sin 2(\Theta+\beta)$ the optical force. K is an elastic constant in a one elastic constant approximation, γ_1 the rotational

viscosity, γ_s and γ_2 are flow viscosities, $\varepsilon_a = \varepsilon_\parallel - \varepsilon_\perp$, ρ_o the mass density and E the optical field. μ is an inertial moment connected with reorientation phenomena. v_x is the x-component of a flow velocity which is excited by forces due to Maxwell-stresses $\vec{F} = (\vec{D}\cdot\nabla)\,\vec{E}^* - \frac{1}{2}\,\vec{\nabla}\,(\vec{E}\cdot\vec{D}^*)$ in the treated case of a polarization grating mainly. As indicated by restriction on the x-component of the flow velocity the main stress is given by the x-component of $\vec{F}$ which holds for $F_x = \frac{1}{2}\,\varepsilon_o\varepsilon_\perp q_t\,E^2\cos q\,y$ in the given polarization-grating geometry, where q is the wavenumber 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. 3b so far. Further a coupling between molecular alignment and laser-heating has been neglected. Eq. 3 provides two possibilities to explain the observed dynamics[5], in particular the delayed maximum of the diffracted signal for times much longer than the excitation pulse duration. First, this delay can be attributed to the inertial moment μ of the rotated areas. Second, a laser-induced flow can provide such delay because the "inertia" of a viscous flow may drag the reorientation further by flow-orientation coupling processes, even when the excitation pulse has left the sample. Although the latter process has already been discussed by Shen et al. (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 the present grating arrangement and therefore cannot explain our results.

If we first discuss the inertial moment term and neglect the flow-coupling effects (i.e. $M_f = 0$), an approximate solution of the linearized eq. 3 under hard boundary conditions $\Theta(z=0) = \Theta(z=d) = 0$ in a plane wave grating experiment is

$$\Theta = \Theta_m(t)\,\cos\{qy\}\,\sin\{\frac{\pi}{d}z\} \qquad (4)$$

where q is the wavenumber of the grating and Θ_m obeys the equation

$$\mu\,\frac{\partial^2\Theta_m}{\partial t^2} + \gamma_1\,\frac{\partial\Theta_m}{\partial t} + D\,\Theta_m = \tilde{F} \qquad (5)$$

with $D = K(q^2 + \pi^2/d^2) - \varepsilon_o\,\varepsilon_a\,E^2\cos 2\beta$ and $\tilde{F} = \frac{1}{2}\,\varepsilon_o\,\varepsilon_a\,E^2\sin 2\beta$. Eq. 5 describes the dynamics of an overdamped oscillator if typical material parameters and experimental data like $\gamma_1 = 0.01\,kg/ms$, $K = 10^{-12}\,N$, $d = 25\,\mu m$, $q = 0.2\,\mu m^{-1}$ and $\mu \leq 10^{-4}\,kg/m$ are used.

If we approximate our pump-pulse by a delta-peak $E^2(t) = E_o^2\,\delta(t)$ (which corresponds to $\tilde{F}(t) = \tilde{F}_o\,\delta(t)$ by using the definition of $\tilde{F}$ given in eq. 5), the solution of eq. 5 is given by

$$\Theta_m = \frac{\tilde{F}_o}{2\,\delta\,\delta'\,\mu}\left(\exp\{-t/\tau_d\} - \exp\{-t/\tau_r\}\right) \qquad \text{for } t > 0 \qquad (6)$$

where $\delta = \gamma_1/2\mu$ and $\delta' = (\delta^2 - \omega_o^2)^{1/2}$ with $\omega_o = (K(q^2 + \pi^2/d^2)/\mu)^{1/2}$. 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. Developping the root δ' results in

$$\tau_r = \frac{\mu}{\gamma_1} \qquad (7a) \qquad , \qquad \tau_d = \frac{\gamma_1}{K(q^2 + \pi^2/d^2)} \qquad (7b)$$

The rise-time is given by the inertial moment divided by the viscosity and the decay-time depends mainly on the visco-elastic behaviour of the nematic fluid. Expression 7b has been also obtained with simple relaxation models where the inertial moment has been neglected [6]. If we take $\gamma_1 = 0.015\,kg/m\,s$ and $K = 7\cdot10^{-12}\,N$ one gets $\tau_d = 36\,ms$ in good agreement with experimental

data (at $T = 25\ ^{o}C$). The observed temperature dependence of τ_d can be explained by the temperature dependence[11] of γ_1/K, confirming that we have induced reorientation effects with ultra-short laser pulses.

A response time of e.g. $\tau_r = 40$ ns can be explained with $\mu = 6 \cdot 10^{-10}$ kg/m following this model. That corresponds to a correlated rotation of many molecules within a sphere of radius $l = 800$ nm but is about 10^6-times larger than the inertial moment of a single molecule which one would expect to be the relevant value concerned with simple director distortions. So optical-torque induced reorientation effects should occur even on a picosecond time scale as already has been demonstrated in self-diffraction experiments recently[7]. As a consequence this "commonly" used inertial moment of the director motion can not be responsible for the observed behaviour.

However, it is reasonable to explain the observed effects and the correlated motion of molecules in terms of flow-alignment as mentioned above. That would require a self-consistent treatment of the molecular reorientation dynamics coupled with the hydrodynamics of the fluid, which is a complex physical problem in general[10].

But the problem can be simplified as indicated with the coupled eqs. 3 above because v_x is the strongest flow-component in a polarization-garting experiment to be excited and the director motion can be described by a single reorientation angle Θ. The previously discussed intertial term will be neglected in the following. If further an approximate solution of the eqs. 3 is restricted to the lowest spatial modes which support each other

$$\Theta = \Theta_m(+)\ \sin q'z\ \cos qy \tag{8a}$$

$$v_x = v_m(t)(1-\cos q'z)\ \cos qy \tag{8b}$$

where $q' = 2\pi/d$, the time dependent flow amplitude $v_m(t)$ can be eliminated from eqs 3 to give once more

$$\tilde{\mu}\,\frac{\partial^2\Theta_m}{\partial t^2} + \tilde{\gamma}\,\frac{\partial\Theta_m}{\partial t} + \tilde{D}\,\Theta_m = F_{(t)} \qquad (9)$$

now with

$$\tilde{\mu} = \frac{2\rho_O\gamma_1}{qq'\,(\gamma_1-\gamma_2)}$$

$$\tilde{\gamma} = \frac{2\rho_O\,K(q'^2+q^2)}{q'q\,(\gamma_1-\gamma_2)} + \frac{2\gamma_1\,\gamma_s\,(q'^2+q')}{q'q\,(\gamma_1-\gamma_2)}$$

$$\tilde{D} = \frac{2\,K\,\gamma_s\,(q'^2+q^2)^2}{q'q\,(\gamma_1-\gamma_2)}$$

$$F_{(t)} = \frac{F_x}{q} = \frac{1}{2}\,\varepsilon_O\varepsilon_\perp\,E^2(t)$$

In evaluating eq. 9 we have neglected the optical torque (which gives just another contribution on the right 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 flow-alignment model the time-dependent amplitude of the reorientation grating is once more described by the equation of an overdamped oscillator, but now with a different interpretation of the relevant parameters $\tilde{\mu}$, $\tilde{\gamma}$ and $\tilde{D}$. For a δ-like excitationpulse the solution of eq. 9 remains the same as discussed above yielding in a rise- and decay-time as given in eqs. 7 by replacing these parameters to

$$\tau_r = \frac{\rho_O}{\gamma_s(q'^2+q^2)} \qquad (10a), \qquad \tau_d = \frac{\gamma_1}{K(q^2+q^2)} \qquad (10b)$$

As with simple reorientation models the grating decay is given by the visco-elasticity of the liquid crystal whereas the observed rise-times are governed by the damping of the induced flow and the flow viscosity.

The apparent value $\tilde{\mu} \approx \rho_0/(q'^2+q^2)$ can be interpreted as the intertial moment (density) of the correlated molecular motions of the excited regions, which is concerned with the investigated flow and reorientation processes.

To explain the observed energy-dependence of τ_r one have to assume a non-Newtonian flow-viscosity

$$\gamma_s = \gamma_{so} + \gamma' \frac{dv_x}{dz} \tag{11}$$

which results (with $dv_x/dz \sim E^2 \tau_p \sim Wp$) in

$$\frac{1}{\tau_r} = kW_p + \frac{\gamma_{so}(q'^2+q^2)}{\rho_0} \tag{12}$$

where γ' and k are constants, Wp is the excitation energy and γ_{so} is the flow-viscosity at low shear-rates $dv_x/dz \to 0$. By plotting $1/\tau_r$ as a function of Wp one would expect a linear graph which is in good agreement within the uncertinities of the experimental data as shown in Fig. 3. By extrapolating the graph to $Wp \to 0$ we get the value $\gamma_{so} = 0.02$ kg/ms which is in excellent agreement with published data for small shear rates[13].

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 observed rise-times are in the order of 20 to 50 ns, depending on pump energy. The reorientation grating relaxes later with an exponential tail showing decay-times of 30 to 40 ms, which can be explained well by the visco-elastic properties of the liquid crystal. The observed delayed reorientation can be described by a laserinduced flow and flow alignment theory. A correlated motion of many molecules has to be

considered in this case and the rise-time can be interpreted in terms of a inertial moment of the excited regions.

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