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Nonlinear Elastic Response of the Liquid-Crystalline Structure to Large Deformations

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The reorientational process in liquid crystals (LC) is usually described in the frame of fluid mechanics (hydrodynamics) by a "classical" Oseen-Franck – Leslie equation under several simplifying assumptions. One of them is the linearity of the elastic response of a LC-structure to the induced stresses causing its deformation. This equation provides essential parameters of the reorientation process like threshold values and it's dynamics which correspond well to those observed experimentally. However, the description of the LC-elasticity by a single-value elastic coefficient (i.e., by linear Hook's law) is an approximation, since real processes are in general nonlinear. In this work an attempt is made to check if this linearity assumption can be extended to large deformations far above threshold. We expected in this case nonlinear effects to be present in structural elasticity because elastic forces result from molecular interactions, which are generally nonlinear, i.e., fundamental interactions obviously decrease with deformation (distance) like $1/r^2 \div 1/r^6$. *Such a decrease is also likely for orientational deformations of molecular arrangement in the LC-structure "mean" field. So an important question is to what extent the nonlinearity appears in the practical operation of LC-based devices. The elastic nonlinearity, if not negligible, may play an important role in applications. The elastic properties of a liquid-crystalline structure substantially influence the threshold value, sensitivity, and dynamics of the induced reorientation process. The elastic nonlinearity may also contribute to a hysteresis of the reorientation, introducing or enhancing bistable behavior in a LC-structure. Thus the elastic nonlinearity may affect operation of liquid crystalline photonic devices and its detailed examination would help in their improvement and optimization. This paper presents also a simple method of detection of the elastic constant changes in a deformed LC-structure.*

Keywords Liquid crystals; reorientation dynamics; structure elasticity

Introduction

An elastic response of LCs arises if an ordered LC-structure is subjected to external stimuli, like electric or optical fields inducing orientational distortion, provided that somewhere in the structure the LC-molecules are anchored and hence resist orientational changes. When these stimuli disappear, elastic forces related in general with bend, splay and twist deformations tend to restore initial order and a relaxation process begins.

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Orientational free relaxation of the LC-layer, in the case of "strong anchoring" at the layer boundaries, is governed by a simple exponential decay law [1,2]:

$$\theta(t) = \theta_{\max} e^{-\frac{t}{\tau_r}} \tag{1}$$

with characteristic relaxation time τ_r inversely proportional to the effective elastic coefficient K of the corresponding deformation:

$$\tau_r = \frac{\gamma d^2}{K\pi^2}, \tag{2}$$

where γ is the rotational viscosity and d stands for an LC-layer thickness in the case of spatial deformation. For the deformation constrained around a laser beam having cross-section diameter ρ, d should be replaced by an "effective" thickness d′, given approximately by $1/\mathrm{d}'^2 \approx 1/\mathrm{d}^2 + 1/\rho^2$.

By re-writing Eq. (1) one obtains the time t that passes until the relaxing deformation reaches some value θ(t):

$$t = \tau_r \ln \theta_{\max} - \tau_r \ln \theta(t) \tag{3}$$

In the simple experiment to be described later we could only monitor θ(t) for fixed, discrete values θ_n corresponding to the times t_n

$$t_n = \tau_r \ln \theta_{\max} - \tau_r \ln \theta_n \tag{4}$$

However, since we couldn't measure the value of θ_n directly in this experiment, we evaluated the changes of τ_r from the slope of the function $t_n(\theta_{\max})$, i.e.,

$$\frac{dt_n}{d\theta_{\max}} = \frac{\tau_r}{\theta_{\max}} + \frac{d\tau_r}{d\theta_{\max}} (\ln \theta_{\max} - \ln \theta_n) \tag{5}$$

In the linear case, K is a constant and the relaxation time τ_r = const. In such a case the slope is

$$dt_n/d\theta_{max} = \tau_r/\theta_{max}$$

and doesn't depend on the specific deformations θ_n monitored in the relaxation process.

It should be, however, also pointed out that the real re-orientation process is complicated by the coupling between molecular rotation and hydrodynamic motion of the LC, which is usually manifested by backflow and "kickback" effects [1].

Results and Interpretation

Experiment I

To validate our expectations, in the first part of the experiment we induced optically a local deformation of the LC-structure around an (Ar+)-laser beam passing through a homeotropically aligned nematic layer. The degree of the deformation

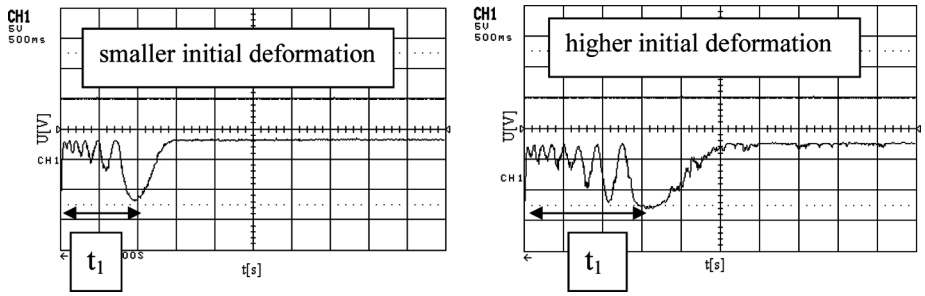

Figure 1. The time evolution of the diffraction rings (detected as minima and maxima in the diffraction image) during the relaxation process of a deformed LC-layer for two different initial deformation magnitudes. The time t_1 ($n = 1$) is indicated on the graphs.

was controlled by changing the beam intensity. The number of self-diffraction rings appearing in the far-field was a measure of deformation magnitude. Thus the number of rings can be attributed to a certain reorientation angle θ_{max} in a given location (for example in the center at $z = d/2$) of the LC-sample. After the deformation reached steady-state, the laser was shut off and the relaxation process from the induced deformation θ_{max} back to the aligned equilibrium state was monitored by observing the evolution of rings as a function of time (Fig. 1).

During this relaxation from various initial deformations θ_{max} we measured the times t_n at which the selected n-th ring reached a fixed detector position. In other words, t_n (θ_{max}) was measured, representing LC-relaxation speed from the various initial deformations θ_{max} to the fixed values θ_n. The results were taken in the separate series for each θ_n.

The measured experimental data obtained with increasing laser power P_{max} (related to increasing deformation θ_{max}) are shown in Figure 2. The variation of t_n with laser power t_n (P_{laser}), presented for $n = 1,2$, and 5, is based only on a few values of power, because stable deformation was only observed in a narrow range of exciting fields. Nevertheless the slopes of $t_n(\theta_{max})$ are clearly a decreasing function of n, so that they decrease correspondingly for t_1, t_2, and t_5 at each particular P_{laser} (θ_{max}).

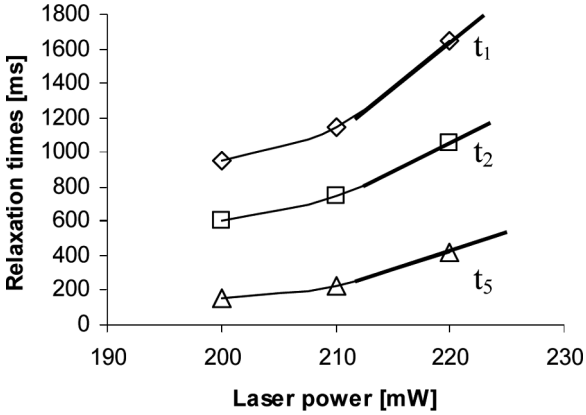

Figure 2. The variation in the decay time t_n during relaxation of the LC-layer, deformed by the laser beam, versus the laser power, P_{laser}, shown for $n = 1$, 2, and 5.

According to Eq. (5) this implies that

$$\frac{d\tau_r}{d\theta_{\max}} > 0$$

i.e., that the characteristic relaxation time τ_r increases with deformation $\theta_{\max}$. However, based on Eq. (2), τ_r is affected not only by the elastic modulus K, but also by the rotational viscosity γ and the deformation diameter d (effective LC-layer thickness). Since all three parameters may be dependent on beam intensity, one has to be careful in drawing conclusion from the above observation about changes in the elasticity.

Experiment II

In the next part of our experiment the same measurements were repeated for deformations induced by voltage. Now the magnitude of the LC-layer deformation, which is spatial and homogeneous "in-plane", was detected by observing changes in the layer birefringence (Fig. 3).

The samples with different textures – either with hometropic (initially bend deformation) or with planar texture (initially splay deformation) were investigated.

After the voltage applied and removed, relaxation times $t_n(V)$ were measured for various voltages as discussed previously. Results are shown in Figure 4. Now, increasing the voltage causes an increasing deformation $\theta_{\max}$.

In this case, for all examined geometries the effect of the τ_r-increase with increasing deformation was also observed, although it was smaller. The changes are more apparent for smaller voltages. It may result from the birefringence method of deformation detection used, which is less sensitive than diffraction. Moreover, for higher voltages the deformation inside a LC layer quickly saturates, canceling elastic stress, and only thin distorted surface regions remain, which don't contribute much to the layer birefringence.

The changes in the characteristic relaxation time τ_r observed in our experiments can be a result of several effects, including those not related to variations of K with deformation magnitude $\theta_{\max}$. In particular, in the first experiment thermal effects caused by the laser beam should be taken into account. The temperature rise influences both the elastic coefficient K and viscosity η in Eq. (2). Hence both K and

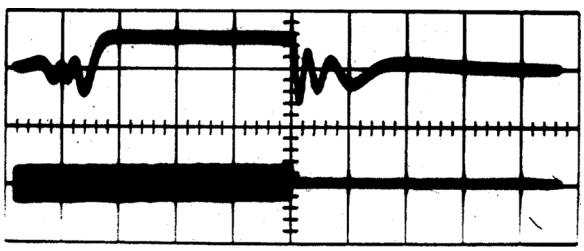

Figure 3. The record of the birefringence changes induced by 100 Hz A.C. voltage in a LC-layer. Upper trace: the intensity of the transmitted light, observed through crossed polarizers. Lower trace: the amplitude of the applied voltage. The time scale (horizontal): 0,5 s/div, the voltage scale (vertical) 2 V/div.

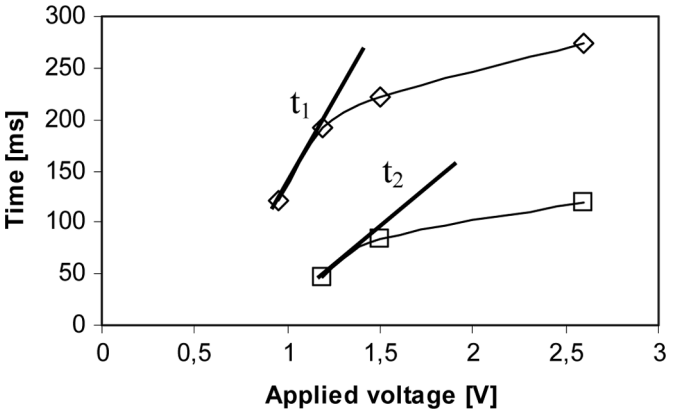

Figure 4. The relaxation times t_n of the LC-layer, deformed by an A.C. voltage, versus increasing deformation (voltage) for $n = 1, 2$.

η decrease with increasing temperature. Since in addition the laser power during these measurements has changed no more than 10%, so one can expect the resulting change in the relaxation time τ_r to be rather small. Moreover, thermal effects in such experiments are usually much faster (ms) than that observed here (hundreds of ms). In the second experiment thermal effects can be excluded completely.

In the both experiments, however, the relaxation process investigated is affected by the LC-backflow and kickback effects [1,3], the strength of which may also depend on the deformation magnitude and the laser power. Potentially they can mask the elastic response of interest here. In our measurements we have used LC-cells of various thicknesses, even as small as 11 μm. In such a thin cell the hydrodynamic motion contributes significantly to LC-layer relaxation at the beginning and vanishes rather quickly (ms) [4] as comparing to the processes measured here. Moreover, the backflow, if present, causes a decrease in the effective viscosity of molecular rotation [1,5]. This would be manifested in our measurements as a decrease of the relaxation time τ_r, quite contrary to the observed rise in τ_r.

The third parameter that may influence the relaxation time τ_r in our first experiment is the effective LC-layer thickness d_{eff} (see Eq. (2)). It depends on the laser power via changes in the active cross-section diameter ρ of the laser beam. Laser beam intensity had a gaussian radial distribution. Then the illuminated LC-area, where the optical field intensity is higher than deformation threshold intensity, increases with laser power and d_{eff} increases consequently. However we don't expect that this effect could have influenced our measurements significantly, because of the narrow range of laser power utilized (change of less than 10%). Again, this effect doesn't exist in the second experiment.

At last, it is highly probable, that the LC-layer relaxation is a complex process comprising of several effects, characterized by different relaxation coefficients. In such a case, the effects involved in the process with different dynamics may contribute to the total relaxation time at different deformation magnitudes. For example, the LC-structure deformation may be combined with the distortion of surface orientation. It also can result in changes of the total τ_r as observed in our experiments. We have used LC-cells treated with various surfactants, giving various

anchoring energies, in order to investigate to what extent this could be the case in our measurements. No influence has been observed.

Conclusions

In this work the elastic response of a liquid-crystalline nematic structure to induced deformations was investigated. Structural deformation of the ordered nematic phase was induced locally by the optical fields associated with a laser beam, and also by a homogeneous electric field. The combined results of both experiments indicate that the observed rise in the characteristic relaxation time τ_r is caused by a decrease of the elastic constant K at large deformations rather than by changes of other parameters such as viscosity or beam size. This implies a nonlinear elastic response of the LC-structure.

Acknowledgments

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