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Laser Induced Reorientation of Nematic Liquid Crystals

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A significant increase of the divergence and appearance of a fringe system has been observed by illuminating of MBBA (p-n-methoxybenzilidene-p-butylaniline) and OCB (octyl-cyano-biphenyl) nematic liquid crystalline samples with a collimated beam of an argon ion laser. The dependence of this effect on laser power, beam polarization, and angle of incidence has been studied in homeopolar and planar sandwich-like cells of 50–150 μm thickness, in the nematic phase. At a homeopolar cell of MBBA (150 μm thickness), at normal incidence a threshold laser power of 45 mW was found. The phenomena can be explained as deformation of the orientation by the Fredericks effect due to light fields. The observations are in good agreement with the theoretical predictions including an estimation of the laser power threshold.

INTRODUCTION

The reorientation of the director of a nematic liquid crystal (NLC) in an external homogeneous magnetic field was discovered by Fredericks many years ago. This effect—called often as Fredericks transition—was then found to exist also in quasistatic electric and magnetic fields.[1] Fredericks effect due to the electric field of a laser beam has been reported by us recently.[2]

Optically a NLC acts like a uniaxial crystal with its optical axis lying parallel to the director *L*. Thus the reorientation of the director at any point of the crystal is accompanied by the reorientation of its optical axis causing a change in the extraordinary refractive index. For NLC, which have a positive optical anisotropy, the change of the refractive index may lead to the self-focusing of the transmitted beam. This self-focusing effect has been studied theoretically and experimentally by Zeldovich *et al.*[3] in a planar sample with a He-Ne laser. They observed more than twofold increase of the divergence of the beam.

The aim of this work is to present the main results of our investigations on the laser induced reorientation of nematic liquid crystals. In our experiments the laser field intensity was much higher than in that of Zeldovich *et al.*[3] As a consequence the divergence of the laser beam was much larger and the transmitted light showed a characteristic ring structure. Furthermore the intensity in our case was high enough to induce a Fredericks transition with a definite threshold in a homeotropic sample with normal incidence of the laser beam.

In the experimental part of our paper we describe in detail our findings on MBBA and OCB liquid crystalline samples in different geometries. In particular these investigations show that in some geometries, where by a simple analogy to the Fredericks transition a threshold effect could be expected, the laser beam does not produce any effect. In the theoretical part we show that the key for understanding the difference between the different geometries is to describe properly the propagation of light through the nematic cell. This can be done in most cases by using the "adiabatic" approximation, i.e. by considering the ordinary and extraordinary component of the light to be uncoupled. As shown in the paper this approximation provides a simple explanation of the absence of reorientation effects in certain geometries.

In the present paper we mention only briefly some quantitative data. Detailed quantitative analysis is postponed to forthcoming papers.

EXPERIMENTAL

In our experiments the beam of an argon ion laser with Gaussian intensity distribution (TEM_{00} mode) and of 515 nm or 488 nm wavelength was focused into the sample with a lens to a spot of $50 \div 60$ μm diameter. The plane of polarization of the linearly polarized incident beam could be changed continuously by a Spectra Physics Model 310-21 polarization rotator, while circularly polarized beam was produced by inserting a Fresnel prism into the beam path. The transmitted beam structure was studied on an opaque screen at a distance of about 20 cm.

The samples had a sandwich like cell structure; a thin layer of MBBA or OCB was filled between two glass plates separated with a teflon spacer. The thickness of the teflon sheet amounted 50–150 μm. The planar orientation was achieved by oblique deposition of a thin SiO film onto the internal glass surfaces. For getting homeotropic orientation the glass surfaces were immersed into a weak solution of CTAB (cetyltrimethylammonium bromide).[4] The monodomain structure was tested carefully at each cell by the conoscopic method.

The experiments were carried out partly at 36–37°C (4–5°C below the nematic-isotropic tansition temperature), partly near the room temperature (21–22°C).

RESULTS

The main features of the effect observed by us in the transmitted laser beam can be described as follows: At very low laser power level, the divergence and intensity distribution of the laser beam remains unchanged. By increasing the laser power, however, the beam divergence begins to increase rapidly and a complex fringe system appears in the beam.

The effect strongly depends on the laser power, on the angle α between L and k (L being the director of the cell and k the wave vector of the beam) and on E (the polarization direction of the incident beam) but does not depend significantly on the wavelength of the beam, on the cell temperature (in the middle of the nematic range), and on the NLC material.

Homeotropic cells (Surface orientation in the Z direction) We take the (x,y) plane parallel to the cell boundaries (vertical plane), and the light beam with wave vector k propagates in the (x,z) plane (horizontal plane), α denotes the angle between k and L (Figure 1).

(1) If $k \nparallel L$ (typically $\alpha \sim 30 \div 45°$) and E horizontal then the increase of the divergence and the fringe system *appeared without any threshold* it was observable already at 10–20 mW laser power.

At a fixed angle α, with increasing laser power the divergence and the fringe number increased (Figure 2). Above a certain laser power (50–150 mW, depending on α) this increase weakens, saturation occurs and the fringes get blurred.

A similar process takes place, if at a fixed power level P the angle α is increased ($10° < \alpha < 45°$); the only difference is, that the power scale goes down and saturation occurs at lower laser levels.

The fringes have a more or less pronounced ellipticity; their size in the y direction is larger with about $10 \div 40\%$. Both the increase of the divergence

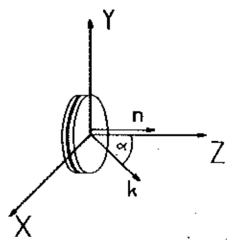

FIGURE 1 Cell position and beam propagation scheme. n normal vector of the cell, k wave vector of the beam. The beam propagates always in the (x,z) plane (horizontal plane).

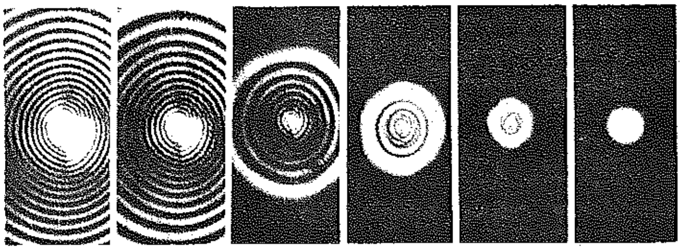

FIGURE 2 Fringe patterns from OCB, at different incident laser powers (Homeopolar cell, $\alpha = 30°$).

and the fringe pattern appear with a time delay T_d. This T_d strongly depends not only on α and P, but also on the temperature, thickness and orientation of the sample. The order of magnitude of T_d is 10 s $\div$ 5 min. After the appearance of the first fringe, the whole pattern develops more quickly, this stabilization time T_{st} being 2 s $\div$ 2 min. The higher is the temperature and the larger is the angle α, the shorter are T_d and T_{st}.

(2) If $k \nparallel L$, ($\alpha > 10°$) and E vertical, neither a significant increase of the divergence, nor fringes were observed, till about 200 mW laser powers. (At high powers, occasionally some deformation of the beam could be seen.)

(3) If $k \parallel L$ ($\alpha = 0$), the effect had a definite *threshold* power, which proved to be *independent* from the direction of E (i.e. from the polarization direction of the incident linearly polarized light beam). In the case of MBBA, thickness 150 μm, temperature 21,5°C, and at a spot size of 65 μm, the measured threshold was 45 $\pm$ 10 mW.

Otherwise the features of the fringe pattern were similar to that of the Case 1. The ellipticity of the fringes always followed the actual direction of the incident beam polarization.

For achieving reproducible results, a well oriented sample, accurate adjusting of the $\alpha = 0$ position and much patience was needed, then at room temperature and near the threshold the first ring appeared only after several minutes.

In this case and near the threshold the pattern was not very stable; it jumped large and then again disappeared.

Experiments with circularly polarized laser light gave remarkable results. In MBBA, 150 μm and 21,5°C, by accurately adjusting the $\alpha = 0$ position, no effect could be observed till to about 150 mW laser powers. But if the sample was first illuminated with linearly polarized light of about 150 mW power resulting in a large divergence and a great number of rings, and then the linear polarization was turned to the circular one; in this case the ring system—though reduced in its size—remained and a well defined threshold intensity could be measured. It amounted 80 ± 10 mW, roughly two times higher, than the threshold at linear polarization. The effect appeared at any initiating polarization direction and as before the ellipticity of the fringes, produced by circularly polarized light, followed the first illuminating polarization direction.

Planar cells (surface orientation in the x or y direction) We choose the coordinate system and the beam propagating direction as before and n denotes the normal vector of the cell, ($n \parallel z$, and $L \perp n$).

The measurements were carried out mostly with an MBBA cell thickness 180 μm at 21,5°C.

(1) If $kL = 0$, (normal incidence) there was no effect independently from the direction of E, up to about 150 mW. At high power levels, some deformation of the beam could only be observed. (Figure 3a)

(2) If $kL \neq 0$, $L \parallel x$ and E is the (x, z) plane, (oblique incidence, L and E horizontal), the fringes appeared without threshold, though the number of fringes were less and they were more blurred, than those in the corresponding homeotrop case. (Figure 3b). The number of fringes at saturation increased with increasing angle of incidence.

(3) If $kL \neq 0$, $L \parallel x$, but $E \parallel y$ (oblique incidence, L horizontal, E vertical), no effect could be observed up to about 150 mW.

(4) If $kL \neq 0$, $L \parallel y$ (oblique incidence, L vertical) there was no effect, independently from the direction of E, up to 150 mW.

THERMAL EFFECTS

As it was already mentioned, at high power levels, occasionally small deformations of the beam (dark spot in the middle of the beam, some rings across

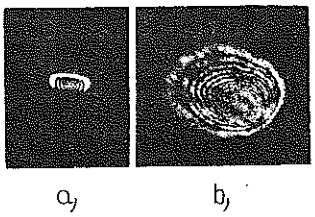

FIGURE 3 (a) Thermal effect in MBBA (planar cell, normal incidence, both L and E are vertical, laser power 120 mW). (b) Fringe pattern from MBBA (planar cell, both L and E are horizontal, $\alpha = 52°$, laser power 25 mW).

the beam, a weak halo around the central spot, or a little increase of the divergence) could be observed. These can be attributed to thermal effects.

A more pronounced thermal effect was found in a homeotropic MBBA cell after strong illumination (over 100 mW) for several hours. The fringe pattern has been changed essentially: a strong broad ring system appeared, which was overlapped by a weaker system with equidistant fringes.[5] The change of the pattern can be explained by increased heating of the sample, which could be a consequence of an increased absorption due to the decomposition of MBBA molecules. Especially the appearance of the weak equidistant fringes can be explained by diffraction on a small bubble of isotropic liquid in the middle of the illuminated spot. This assumption is supported by the fact, that the distances between the adjacent fringes decreases with increasing intensity.

THEORETICAL CONSIDERATIONS

The observations described in the previous section, which are supposed not to be of thermal origin, can be explained with the assumption, that a Fredericks transition takes place here, which is induced by the electric field of a laser beam. The increase of the divergence and the complex fringe pattern observed in the transmitted beam is then a consequence of the accompanying local change of the refractive index which thus reflects the spatial intensity distribution of the inducing laser beam.

For the interpretation of the phenomena, we investigate the sandwich cell of a nematic liquid crystal, in which the director L is fixed uniformly at the boundaries. In the absence of external fields the molecules are arranged in a monodomain structure, determined by the molecular alignment at the cell boundaries. We discuss in the following that in what cases can it be possible to deform the monodomain structure by a light beam, passing through the cell. The light beam interacts with the molecules through it's electric field. The torque $\Gamma^{(E)}$ induced by an electric field E in a nematic is[1]

$$\Gamma^{(E)} = D \times E = \epsilon_0 \epsilon_a E L \, (E \times L) \tag{1}$$

where ϵ_a is the anisotropy of the dielectric constants. For the optical frequencies studied here $\epsilon_a = n_e^2 - n_o^2$ (n_e, n_o extraordinary and ordinary refractive indices).

The light beam can be decomposed at every point inside the cell into an ordinary and extraordinary component. In the ordinary component the electric field is perpendicular to the plane determined by the director L and the wave vector k, while in the extraordinary component the electric field lies in this plane. From Eq. 1 it follows that torque is induced only by the extraordinary component, because for the ordinary one $EL = 0$.

In the cases, when $L \not\parallel k$ (the director and the wave vector of the light beam are not parallel or nearly parallel) the light propagates "adiabatically" in the cell, i.e. the ordinary and extraordinary components propagate independently.[6,7] This is true both for the undeformed and the deformed configuration provided that the director does not vary much at distances comparable with the wavelength of light. From the above considerations we conclude that *deformation can occur only if there is an extraordinary component at the boundary, where the light enters the cell.* Should the "input" be purely ordinary, the light remains ordinary inside the whole cell and it does not induce any deforming torque.

Let us consider in detail the different cases. We use the same notation here as in the previous section.

(1) *Homeotropic cell* (surface orientation in z direction). For oblique incidence the extraordinary input is the horizontal polarization; it induces deformation without threshold. The vertical polarization is the ordinary input which does not deform the orientation. As the incidence approaches to normal, L and k become parallel, and the ordinary and extraordinary components become coupled. In this case we may expect deformation with ordinary incidence too. For strictly normal incidence ($k \parallel L$) there is no deforming torque in the unperturbed structure since in this case $E \perp L$ for arbitrary polarization. However for $\epsilon_a > 0$ i.e. $n_e > n_o$ (this is the case for most of the nematics including MBBA and OCB) the homeotropic alignment is only stable below a threshold intensity, just as in the case of the electric or magnetic field induced Fredericks transition. For $\epsilon_a < 0$/ e.g. for disk like nematics[8]/the homeotropic alignment is stable for arbitrary intensity.

(2) *Planar cell with surface orientation in x direction* The ordinary input corresponds to vertically polarized light beam (E in y direction). At this polarization no deformation can be induced. Deformation can occur with horizontal polarization (E in x direction). With oblique incidence the deformation develops without threshold. At normal incidence there is again no torque in the underformed configuration since $E \parallel L$. The planar structure is stable for $\epsilon_a > 0$ and unstable above a threshold intensity for $\epsilon_a < 0$.

(3) *Planar cell with surface orientation in y direction* Now the horizontal polarization is the ordinary input which does not deform the planar structure. For extraordinary input (vertical polarization) we have for both normal and oblique incidence $E \parallel L$, and therefore no deformation takes place again. As above, for $\epsilon_a > 0$ the planar structure is stable; for $\epsilon_a < 0$ it becomes unstable above a threshold intensity.

The above cases have been studied experimentally (see previous section) and the observations were in full agreement with the theoretical predictions.

In addition we mention two quantitative results without going into details. A calculation, taking into account the finite size of the focused light spot in the sample,[9] gives for the homeotropic cell, and at normal incidence (MBBA, thickness 150 μm, Gaussian intensity distribution, with 65 μm FWHM in the light spot) a threshold intensity value of 35 mW, which is in good agreement with the observed 45 ± 10 mW value.

The other result is, that using the selffocusing model, with simple considerations, the maximum number of fringes can also be estimated. The calculation gives:[10]

$$N \sim (n_e - n_o)\frac{l}{\lambda} \tag{2}$$

where l is the sample thickness, and λ is the wavelength of the light. Taking $l = 150\ \mu$m, $\lambda = 0{,}5\ \mu$m and $n_e - n_o \approx 0{,}15$ we get $N \approx 45$ which is again in fairly good agreement with the observed fringe numbers (38–40 fringes) at high laser powers.

CONCLUSION

In the work presented here we demonstrated the possibility of inducing Fredericks transition with light fields. The effect shows some analogy to the well known Fredericks effect produced by quasistatic fields, however there are some marked deviations too. The most important reason of these deviations is that for quasistatic fields the wavelength of the corresponding electro-magnetic field is much larger than the sample thickness ($\lambda >> l$); on the other hand light waves correspond to the opposite limit, $l >> \lambda$.

An excellent example of the difference between the two cases is given by a planar cell with surface orientation in x direction. Applying a *quasistatic* electric field in the y direction we get a deformation above a threshold field; on the other hand a *light beam* polarized in y direction does not deform the planar orientation at any intensity.

As demonstrated if k and L are not parallel or nearly parallel the dependence of the deformation on the direction of polarization can be explained by taking into account the adiabatic propagation of light. It remains however to understand the behavior of NLC-s at small angles between k and L, including the case of circularly polarized light beam. Another problem is to separate electric field effects from thermal ones, which may become important near the clearing point of nematics.

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