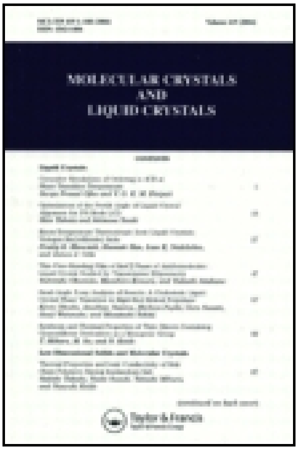

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NONLINEAR OPTICAL EXPERIMENTS ON LIQUID CRYSTAL CHIRAL STRUCTURES

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Abstract Up to now there are only few reports in the literature on experimental examination of nonlinear effects in chiral liquid crystals. The presented data are sometimes confusing. We proposed a simple experiment in a probe+pump laser technics for studying periodic liquid crystalline structures. The paper presents initial results of examination of a cholesteric material in the region of selective light reflection. We have been interested if any nonlinear effect of the collective nature appeared by usually accessible light intensities, i.e. non-thermal but of optical reorientation origin. The investigation was oriented toward potential application in all-optical light modulators.

Key words: optical nonlinearity, liquid crystals, chiral nematics

INTRODUCTION

Nonlinear optical properties of liquid crystal (lc) chiral structures (i.e. chiral nematics and smectics) are relatively weakly recognised, although they are investigated over twenty years[1÷3]. The examination of those media, so theoretical as well as experimental, meets much more difficulties as in the case of simpler nematic structures. From one side it is caused by more complex structures of cholesterics and smectics, but also by significantly higher light intensities for generating the nonlinear optical response (as expected from theoretical evaluations) on the other hand.
Nevertheless, the results of theoretical and experimental works done in the past, mainly for cholesterics, revealed a series of nonlinear effects, as intensity-dependent optical activity and polarisation relations of the propagating light components, nonlinear changes in Bragg ("selective") reflection, induced nonlinear light scattering, self-diffraction effects, etc.[4÷8]. Illustrative view of distortion of cholesteric helix in a static field F is shown in fig.1; similar effect can be expected to occur also in the optical field.

The main difficulty in the nonlinear experiments with those media is the problem of registration of week changes in an intense laser beam activating the effect. Additional

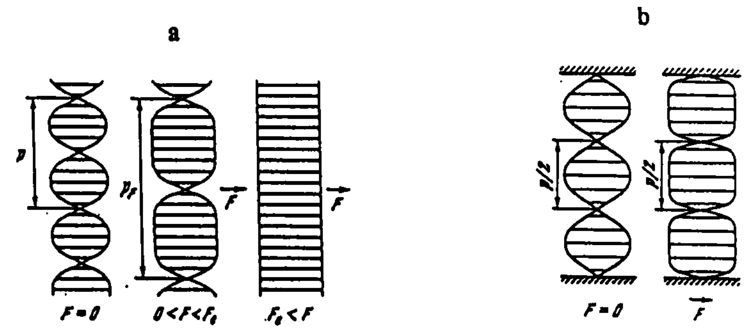

FIGURE 1. Illustrative representation of cholesteric structure deformation under a static field F; a: for unlimited helix, b: for thin cholesteric layer with imposed boundary orientation

problems deliver strong parasite reflections from glass holder of the lc material and usual necessity of focusing the light beam, which then impinges on the structure at various angles in a cone, darkening the physical picture. In such a cases usually the pump+probe beam technics is used. This is normal procedure of recording of small optical changes of a medium under intense illumination. We have used this technics for examination of periodic lc structures in the reflection mode of the probe beam positioned at the Bragg scattering angle. In the experiment the periodicity changes of a cholesteric structure under 1 W-argon laser beam were measured in the region of selective light reflection.

EXPERIMENT

The experimental cells were made as typical sandwich cells with Grandjean - planar alignment and thicknesses varying from 10 μm to 25 μm. The glass plates constrained the lc layer were covered by polyimide surfactant without imposing any preferred surface orientation. They were prepared from semi-heavy glass material (n=1.59) to avoid additional ordinary reflections at the glass - cholesteric interface. The examined cholesteric phase was formed in an isothiocyanate nematic mixture denoted as 979A[12], doped with a chiral right-handed (optically active dextro) additive[13] to fit the pitch p=510 nm to the pump laser wavelength.

The geometry of the experimental setup is shown in fig.2. The intense argon laser beam is thrown at normal incidence on the planar sample. The beam passes through λ/4 plate controlling its polarisation and is focused to about 0.2 mm^2 spot. The normal incidence of the pump beam is essential for theoretical analysis of its interaction with lc structure, since there are exact solutions for propagating light in this case. The linearly polarised probe beam from laser diode λ=670nm enlightens obliquely the activated area of the sample through the polarisation rotator (P + λ/2) and, after cutting off parasite side reflections, the part of it reflected by the cholesteric layer is recorded by the photomultiplier. The angle of incidence Θ, equal the angle of reflection was fixed in the Bragg scattering region (1/2 or 1/3 reflection order).

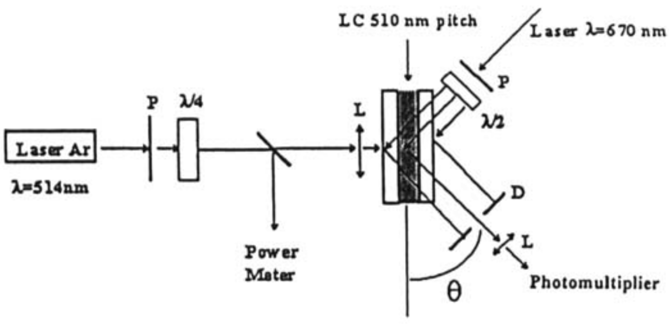

FIGURE 2. The experimental arrangement; Ar - pump argon laser, P - polariser, λ/2 and λ/4 - half-wave and quarter-wave plates, L - lens, D - diaphragm, LC - cholesteric liquid crystal cell

For interpretation of the measured signal let assume that the pitch of the optically deformed structure increases under illumination. The Θ_{max} obeying exactly Bragg condition is related to the periodicity of the structure p and the probe beam wavelength λ via $p \sin\Theta_{max} = n\lambda$. For fixed wavelength of the probe beam, what is the case of the experiment, the Bragg angle Θ_{max} must than decrease. Thus, the angular dependence of the Bragg scattering intensity shown in fig 3 shifts for larger pitch p towards lower angle

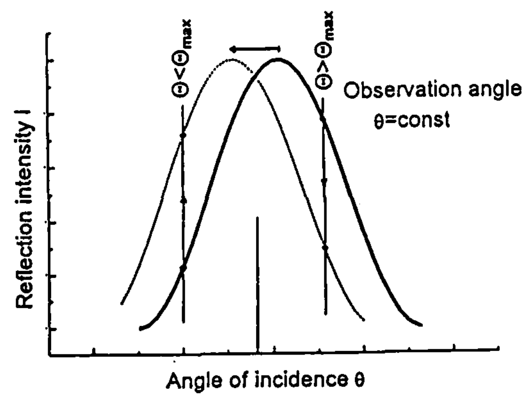

FIGURE 3. The angular intensity dependence of the Bragg scattering

values (i.e. to the left in fig 3). Since in this experiment the probe signal was measured by a fixed angle Θ, the "working point" of the measuring setup moves along continuously shifting curve, as it is pointed in fig.3. It gives the signal increase by $\Theta<\Theta_{max}$ as well as the decrease in the case $\Theta>\Theta_{max}$, both indicating enlargement of the pitch. To determine the pitch as a function of pump beam intensity in this experiment, the values of Θ_{max} should be recorded point after point.

RESULTS AND DISCUSSION

Fig.4 represents the records of the probe signal reflected from the sample versus the pump beam intensity, taken by the angle Θ above and below the Bragg angle Θ_{max}. The data were recorded for several samples of different thicknesses and for various polarisations of the pump beam. The samples always showed the increase of the pitch with rising light intensity and, apart from a certain differences in magnitude, equal behaviour for any polarisation of the pump beam. From the plots in fig.4 following can be also noticed:

- relatively low light intensities inducing structural changes, what is in contrast to the previous theoretical predictions [1],
- lack of any intensity threshold value in the effect,
- different contributions to the effect from the right circular polarisation of the pump beam almost totally reflected and from the left one, almost fully transmitted, what is in accordance with theoretical suggestions [1, 4].

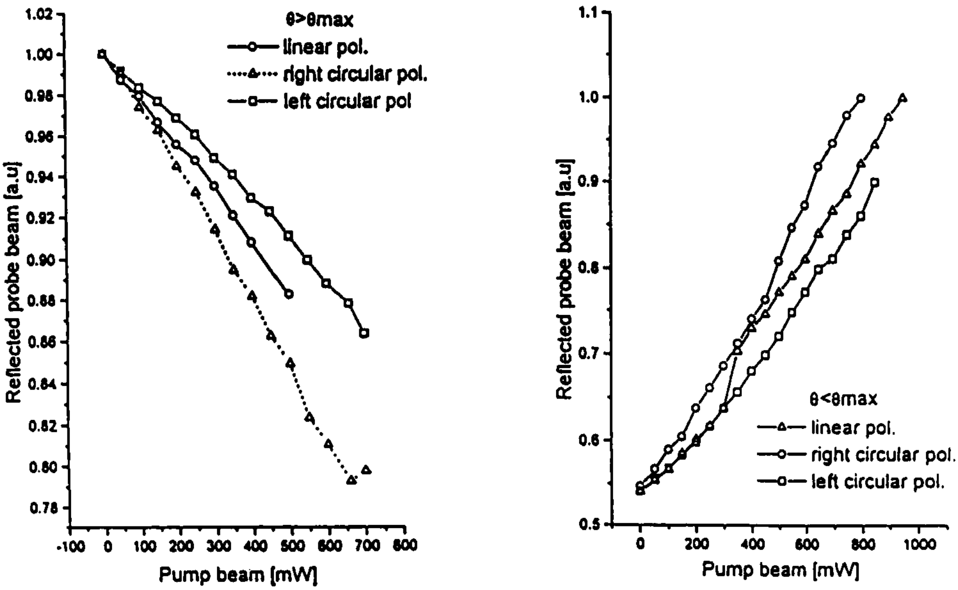

FIGURE 4. The probe beam intensity versus pump beam intensity for observation angles Θ above and below the Bragg angle Θ_{max}

Before continuing the experiment, for further discussion of the results is necessary to determine which mechanism - thermal or orientational - is responsible for what we have observed. Unfortunately, the p(T) behaviour of this particular lc material we used in the experiment is similar to p(I), i.e. increased temperature elongates pitch p, so as it is expected also for optical electric field (see fig.1). However, for optical reorientation, theoretical considerations predict a linear pitch - intensity dependence [9] :

$$\delta p/p_o = k L^2 [p_o/\lambda]^2 E^2, \quad \text{with} \quad I \sim E^2,$$

and E as the pump optical field, L-layer thickness, $k=[\varepsilon_a]^2/[64\,\pi K_{22}]$ -material constant,

while thermal behaviour of the pitch should be in general nonlinear through nonlinear functions p(T)[10] and T(I). However for small p-changes observed in the experiment this distinction could be not enough pronounced and would appear only for very precise pitch measurements. Than the separation of thermal and orientational mechanisms seems to be done much simpler by using another lc material with negative thermal coefficient dp/dT which will be opposite to optical dp/dI expected to be positive, at least for wavelength region in Mauguin limit[9]. Although the difference of sensitivity of the observed effect to the both counterpart-polarisations is in agreement with theoretical suggestions for optical reorientation, it does not exclude thermal mechanism, because this feature can be attributed in general also to the latter. Since the initial data do not allow to determine with sufficient certainty what mechanism is involved in the observed effect, we have examined dynamic response of the lc structure to the switching of the illumination. This response is shown in fig 5. The reaction was slow, of the order of seconds. The dynamics of the observed effect coincides with the reaction times for structural changes measured in cholesterics in static electric fields[11]. The same sample, when heated as a whole and than illuminated, showed apparently more pronounced and much faster response.

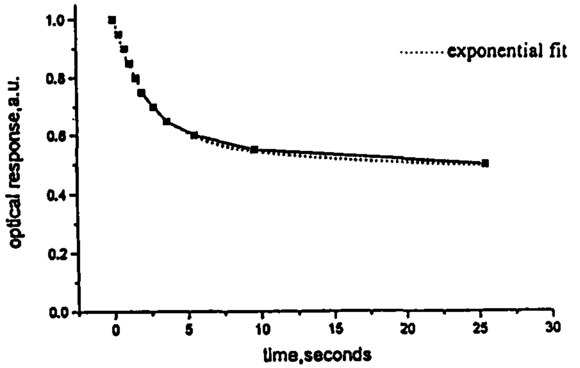

FIGURE 5. The dynamic response of the sample on the pump beam pulse

This stronger and faster dynamic response to an optical pulse in elevated temperature probably results from lower viscosity and weaker elastic torques of lc medium, so indicating more clearly orientational mechanism of the observed deformation. In contrast, thermal sensitivity dp/dT of the pitch changes decreases if T rises[10].

By consideration of molecular picture of structural deformations there is additional complication which must be taken into account for thin lc cholesteric layers with surface orientation imposed by boundaries in a preferred direction, what is the case, to more or less extend, in practice. In the extremal cases structural periodicity could be "frozen" and only a certain deformation could appear (as it is indicated in fig 1b). At this point of the experiment however we need more detailed examination to understand what is going on.

CONCLUSIONS

We have demonstrated the experiment examining the nonlinear optical structure changes in chiral nematics. The presented pump+probe beam technics is simple and sensitive to structural changes in periodic media like cholesterics and smectics . The method allows detection of small pitch changes and molecular reorientation (by polarisation relations of reflected probe beam). We have shown initial results of examination of a cholesteric liquid crystal in the region of selective light reflection. The rise of the pitch in cholesteric thin layers was observed by relatively very low intensities of activating beam, depended on its polarisation. No threshold intensity value was noticed in the effect. The obtained results suggest orientational optical mechanism of liquid crystal structure changes.

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