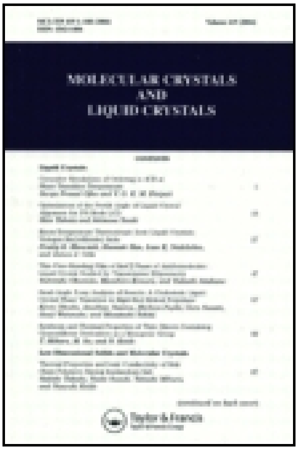

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Study of Structural Optical Nonlinearity in Chiral Nematics by Reflected-Probe-Beam Technique

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Study of Structural Optical Nonlinearity in Chiral Nematics by Reflected-Probe-Beam Technique

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Interaction of intense laser light with chiral nematic structure was examined by use of a reflected-probe-beam experimental technique. The technique is sensitive to structural changes in periodic media and appears to be as valuable for chiral nematics investigation as rentgenografic diffraction for crystal lattices. The paper presents partial results of experiment which is just now continued. They suggest optical nonlinearity in chiral nematics of the collective nature rather than of thermal or photochemical origin. The experimental data obtained up to now are discussed in details, although at present some essential questions remain not clear and therefore no theoretical analysis is given.

Keywords: optical nonlinearity; chiral nematics

INTRODUCTION

The measurements we have done previously[1] showed an interesting response of chiral nematic (CHN) cell on laser illumination. We have studied reaction of its helical structure under illumination of argon laser by reflected-probe-beam technique. In this method the area illuminated by intense laser light, expected to activate a local deformation of structure is additionally enlightened by weak probe-beam at a glancing angle close to Bragg reflection. Reflexes of the probe beam were measured and analysed for various sample parameters and configurations in order to obtain data concerning behaviour of CHN structure in optical field. The observed reaction of CHN cells as detected by the technique used in the experiment started in certain conditions without any noticeable threshold at low activating light intensity. As we believed we have observed optically induced changes of the helix pitch and accompanying optical effects. For measured signal varied almost linearly in certain range of laser power and had good repeatability we have even suggested the possibility of using this effect and detecting method for light intensity measurements[2]. Since our recent report on this observations the experiment was continued. But the more new observations we made, the more questions appeared. Some of them were quite basic:

1. does the probe beam really detect helical pitch changes of the structure or it is simply reflected from the layer boundaries,

2. what kind of nonlinearity we have to do with or which prevails at least - thermal, which is optically non-specific or orientational,

3. why the reaction of CHN structure starts at relatively low power often without threshold, while theoretical considerations[3] are in contrast to that statements.

EXPERIMENT

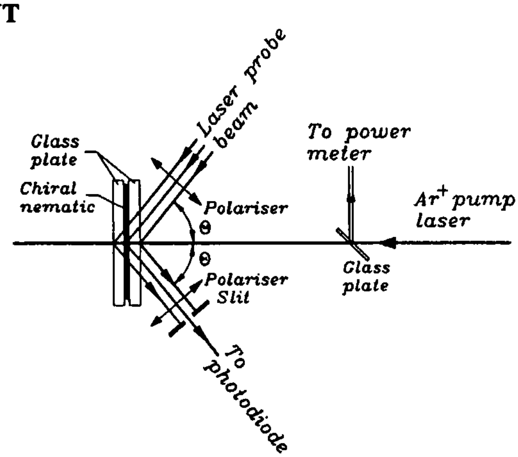

FIGURE 1, Experimental optical configuration

Continuing the experiment the same optical arrangement was used. The experimental set-up is sketched in Figure 1. Argon laser ("pump" laser) beam impinges at normal incidence on planarly oriented CHN layer to activate structural deformation. We have used the argon laser working on the spectral line 514 μm in power range 0÷1.5W. The beam was neither focused nor formed anyway and had natural linear polarisation. If the optical field of the beam interact with structure of CHN-medium, it will induce local deformation of the CHN-helix and can influence structural periodicity. The second semiconductor laser ("probe laser") enlightens the deformation area at a glancing angle close to the Bragg angle Θ_B. Intensity of reflected probe beam from the CHN structure is measured as the detecting signal. Relatively strong parasite side reflections of the probe beam from glass holder we have cut off by setting into the optical path two polarisers and a slit. So the part of the beam which was reflected from the CHN structure after passing through the slit could be additionally selected due to its elliptic polarisation in contrast to linearly polarised reflexes from the glasses. CHN-substance used in experiment was prepared by MAT[4] as a mixture of nematic cyanobiphenyl basis (CHBT) with chiral dopants having helical structure. Experimental cells were prepared with structural periodicity (pitch p) either close to argon laser wavelength for measurements in selective reflection region or far outside it (p≈1000nm). We have marked them as "resonant samples" and "nonresonant samples", respectively. Examined samples consisted of CHN-layers oriented planarly by standard surface treatment (polyimide coating + rubbing). Thickness of the CHN-layers as imposed by spacers varied from 7μm to 35μm.

RESULTS AND DISCUSSION

The dependence of photodiode signal which correspond to probe-beam intensity on argon laser power activating the effect is exemplified in Figure 2. The reflection of probe beam rose or fell down or the both in a sequence. Apparent hysteresis was observed but not bistability. However measurements of intensity of the transmitted argon laser beam did not reveal any nonlinearity, neither for nonresonant samples, even nor for resonant ones.

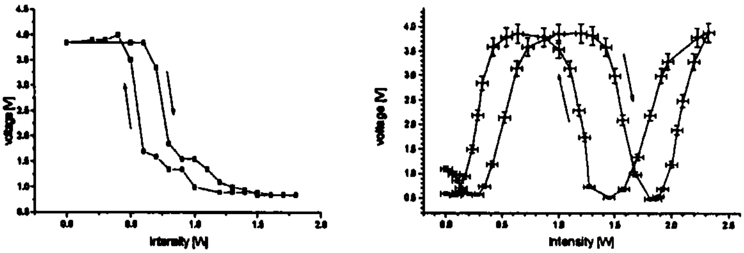

FIGURE 2, Reflected probe-beam intensity (= photodiode voltage V) versus pump beam power for : a) nonresonant sample ($p \neq \lambda_{pump}$) and b) resonant sample ($p \approx \lambda_{pump}$) - full cycle.

Assuming optically induced pitch changes of CHN structure, observed probe-beam variations could be explained by analogy to the X-ray scattering theory in crystals[5]. However, the effect can have also another origin. As it was mentioned, optical path of probe beam was matched to fulfil the Bragg condition. This condition is equivalent also with requirement for constructive interference of two reflexes from the both cell glasses, since their distance d (=thickness of CHN layer) is a multiplication of the helix pitch p. Such a interferometer works nonlinearly in regard of liquid crystal medium inside the resonator, where optically induced changes in effective index of refraction can shift its Airy function. Thus, even if the probe beam does not "see" the structure of the CHN layer, reflexes from layer boundary could vary with power of incident light in the similar way as it was observed in the experiment.

For verifying our previous interpretation and for checking which mechanism is involved in observed effects, the following has been done:

1. ellipticity of reflected probe beam was examined as a function of glancing angle θ by measuring intensities of the both principal vibrations I_{max}, I_{min}. The results are shown in Figure 3. Clearly, all the dependencies show dramatic changes in the same angular region near Bragg reflection. The both intensities suddenly rise near Bragg angle but the polarisation linearity (being I_{max}/I_{min} ratio, i.e. reciprocal of ellipticity) simultaneously fall down to values indicating that the beam approaches circular polarisation. This behaviour is characteristic

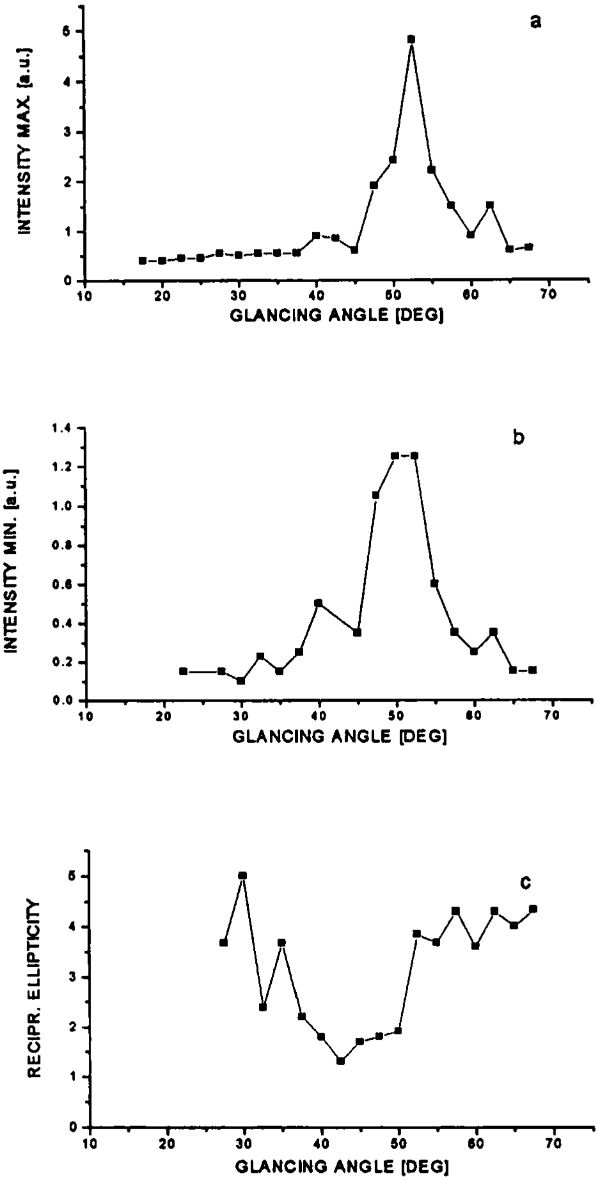

FIGURE 3. Principal components of probe beam a), b), and its ellipticity c) as dependent on angle of reflection from CHN layer.

for selective reflection in CHN structure.

2. ellipticity of argon laser beam obliquely reflected from the CHN layer was measured as a function of its power. The results represents Figure 4a, while Figure 4b illustrates analogous dependence for the same experimental cell but filled with pure CHBT basis, which is pure nematic. The cell was also planarly oriented. The curve in Figure 4b shows that ellipticity of the beam reflected from non-chiral nematic layer is not influenced by illumination and is nearly constant, just in contrast to oscillating dependence for CHN layer. The only exception is a narrow range at low intensity. This difference also suggest the crucial role of the chiral structure by measured reflection.

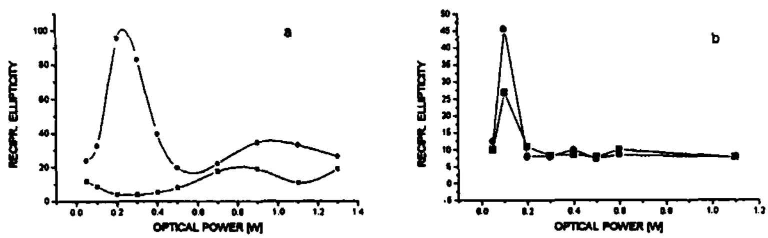

FIGURE 4. Ellipticity of the pump beam obliquely reflected from: a) CHN layer and b) ordinary nematic, versus its power.

The linearity peak in Figure 4b which is seen only for moderate beam intensity can be clearly understood if we consider distortion which can be caused by linearly polarised light in a planar nematic layer. The only deformation in the experimental configuration can be twist. It can appear when the initial angle between direction of optical field and principal axes of nematic is non-zero. For very low light intensities, if no deformation occurs, each of the both principal polarisation components of the beam are affected at the layer boundary by different refraction indices and so they undergo different reflections, according to Fresnel formulae[6]. Therefore they reflect with different intensities. The effect is repeated at second boundary of the layer, where the components travelling in birefringent medium additionally require a phase shift. After recombination it results in elliptic polarisation of the reflected beam. For higher light intensities in the layer starts twist deformation. Initially the deformation is smooth - i.e. adiabatic, then the beam, partially reflected from the first boundary travels across the layer in conditions known as a Mauguin limit[7]. Thus the polarisation directions of the two vibrations rotate with the principal axes of the deformed structure and reach the rear layer boundary with azimuths closer to the axes. It changes reflection at this boundary (one can show that extraordinary vibration will be privileged) and finally affect the ellipticity of the total reflex. However for still higher light intensity optical deformation become sharp; layer goes out of Mauguin limit and light wave can not follow it. Now the nematic layer presents two narrow interfaces near boundaries with dramatically changing molecular orientation separated by the bulk almost homogeneously oriented. This situation persist more or less up to the highest light intensities used. Incident light meets now at the both boundaries new reflection conditions. They change again ellipticity of the reflected light and remain nearly constant in the high intensity range.

In order to avoid uncertainty and suppress eventual addition of "non-structural" interference to the main reflex from CHN structure, a series of measurements was also made for 50-μm thick cells. Moreover, the probe laser was replaced by much less coherent halogen lamp with filters. In this conditions the non-structural interference should have low visibility. As expected, the results of this measurements did not essentially differ from that previously made.

CONCLUSIONS

Taking all above mentioned into account we tend to following interpretation:

1. The reflected probe-beam technique is sensitive to structural changes in chiral liquid crystals,

2. The nonlinear effects observed have orientational origin rather than thermal. It is suggested by two facts: a) reaction of CHN structure is of the order of seconds to tenth of seconds, b) for thin samples (<10μm) with imposed strong surface anchoring no effect was detected.

3. The probe-beam variations measured by relatively low illumination are connected with certain imperfection of ordering in thicker samples. It could be

expected, that they contain tilted-planar layer with helix slightly inclined to the layer normal. Under illumination, at the beginning the probe-beam signal probably detect reorientation of the helix as a whole, which resembles a kind of optical electroclinic effect. Than, for much higher light intensity the helix starts to change its pitch.

However, the results here presented concern to experiment which is still in progress; therefore, now, on the basis of partial data we would be afraid to risk any detailed theoretical analysis, although we have already started to do this in previous publication[5].

Acknowledgments

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