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Dye-enhanced nonlinearity threshold measurements in liquid crystals

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Since early eighties it is well known, that optical field can interact with liquid crystalline structures inducing molecular motion in similar way to quasi-electrostatic fields. This nonlinear optical effect (NOE) causes changes of refractive index distribution in the structure and in this way leads to several optical phenomena, like self-diffraction of the propagating beam, harmonic generation, four-wave mixing, nonlinear wave-guiding, etc.

At first, NOE was observed in nematics for relatively high optical power densities. In the middle of nineties, Istvan Janossy discovered, that small addition of specific dopants to LC-material can significantly reduce the light power required for NOE. At present, NOE can be induced in certain LC-materials by sub-miliwatt laser beams. This achievement opened a way to many applications of the accompanied optical phenomena.

Mechanism of the Janossy effect is still not quite clear. NOE is generally a threshold effect, which means that it starts after a certain threshold value of the optical power. In this work we examine the threshold of the self-diffraction of light in doped nematics in selected conditions, trying to understand its nature. Final goal of the work is examination of NOE in chiral liquid-crystal structures (cholesterics).

Keywords: optical nonlinearity, liquid crystals, Janossy-effect.

1. Introduction

Liquid crystals (LCs) are developed for more than 100 years. During this time many researchers have been working on this subject discovering plenty of fascinating optical and electro-optical phenomena and bringing to light their mechanism. The story of nonlinearity investigation in LCs is not so long, nevertheless it was clear since early eighties of the passed century that optical field of light can interact with LC structure exerting changes similar to those which quasi-electrostatic field does. The phenomenon was called as structural nonlinear optical effect (SNOE).

General understanding of the SNOE is quite obvious. If the electric field is introduced in the structure, it gets additional portion of energy. To minimize it, LC-molecules start to rotate (reorient) collectively distorting the structure. In other words, electric field of the light exerts a torque on anisotropic LCs molecules what causes that they tend to orient along a field vector. The final balance is reached for minimum of total energy and for certain distortion, corresponding to the field strength.

Distortion of an anisotropic structure induces the spatial distribution of refractive index of the medium. This is the main point of the SNOE. However, exact mathematical description of the SNOE is now done only for the simplest cases. Theory predicts differences for particular geometries. For geometry used in our experiment, the SNOE may have a threshold character, what means that it starts above some threshold value of intensity, or can appear without threshold, depending on light polarization [1]. For normal incidence both polarization components are equivalent, and for both cases the threshold exists. Threshold value is then given by LCs material parameters, as (in the case of strong anchoring) [2,3]

$$I_{th} \cong \frac{4\pi^3}{d^2}\left(\frac{K_{33}}{\Delta\varepsilon}\right), \tag{1}$$

where d is the LC-layer thickness, K_{33} is the elastic constant for bend deformation, and $\Delta\varepsilon$ is the LC dielectric anisotropy.

Equation (1) was obtained for pure nematics and it does not take dopants existence into account. However, here, the influence of small amount of additives can be considered through modification of the elastic constant value K_{33}.

For an oblique incidence, one polarization component of the light yields threshold, while the others do not. In this case, for various polarization states the SNOE may appear for lower or higher intensity of the incident light, i.e., changes of the threshold value can be observed. This changes are importan thint-mark of the SNOE, since the structural nonlinearity is always accompanied more or less by the thermal nonlinearity. These two effects are hardly separable since a part of passing light is always absorbed and local heating by the light beam gives similar optical effects as structural nonlinearity [2,4].

LC-nonlinearity is experimentally demonstrated by several optical effects. The simplest one for observation is the

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self-focusing of the passing light beam. It occurs when the light beam has non-homogeneous power density distribution in the cross-section (x,y-plane). It causes non-homogeneous distribution of the refracting index $n(x,y)$, what results in spatial phase shift, and diffraction or refraction of the light.

The phase change of the light wave travelling along z-axis in a medium depends on $n(z)$

$$\varphi = \int_0^d \frac{2\pi}{\lambda} n(z)dz. \tag{2}$$

For the Gaussian laser beam

$$I_{opt} = I(r) = I_0 \exp(-2r^2/w_0^2); \quad r^2 = x^2 + y^2, \tag{3}$$

the phase distribution is also Gaussian and the geometry has axial symmetry, i.e., $n = n(r)$, $\varphi = \varphi(r)$ [2]

$$\varphi(r) = \int_0^d \frac{2\pi}{\lambda} n(r,z)dz.$$

Diffracted light interferes giving a pattern of concentric rings in the far-field (see Fig. 1). Obviously, the diffraction pattern depends on the light intensity.

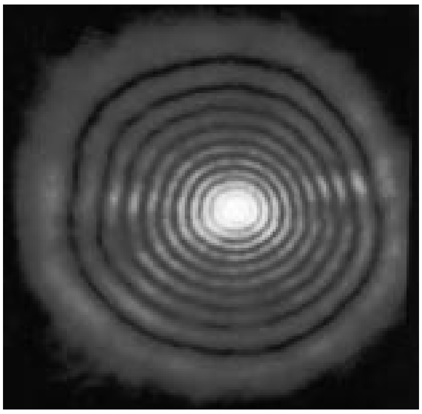

Fig. 1. Self-diffraction effect in homeotropic nematic layer.

2. Experiment

In our measurements we used the effect of self-focusing. We examined the threshold values of the optical nonlinearity using self-diffraction of the light in pure and doped nematics in selected conditions. The experimental set-up is shown in Fig. 2.

Argon laser beam of the wavelength λ = 514 nm passed through intensity controller and polarizer. The beam was split and its small part, controlled part was measured by the DetI detector. The signal from DetI was consequently denoted as light power. The main part of the beam was focused directly on the examined LC-cell in the experiments with linearly polarized light, while in the case of circular polarization a quarter-wave plate was additionally used. The diffracted beam coming out from the LC-cell was projected on a screen; in the centre of diffraction area (i.e., in the point where the first diffraction ring starts to arise) DetO detector was placed for measuring of the output power. In our experiment, the threshold values for the formation of the intensity-dependent diffraction pattern was measured. Characteristic experimental plot of the both DetI and DetO signals (i.e., light and output power, respectively) during one measurement cycle is presented in Fig. 3.

The solid line is the DetI-record and shows the programmed cycle of monotonically increasing and decreasing the laser-beam power. Dots (DetO-record) represent power of the beam passed through the LC-cell, where the decrease in the output light power corresponds to the formation of

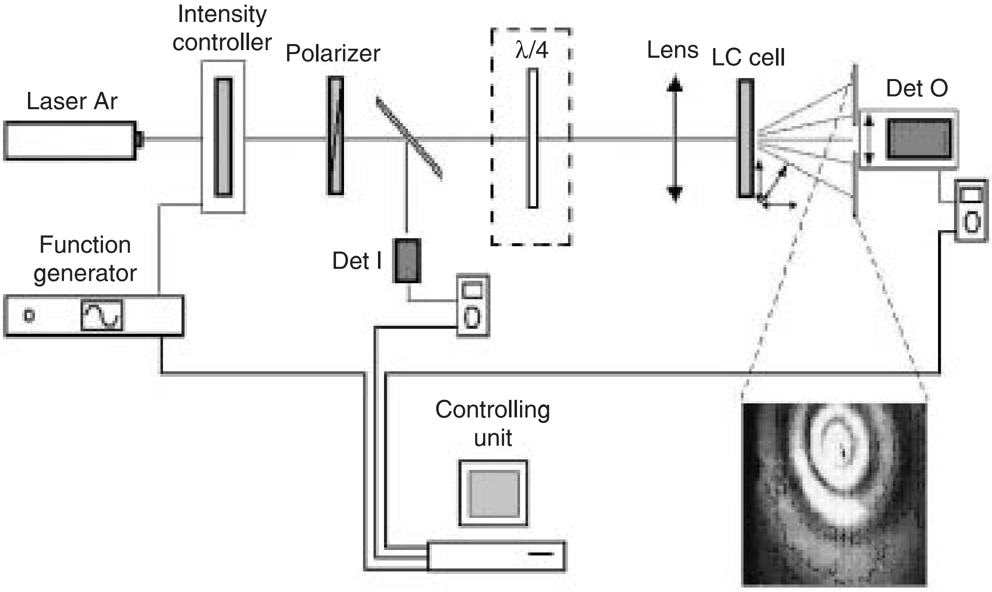

Fig. 2. Measurement set-up.

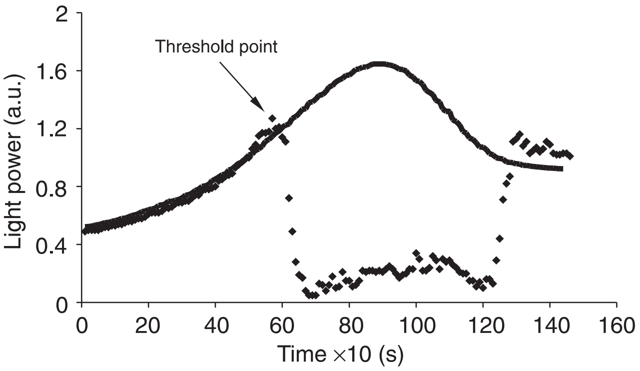

Fig. 3. Experimental data showing the threshold point of the SNOE: input light power (solid line) and output light power (dots).

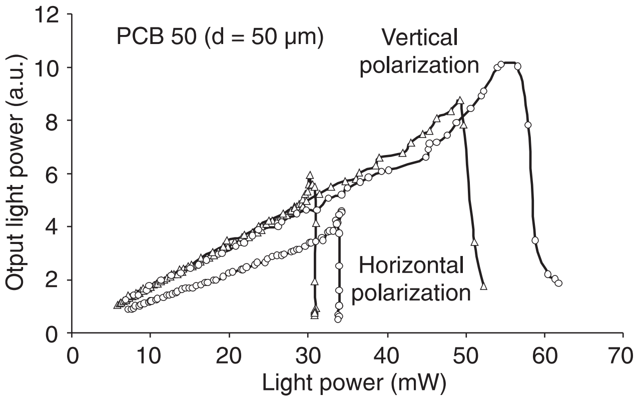

Fig. 5. Optical reorientation for linearly polarized light: dependence on the polarization plane; triangles and circles correspond to different measurements for two similar LC-cells.

the first diffraction ring and gives the threshold value of the SNOE.

Experimental samples were typical liquid-crystalline sandwich cells with the homeotropic alignment. Orientation of LC-layer was imposed by surfactants, either lecithin or commercial polyimides were used. The thickness of the LC-layers ranged from 20 μm to 100 μm for various samples.

The measurements were carried out using standard nematics PCB and 6CHBT. For Janossy-effect, they were doped with dye materials having similar molecular structures but different colour centres. The dyes are shown in Fig. 4.

The doped samples were prepared with saturated solutions of dyes in nematic materials at 22°C. It gave concentrations of the dyes of the order of 1%. Spectrometric measurements showed 70% absorption for the red and 2% for the blue dye-doped samples (6CHBT- 50-μm thick) for the used laser wavelength of 514 nm.

3. Results and discussion

Typical results obtained for 50-μm cells filled with pure nematic PCB and for the linearly polarized light of various polarization planes are presented in Fig. 5, while for the circular polarization in Fig. 6.

Although LC-cells were prepared as homeotropic, the threshold strongly depends on the plane of polarization (Fig. 5). Average threshold values vary up to 70% for different polarization planes. It can derive from the presence of small initial tilt of the LC-molecules at the boundary plates. Other measurements, not shown in this paper, indicate that such changes of a threshold value can be observed for small changes of tilt. This behaviour indicates the reorientational, but not thermal nature of the observed nonlinearity, because power density of light beam by different polarization planes remains constant and no pleochroism was observed.

Similar conclusion can be obtained basing on the results obtained for circularly polarized light, as illustrated in Fig. 6.

The threshold values for the circularly polarized light are more than twice higher than for the linear polarization by the equal beam power. On one hand it is caused by the fact that reorientational nonlinearity is affected not by total power of beam but by the intensity of the optical field in the certain plane, and on the other hand by independence principle, i.e., by the principle which says that the movements (molecular rotation) in orthogonal planes are independent. It also means that two light beams polarized in orthogonal planes do not enhance each other to induce the SNOE. Through the circular polarization here provided the same light power as the linear one, in the former case due to the quarter wave plate, the optical field intensity in any fixed direction is

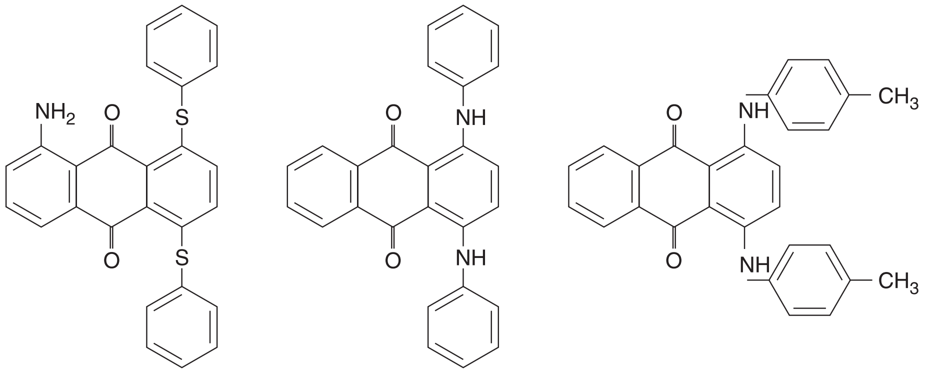

Fig. 4. The three of dye-dopants used in experiment.

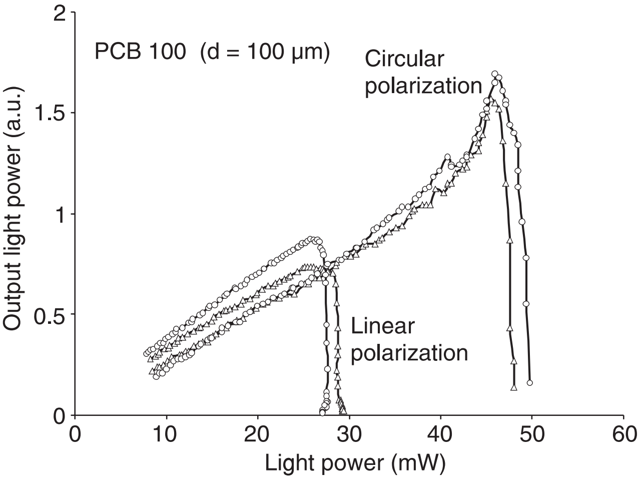

Fig. 6. Optical reorientation: comparison of the circular and linear light polarization. Triangles and circles correspond to different measurements for two similar LC-cells.

$(1/\sqrt{2})E_{linear}$ and the mean power is then $P_{linear}/2$. It requires two times higher power to induce the molecular rotation in any plane. Again, these results indicate the reorientational nonlinearity, as thermal effect is not orthogonally independent, the partial effects induced by the different polarization components summarize. These statements are important hints for SNEO in chiral LCs structures.

Examination of dye-enhanced nonlinearity (Janossy-effect) shows, in general, for given configuration that, the starker is enhancement, the weaker are surface interactions of LC-layer. It means that for strong anchoring as well as for thin LC-layers SNOE threshold-changes are less distinct than for week anchoring or thicker layers.

We observed decrease in the threshold value of about one order of magnitude for the examined dye additives as it is shown in Table 1.

Table 1. Optical threshold values for various samples, (s.s) – saturated solution of dye and (p.c.) – weight concentration of dye.

Configuration	Threshold power P_{th} (mW)	Intensity I_{th} (MW/m^2)
6CHBT 50	12.7	96
6CHBT 50 + red 916 (s.s.)	1.7	13
6CHBT 50 + blue 917 (s.s.)	3.4	26
6CHBT 50 + brown 921 (s.s.)	2.9	21
6CHBT 50 + red 2909 (p.c. 2,57%)	9.0	67
6CHBT 50 + blue 2590 (p.c. 1%)	3.4	26
6CHBT 50 + violet LC13 (p.c. 1%)	1.3	10
6CHBT 50 + violet LC13 (p.c. 1.88%)	1.2	9

Although these dyes are not particularly effective in Janossy-effect, they enable an observation of some details of the analysed mechanism. Among the investigated dyes, the three mentioned above – red, brown and blue – reduce threshold value almost by the same degree, approx. 13:2, 13:3, and 13:3, respectively. These dyes have different absorption for the wavelength used in the experiment, but they have similar structures and solubility [5]. In this way we can assume important role of steric properties of the dye molecules.

Our results point out also negligible influence of the thermal mechanism on the observed effect, regarding that absorption relation for example red/blue is greater than 35:1 by similar threshold reduction.

One of the possible modification processes of optical transition by dopants, where a molecular shape is crucial but bulk concentration is secondary, is adsorption of solved molecules at LC-layer boundaries. This can result in changes of interfacial energy, in other words resolved dyes can act as surfactants in an LC-cell [6]. However, such a mechanism does not seem to be observed in our samples. If it took place, one could expect greater threshold reduction for thinner samples and for strong anchoring, what is just in contrast with ours observations. Ours results (above mentioned as well as those unpublished) rather suggest Janossy-effect to be a bulk effect. Threshold Eq. (1) shows, that a bulk process, reducing threshold value, could decrease the elastic constants of LCs by dopants existence. However, it is only a general, phenomenological description of LC molecular interactions [7] and actually does not explain anything. In literature [8–15] there are some concepts trying to explain how it happens. Presumably Janosssy effect involves more than a single mechanism. Its complex dependence on the dye concentration supports this possibility. This problem still needs more work to be solved [5].

4. Conclusions

In the work, the nonlinear optical effect in nematic liquid-crystal layers was examined for various polarizations of the incident light.

It was shown, that such measurements could help to separate effects of the thermal and the reorientational nonlinearities. The obtained results suggest also, that dye-enhancement of reorientational nonlinearity in LCs is a bulk effect and is controlled apparently by the steric properties of dopant molecules. The complex dependence of the effect on the dopant concentration suggests, in our opinion, more than one single mechanism of the effect.

Acknowledgements

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References

1. M. Sierakowski, "Nonlinear liquid-crystalline optical cells", *Optica Appl.* **24**, 219–224 (1994).
2. I.C. Khoo and S.T. Woo, in *Optics and Nonlinear Optics of Liquid Crystals*, Series in nonlinear optics – Vol. 1, World Scientific Publishing, 1993.
3. C.M. Arakelyan and Yu.C. Tchilingaryan, *Nieliniennya Optika Zhidkich Kristallov*, Nauka, Moskva. (in Russian)
4. L. Marrucci, "Mechanism of giant optical nonlinearity in light absorbing liquid crystals", *Liquid Crystals Today* **11**, 6–33 (2002).
5. Even if the concentration of the dyes is a little different, the differences do not have to be concerned because in our experiment we noticed week sensitivity of the observed effect to the dye concentration in the examined range.
6. It should also result in reduction of the threshold of electro-optical Frederiks transition.
7. The analogical action of the chiral additives on molecular structure takes place by the forming of the cholesteric structure from the nematic phase.
8. I. Jánossy, "Molecular interpretation of the absorption-induced optical reorientation of nematic liquid crystals", *Phys. Rev.* **E49**, 2957–2963 (1994).
9. I. Janossy and A.D. Lloyd, "Low power reorientation in dyed nematics", *Mol. Cryst. Liq. Cryst.* **203**, 77–84 (1991).
10. L. Marrucci, D. Paparo, M.R. Vetrano, M. Colicchio, E. Santamato, and G. Viscardi, "Role of dye structure in photoinduced reorientation of dye-doped liquid crystals", *J. Chem. Phys.* **113**, 10361–10366 (2000).
11. L. Szabados, I. Janossy, and T. Kosa, "Laser induced bulk effect in nematic liquid crystals doped with azo-dyes", *Mol. Cryst. Liq. Cryst* **320**, 239–248 (1998).
12. L. Marrucci, D. Paparo, P. Maddalena, E. Prudnikova, and E. Santamato, "Role of guest-host intermolecular forces in photoinduced reorientation of dyed liquid crystals", *J. Chem. Phys.* **107**, 9783–9793 (1997).
13. E. Benkler, I. Janossy, and M. Kreutzer, "Control of the orientational nonlinearity through photoisomerization in dye doped nematics", *Mol. Cryst. Liq. Cryst.* **375**, 701–711 (2002).
14. M.I. Barnik, S.A. Kharchenko, V.F. Kitaeva, and A.S. Zolotko, "Reorientation of director of nematic liquid crystals doped with azodyes under light and low-frequency fields", *Mol. Cryst. Liq. Cryst.* **375**, 363–372 (2002).
15. T. Kosa, P. Palffy-Muhoray, H. Zhang, and T. Ikeda, "Large optical torque enhancement by oligothiophene dye in a nematic liquid crystal host", *Mol. Cryst. Liq. Cryst.* **411**, 107–131 (2004).

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