Contributed paper

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Photorefractivity of dye-doped NLC layers and possibility of their application

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Photorefractive properties and possibility of guest-host effect application for real-time holography have been described. Isothiocyanate nematic liquid mixtures with positive dielectric anisotropy as hosts while antraquinone dyes as guests were used. In dye-doped nematic liquid crystals high diffraction efficiency approaching 20% were obtained. Fast optical grating formation with the time constant of a few ms have been observed. The dependence of grating formation on strength of applied dc and low frequency of ac electric field, configuration of light polarisation and nematic director orientation were reported.

Keywords: photorefractivity of dye-doped nematic liquid crystals, holographic gratings, real-time holography.

1. Introduction

Photorefractive phenomena are the most popular effect for writing dynamic phase and amplitude (flat and volume) holograms in real time. For increasing the photorefractivity of thin liquid crystal (LC) layers the dyes as a dopants (guest-host effect) or photosenstive layers in the cell are used. These phenomena give possibility to work with lower light intensity. Properties of guest-host effects in nematic liquid crystals were described in many papers. The fundamental works in this area have been done by Heilmeier [1]. Guest-host effect is typically used now in colour and black-white displays [2]. An interest in nonlinear photorefractive properties of dye-doped liquid crystals was arisen after discovering by Janossy a decrease in optical Freedericksz threshold (by two to three levels) [3]. The nonlinear optical processes in nematic liquid crystals are connected with the mechanism of orientational nonlinearity [4]. These processes are determined by electron structure of liquid crystal molecules and dopants [5,6]. In optical Freedericksz effect, the reorientation of nematic liquid crystal director in cell is proportional to E^2. A refractive index is a function of light intensity. In this paper, the induced photorefractive effect in which the director of LC molecules is initially planar and then reoriented by electric field to the homeotropic position is considered. The grating formation process in LC cells of this kind is connected with charge generation and transfer inside the cell [7,8].

Dynamic optical holography is rapidly expanding field of optical computing. Holographic elements are especially important when used in optical interconnectors for fan-in and fan-out elements. Real-time devices used in the area of optical image processing, including pattern recognition, moving object extraction etc., demand sensitive and fast recording media.

2. Theoretical and experimental background

Nonlinear processes in nematic liquid crystals are connected with reorientational photorefractivity. Light pass through the liquid crystal layer influence with liquid crystal layer and trigger nonlinear optical phenomena [4,5]. One of them is optical Freedericksz effect [5,12]. Optical Freedericksz effect is deformation of field director nematic oriented layer under electromagnetic field influence. In this paper, interaction between optical field and homeotropic nematic director ***n*** in director formed by static electric field will be considered (Fig. 1).

Nonlinear effects in nematics, considered in this paper, are connected with orientational nonlinearity. Nonlinear susceptibility of nematic liquid crystals is determined by electron molecule structure. Nonlinear optical answers are determined by delocalised π orbitals in polar nematic molecule core which contains one or more benzene rings. Reaction times of this nonlinearity are proportional to liquid crystal viscosity. Interaction with magnetic field can be neglected in this case due to low anisotropy of magnetic susceptibility for optical field frequency.

Interaction energy density of liquid crystal with electromagnetic field f_{opt} is equal to electric field interaction energy and can be given by the following equation

$$f_{opt} = -\frac{\varepsilon_0 \varepsilon_\perp}{2}\left\langle E^2 \right\rangle - \frac{\varepsilon_0 \Delta\varepsilon}{2}\left\langle (nE^2) \right\rangle. \tag{1}$$

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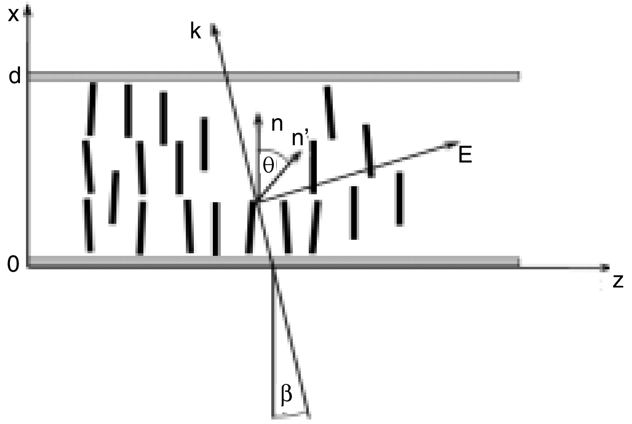

Fig. 1. Nematic director reorientation in planary oriented LC cell under dc voltage and optical field of laser beam.

Total free energy of the sample $\int f d^3 r$ in the normal state at given temperature (T = const) has minimum value and then free energy density fulfils Euler-Lagrange equation [4]. Considering this equation for two-dimensional (Fig. 1), where nematic director and electric vector of electromagnetic wave lie in x,z plane, we can obtain $\boldsymbol{n} = (\cos\theta, 0, \sin\theta)$. Then equation for the orientation angle θ has the following form

$$\begin{aligned}&\frac{\partial^2\theta}{\partial x^2}(K_{33}-K_{11})\sin^2\theta)-\frac{1}{2}\left(\frac{\partial\theta}{\partial x}\right)\times(K_{33}-K_{11})\sin 2\theta + \\ &+\upsilon(2|E_x E_z|)\cos 2\theta + EE\sin 2\theta = g\frac{\partial\theta}{\partial t})\end{aligned}, \quad (2)$$

where $\upsilon = \varepsilon_0 \Delta\varepsilon/4$.

In this dependence only splay and bend elastic constants occur while there is no twist elastic constant. This dependence can be true when we consider deformation in y,z plane. Equation (2) has a solution for chosen boundary condition, e.g. for homogeneous ($\theta = 0$) and homeotropic ($\theta = \pi/2$) orientation. These conditions must be constant for any field value (strong and weak ancoring energy). Nematic director reorientation leads to local dielectric anisotropy changes. Dielectric anisotropy tensor can be defined by the following equation

$$\varepsilon = \begin{pmatrix} \varepsilon_\perp + \Delta\varepsilon\cos^2\theta & 0 & \Delta\varepsilon\sin\theta\cos\theta \\ 0 & \varepsilon_\perp & 0 \\ \Delta\varepsilon\sin\theta\cos\theta & 0 & \varepsilon_\perp + \Delta\varepsilon\cos^2\theta \end{pmatrix} \quad (3)$$

where $\Delta\varepsilon = \varepsilon_I - \varepsilon_\perp$. It is better to use approximate effective dielectric anisotropy instead of dielectric anisotropy tensor. For the field polarised along z axis an effective dielectric anisotropy $\varepsilon_{ef} = 1/(\varepsilon^{-1})_{zz}$ is defined as

$$e_{ef} = \frac{\varepsilon_\perp \varepsilon_{II}}{\varepsilon_\perp \sin^2\theta + \varepsilon_{II}\cos^2\theta}. \quad (4)$$

Because reorientation of the nematic director depends on square of the electric field intensity $\boldsymbol{EE}$, the dielectric anisotropy and connected with it refractive index ($n^2 = \varepsilon$) is a function of the light intensity $I \sim E^2$. Taking into account the angle β (Fig. 1) between the electric field direction and z axis ($tg\beta = |E_x/E_z|$), as well as simplifying the problem to one Frank elastic constant ($K_{11} = K_{33} = K$) one can transform Eq. (2) to the form

$$K\frac{\partial^2\theta}{\partial x^2} + \upsilon EE\sin 2(\theta+\beta) = g\frac{\partial\theta}{\partial t} \quad (5)$$

For light propagation perpendicular to the walls of the cell ($\beta = 0$) and for homeotropic liquid crystal director orientation (boundary condition $\theta(0) = \theta(d) = 0$), Eq. (5) has a solution for small angles $\theta << 1$, that is $\sin\theta \approx \theta$ in a form

$$q(x) = q_0\exp(t/\tau)\sin(\pi x/d), \quad (6)$$

where

$$\frac{1}{\tau} = \frac{1}{\gamma}\left(2\upsilon E^2 - \frac{\pi^2 K}{d^2}\right). \quad (7)$$

Liquid crystal layer reorientation occurs only, when the time constant $1/\tau$ is positive and the value of θ angle increases with time until to maximum value (for which small angle approximation is not valid). It means that E^2 must be higher than the threshold value E^2_{pr}, for which $1/\tau$ equals zero

$$E^2_{pr} = \frac{2\pi^2 K}{\varepsilon_0 \Delta\varepsilon d^2} \quad (8)$$

Director reorientation is possible when intensity of light is higher than threshold value. This phenomenon is called induced optical Freedericksz effect. It is possible only for cell configuration in which a light wave field goes to rotate director about $\pi/2$ angle with respect to the initial position. For other cases [for example for ($\beta \neq 0$)] this phenomenon has unthreshold character. The threshold field value is inversely proportional to d^2 which means liquid crystal cell thickness. Also in the case of unthreshold phenomena, nonlinear coefficient increases with square of the thickness of liquid crystal layer. It is necessary to remember that it is impossible to obtain good orientation in the cell which is thicker that 100 µm, i.e., it is impossible to increase freely cell thickness. Satisfactorily good orientation in the cell is possible to obtain in the cell 100 µm thick. The thickness of the cell which has been used in investigation were d = 5–15 µm. In our experiment light wave reacts with homeotropic cell structure which has been obtained by electric field.

For the director reorientation in optical Freedericksz effect, the threshold electric field value is necessary. This field is inversely proportional to the cell thickness d, dielectric anisotropy $\Delta\varepsilon$ and K, where K is connected with Frank elastic constant. It is very well known that it is impossible to reorient the LC sample thicker than 100 μm.

The speed of director reorientation in the sample and speed of nonlinearity increase is described by the equation

$$1/\tau_n = 2\upsilon E^2/\gamma \tag{9}$$

Time of increasing nonlinearity depends on E^2 and can be very short if E is high enough. The relaxation time of these processes is connected with the thickness d^2, γ, and K and can be given by the next equation

$$1/\tau_p = K/d^2\gamma \tag{10}$$

The typical value of nonlinearity coefficients for nematics is in the range ~10^8 W/m^2 and the times 10^{-3}–1 s. For homeotropic orientation ($\beta = 0$), where $\boldsymbol{E}$ is proportional to the director $\boldsymbol{n}$, the observed director reorientation changes and connected with them changes of refractive index are a function of light intensity

$$I_{opt} = \frac{\varepsilon_0 nc}{2}E^2. \tag{11}$$

Result of the director reorientation $\boldsymbol{n}$ to $\boldsymbol{n}'$ in the plane x,z is the change of the refractive index n_e which can be described by the equation

$$\Delta n_e = n_e(\beta+\theta) - n_e(\beta) \tag{12}$$

where $n_e(\beta + \theta)$ is the extraordinary refractive index

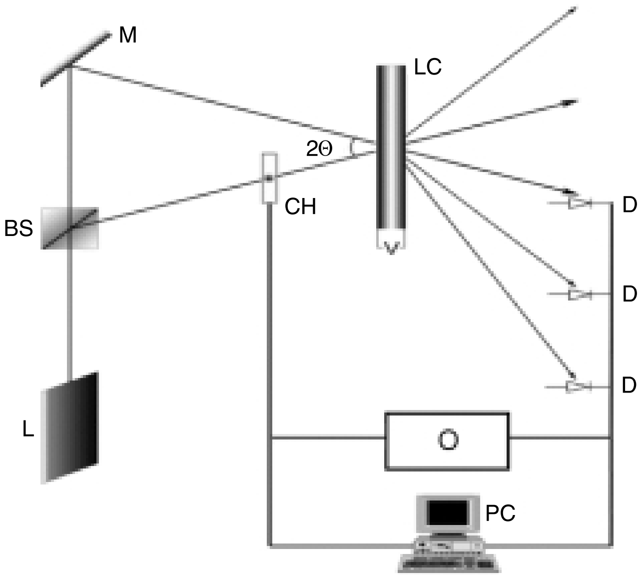

Fig. 2. Experimental setup (DTWM) for writing dynamic holographic gratings and measurement diffraction efficiency: L – laser, M – mirror, BS – beam splitter, LC – dye-doped liquid crystal cell, D – detectors, O – oscilloscope, CH – chopper, and PC – computer.

$$n_{ef}(\beta+\theta) = \frac{n_o n_e}{[n_e^2\cos^2(\beta+\theta) + n_o^2\sin^2(\beta+\theta)]^{1/2}} \tag{13}$$

Nematic director reorientation in LC cell is proportional to E^2 and is a function of light intensity. From Eq. (11), connecting E^2 with I_{opt} one can find that I_{opt} electric field value, which reacts with director of nematic liquid crystal with refractive index $n = 1.6$ is less then 1 V. This electric field value is too small to reorient the LC director and it is comparable with the field in classic Freedericksz effect. In 1994, Janossy shown, that the optical Freedericksz transition can be decreased by addition of dopants of some kind, e.g., dyes [3]. In this way, it is possible to decrease optical field necessary to reorient liquid crystal director from 10 to 1000 orders of magnitude. It gives a possibility to construct the optical light modulators, which can be addressed by light beams of a very low intensity.

Using this phenomena, the planar oriented cells with guest-host effect for dynamic holography under DC and low frequency AC voltage have been investigated [7–10]. The writing and erasing holographic gratings processes usually were investigated in DTWM setup (Fig. 2). A linearly polarised He-Ne laser $\lambda = 632.8$ nm, 10 mW was applied. Two coherent writing beams make the angle 2θ (with the range 1–15°).

3. Results

Our investigations were concentrated on fast response (writing/erasing) times and high diffraction efficiency cells which can work as optical spatial light modulators (OASLM) in Raman–Nath regime. Induced holographic gratings recorded by light intensity pattern in nematic liquid crystals doped with antraquinone dyes have been investigated. It was possible to obtain the dynamic holograms with a few W/m^2 using this type of cells. Dynamic holograms with recording/erasing speed of 500 cycles per sec-

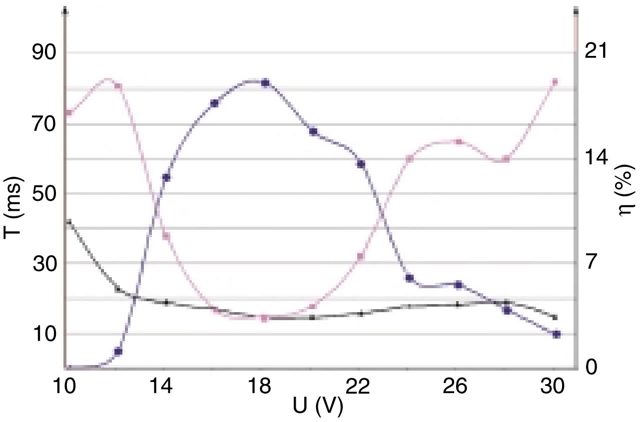

Fig. 3. Dependence of diffraction efficiency and writing/erasing times vs. voltage (● – difraction efficiency, ▲ – writing time, ☐ – erasing time (temp. 20°C, LC mixture W-1294 with $\Delta n = 0.35$, antraquinone dye B_7).

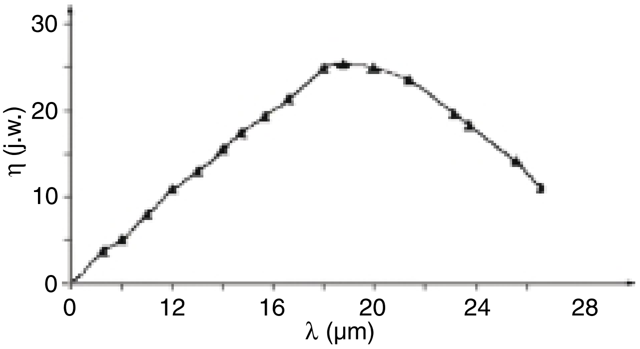

Fig. 4. Dependence of diffraction efficiency versus the grating period Λ (d = 9 m).

ond for 2% diffraction efficiency was obtained. Typical dependencies for investigated cells are shown in Fig. 3. The diffraction efficiency rapidly increases with a voltage increase. The maximum diffraction efficiency value depends on construction parameters of the cell (thickness and resistance of the orientation layers). The writing time is shortest for the maximum of diffraction efficiency but erasing time is practically constant in all applied voltage range.

We noticed that these parameters depend not only on materials, i.e., liquid crystal and dyes but cell construction is also very important (orientation and protection layers). Studying the gratings formation for various angles between the incident writing beams we have found that the position of maximum of the first order of diffraction efficiency is a function of the grating spacing Λ. The maximum value of the diffraction efficiency η (about 20%) measured in the most favourable conditions was obtained for the period Λ which was two times higher than LC cell thickness (Fig. 4).

The same dependencies were observed by Weiderecht and co-workers [6]. On the other hand, if the cell thickness is small, thin grating is possible to write. Our investigations confirm that this dependence is connected with electrohydrodynamical instabilities inside the cell when the voltage is increased [12]. Typical resistance of liquid crystal used in the experiment was about $10^8\ \Omega$. If the resistance of a liquid crystal decreases, the electrohydrodynamical instabilities increase and also the scattering of the cell increases which is disadvantageous from the application point of view [12,13]. Figure 5 shows the scattering of the incident light beam passing across the cell for high (a) and low (b) scattering, respectively. This scattering is connected with rubbing of the orientation layers, domain structure and local order parameter fluctuations. In Fig. 6, the light intensity distribution of laser beam for cells from Fig. 5 is shown.

4. Conclusions

In this paper some experimental results obtained in antraquinone dye-doped nematic liquid crystals, able to act as photoconductivity sensors, have been presented. Application parameters of optical spatial light modulators of this kind are listed in Table 1. The most important parameters are diffraction efficiency, writing/erasing times and revers-

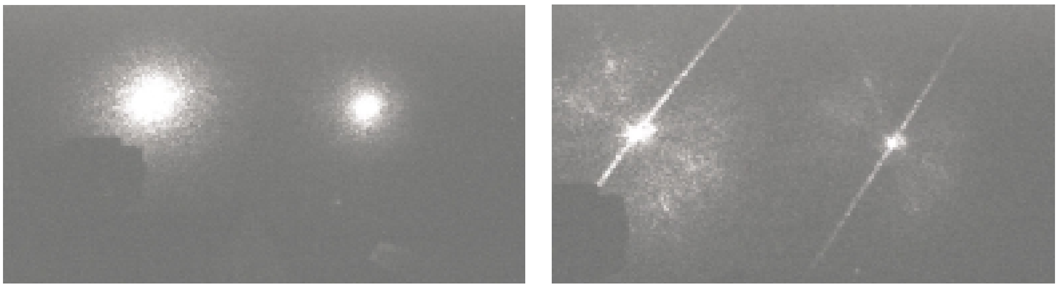
(a)
(b)

Fig. 5. Laser beams images dispersion in the LC cells: high cell dispersion (a), low cell dispersion (b) (d = 9 μm, LC isothiocyanate mixture 1298, antraquinone dye B_7).

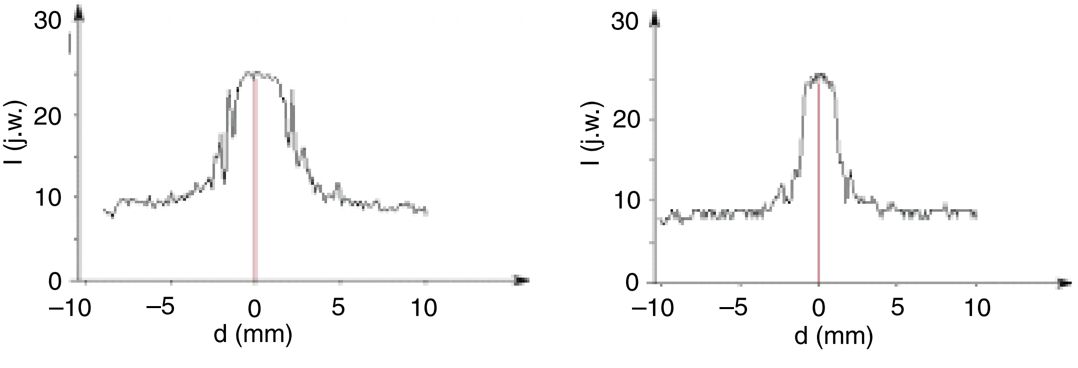

Fig. 6. Light intensity laser beam distribution as a result of dispersion: high cell dispersion (a), low-cell dispersion (b) (conditions as in Fig. 5).

ibility of the process. Fast optical grating formation with the times τ of 2 ms and diffraction efficiency 2% were observed. To obtain better parameters it is necessary to decrease the scattering of the cell. We hope that, using for example photo-alignment, it will be possible to reduce scattering, which occurs in typical rubbed cells. This kind of cells are suitable for writing holograms by the projected method.

Table 1. Parameters of investigated dye-doped liquid crystal OASLM for real-time holography.

Parameter–material	NLC W – 1294 with dye B_7
Type of grating	Thin transmission gratings
Thickness of the cell (μm)	5–30
Diffraction efficiency η (%)	up to 30
Possible change of Δn	~0.2
Change of Δn	$\sim 5\times 10^{-3}$
Resolution (lines/mm)	1000
Power of light (W/m^2)	~1–100
Time of writing τ (s)	$\sim 1\times 10^{-3}$
Repetitivity	Practically no limit
Reversibility of processes	Totally reversible
Sensitivity of material (Jcm^{-2})	~1000
Costs of modulators	low
Applications	Dynamic optical memory, optical processors

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