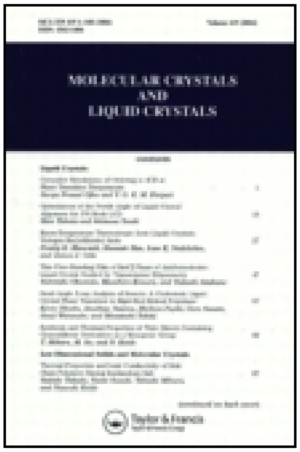

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Photoconductive Polymers in Liquid Crystal Spatial Light Modulators with High Resolution

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PHOTOCONDUCTIVE POLYMERS IN LIQUID CRYSTAL SPATIAL LIGHT MODULATORS WITH HIGH RESOLUTION.

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Abstract Various types of organic polymer photoconductors have been successfully used in optically addressed liquid crystal (LC) light valves based on the electrically controlled birefringence, optical activity, cholesteric-nematic phase transition, dynamic scattering, and polymer dispersed LC. Record regime optimization in projection or holographic manner is necessary to obtain the best device characteristics. The highest parameters obtained in the pulse holographic regime were diffraction efficiency of 36%, limiting resolutions of 1500 mm^{-1}, and spectral sensitivity of 10^{-8} J/cm^2 %. Photoconductive polymers themselves may be used for LC alignment simplifying the light valve construction. High aperture devices were realized side by side with the small ones. Polymer dispersed LC's connected with the photoconductive polymers allow for the realization of entirely solid state valves. High resolution, mechanical and elecrical stability, and the possibility for sensitization make the use of the polymer photoconductors in spatial light valves very perspective. Such valves can be used as phase reversible medium in various optoelectronic devices and processes.

INTRODUCTION

Spatial light modulators (SLM) play the key role in photonic information processing systems such as optical computing, photonic switching, optoelectronics, holography, laser light modulation and so on [1]. These optical devices modify intensity, phase, or polarization of an optical beam as a function of space and time and permit the transfer of two-dimensional images in the cross-section of the coherent laser beam in real time. The SLM may provide an interface between the electronic and optical signals due to electronic or optical control of the modulation functions.

Liquid crystal (LC) SLM are very promising for the above-mentioned application, because of the variety of the the electro-optical effects, small power consumption, and low cost.

Here we are concerned with optically addressed LC SLM. The simplest modulator structure is shown in Figure 1. Thin films of the photoconductor (Ph) and LC are sandwiched between transparent conductive electrodes on glass plates. DC or AC voltages are applied to the electrodes.

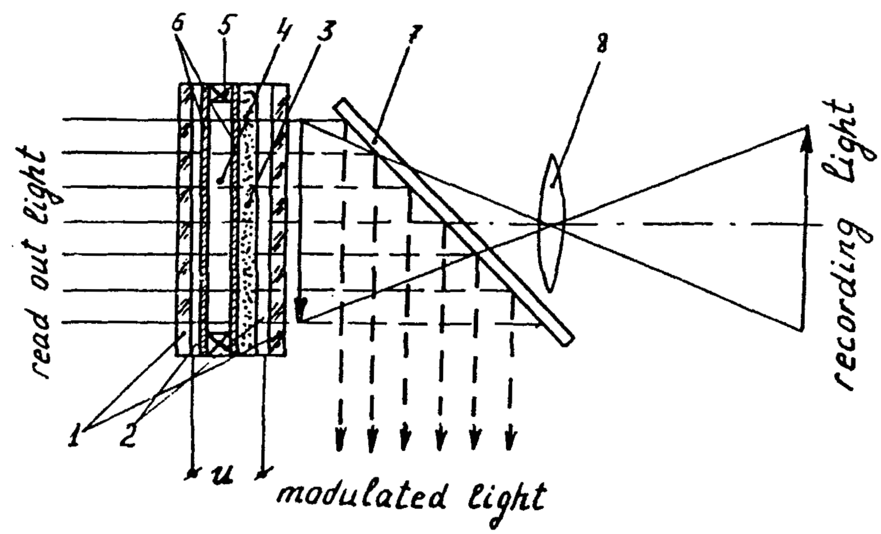

Figure 1 Typical scheme of spatial light modulator. 1 - glass plates, 2- transparent conducting electrodes, 3-photoconductor, 4-liquid crystal, 5-spacers, 6-aligment layers, 7-semitransparent mirror, 8-lens.

Alignment of the LC molecules is secured by rubbing conductive electrode coatings or by vacuum evaporation of thin dielectric films such as silicon or germanium dioxide. SLM's based on electrically controlled birefringence(BR), optical activity (OA),cholesteric-nematic phase transition (PT), or dynamic scattering (DS) have been constructed and tested. LC mixtures of cianobiphenyls and azoxybenzene with positive dielectric anisotropy and anisotropy of the index of refraction of 0,2 were used.

The main features of SLM functioning are the following. Because the photoconductor is highly resistive in the dark an applied electric field drops across the photoconductor thickness. When the recording light is turned on, charge carriers are generated by the incident photons, the effective resistance of the photoconductor drops, and the field across the electro-optical medium increases. The LC medium, in turn, responds to the imagewise-modulated field producing a viewable image in the read out optical beam. It is clear that the photo-conductor determines the main parameters of the SLM , such as energetic and spectral sensitivity, resolution, response time and so on. Inorganic photoconductors such as CdS, GaAs, ZnSe , amorphous Si, and halcogenide glasses have been used in SLM's until recently.[1] New photosensitive materials are needed to increasing the number of excellent modulator features. Some time ago we suggested that organic polymers be used as a photo-conductive layer in LC SLM[2]. High resolution, mechanical and electrical stability, ability for sensitizing the photoeffect within a certain spectral range, small energy consumption, and low cost make the use of the organic polymer photoconductors in SLM very promising. It should be mentioned that application of conjugated polymers is promising in such systems as batteries, capacitors, transparent speakers, electrophotography, thermoplastic devices, and nonlinear optics.

Here we are interested in the photocoductive polymers.[3] A lot of organic photoconductor polymers are available now for various types of devices. The main types of such polymers are characterized by their bonds in the main or side chains of the macromolecules:

1. Polymers with saturated bonds in the main chain. Typical models are poly (N-vinylcarbazole) and its derivatives, charge-transfer complexes with donor-acceptor molecules.
2. Polymers with conjugated bonds-polyacetylene, polydiacetylene, polyphenylensulphide, poly-p-phenylene, polythiophene, polyimides etc.
3. Metal-organic polymers - Copper-organoacetylides.
4. Heterogeneous and molecular doped polymer systems-polycarbonates , plasma and thermo-polymerized compounds.

Practically all of the above-mentioned polymers were tested in our research including poly (N-vinylcarbazole), poly (N-epoxyvinyl carbasole), polyimides , polymers with conjugative bonds obtained by thermal treatment of polyacrilonitrile , polyvinylchloride and so on. The main requirements for the polymers to be used in SLM were resistivities from 10^{7} to 10^{16} Sm cm^{-1},high photosensitivities up to 10^{-8} J cm^{-2} for the recording ligth, transparency for the read out light, simple technology of film making, electrical and mechanical stability, and no interaction between polymers and LC.

We have investigated sensitivity W, switch on and switch off times (τ_{on} and τ_{off}), resolution R, diffraction efficiency η, versus spatial freguency Λ, the influence of the dielectric anisotropy $\Delta\varepsilon$ and the type of LC and Ph on the different parameters of the SLM.

Various write on regimes were tested. Transition from a constant to a commutational and pulse writing regime markedly improved SLM characteristics. Constant regime means that the input image was formed by means of a continuous He-Cd laser light of the 440 nm wavelength or by tungsten lamp. Commutational regime was made by laser light pulses of 530 nm wavelength lasting from 0.1 to 2 sec. Pulse regime was performed by laser light pulses of the 530 nm wavelength lasting for 20 nsec and energy density up to 2 mJ cm^{-2}. The reading of an image was made by a continuous He-Ne laser with the 630 nm wavelength or by a white light in a transparency mode.

The simplest schemes showing the write on and off regimes are seen in Figure 1 and Figure 2. The set up, shown in Figure 2, allows the determination of the holographic properties of the SLM. A sinusoidal grating was constructed on the SLM by the two intersecting interfering laser beams with a 75% modulation depth. The diffraction efficiency, η, was defined as the ratio of the light intensity of the

He-Ne laser passed in the first order diffraction to the whole intensity of the reading light. Response times were defined as the times needed for increasing (decreasing) its maximum values from 0 to 0.9 (from 1 to 0.1) beginning from switching on (off) of the read on light. The spatial frequency Λ, was calculated by the formula $\lambda\Lambda = \sin\theta$, where λ-light wavelength, and θ -first order diffraction angle.

The summary of the SLM parameters obtained can be seen in a Table .

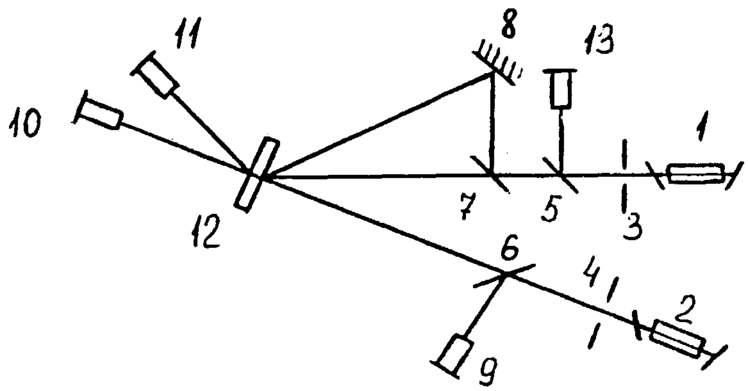

Figure 2 Schematic diagram and experimental arrangement for holographic regime. 1-laser YAG-Nd, 2-laser He-Ne, 3,4- diaphragms, 5,6- glass reflecting plates, 7- semitransparent mirror, 8- mirror, 9-photodiode, 10,11-photoamplifiers, 12-spatial light modulator, 13-photodiode.

RESULTS AND DISCUSSIONS

Now we go on to the experimental and theoretical results obtained. First of all , consider the absorption and photosensitivity spectra of the SLM. Photoconductivity spectra of the polyimide LC structure are shown in Figure 3 with positive (right) and negative (left) bias on the photoconductor.

TABLE I. PARAMETERS OF THE SPACE LIGHT MODULATORS WITH POLYMERIC PHOTOCONDUCTORS.

write-on regime	electro optical effect	λ_{sen} nm	sensitivity	T on(s) min	T off(s) min	contrast ratio	diffraction efficiency %	Resolution mm^{-1}	controlled voltage V	photoconductor	references
CONSTANT	BR	400–500	10^{-4} Jcm^{-2}	0.25	0.7	80			5 – 40	polyimide	2,5
	PP	400–600	10^{-4} Jcm^{-2}	0.2	0.75	100			60 – 140		
	OA	440	$2\cdot10^{-5}$ Jcm^{-2}	0.4	0.3	50			10 – 70		
	OA	440	$1\cdot10^{-6}$ Jcm^{-2}/%	0.1-2	0.1-2		13 1 10^{-3}	20 150 300	1,0-80	–	5
CONTINUOUS	BR	530	$3\cdot10^{-6}$ Jcm^{-2}/% $3\cdot10^{-7}$ Jcm^{-2}/%	0.2 0,015	0.2		23 20	> 90 90	10 40	—	6,7
PULSE	BR	530	10^{-8} Jcm^{-2}/%	0.0001	0.3-1		36 20 12 0.1	100 400 550 1500	10-50	—	8,9
	BR	530	10^{-6} Jcm^{-2}/%	0.005	≥ 0.1		20 0.3%	50 300	10-50	conjugated polymer	10
	PDLC	530	$5\cdot10^{-5}$ Jcm^{-2}	0.0001	0.08	33			5-250	polyimide	15

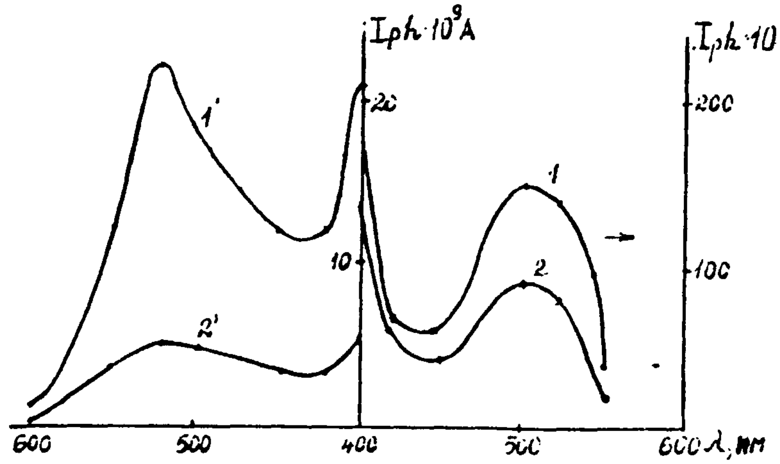

Figure 3 Photsensivity spectra of the polyimide liquid crystal structure with positive (right) and negative (left) bias on the photoconductor. Voltage in V : 80(1,1'), 20(2,2').

The sensitized photosensitivity is spread up to 600 nm and has a maximum value at the 500 nm wavelength at positive and 530 nm at negative bias voltage on Ph. Positive sign of the dominant charge carriers in polyimide was confirmed by comparing the photocurrent at different bias.The shift of the maximum value ca 30 nm with the polarity change is likely to indicate the potential barrier present at the Ph-LC interface. The physical nature of the barrier is probably the same as considered previously[4]. The existence of the barrier was confirmed by examining the current-voltage characteristics of the SLM which were typical for rectifying structures, and by the photoelectromotive force of 0,3 eV, the sign of which was the same no matter which side was lit. The rectification coefficient at 20 V was equal to 20 in the dark and 3 with the recording light. The great influence of this rectifying barrier on all the parameters of the light valve has been established.

The absorption and photoconductivity spectra of the modulator with thermally treated polymers (polyacrylonitrile, polyvinylchloride) were determined by the polymerization process. Figure 4 shows the absorption spectra of the polyacrylonitrile treated at 160 ^{o}C(curve 1),190 ^{o}C(curve 2), and 220 ^{o}C (curve 3). One may see that the photosensitivity in the visual and near infrared spectral range can be reached by such treatment. So we may conclude that the spectral sensitization of the photosensitivity by means of dyeing and thermal or photochemical treatment may be easily reached in organic polymers for the SLM.

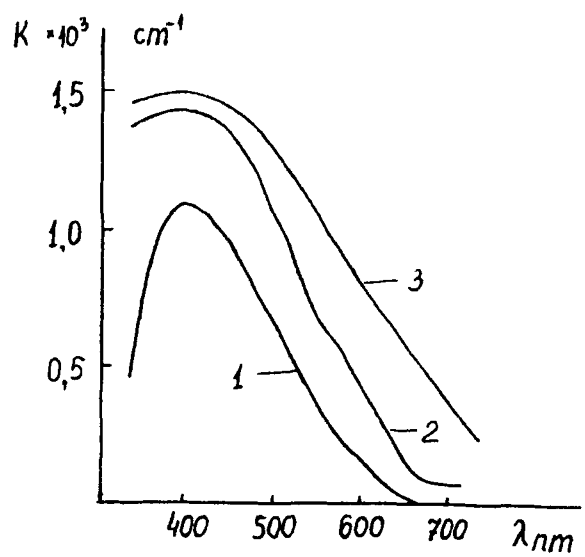

Figure 4 Electronic absorption spectra of the polyacrylonitrile films thermally treated for 3 hours at 160°C (1),190°C (2) and 1 hour at 220°C(3)

SLM CHARACTERISTICS

Constant regime[2,5].

The transparency versus voltage (a) and current-voltage characteristics (b) for the SLM with cholesteric-nematic PT are seen in Figure 5.

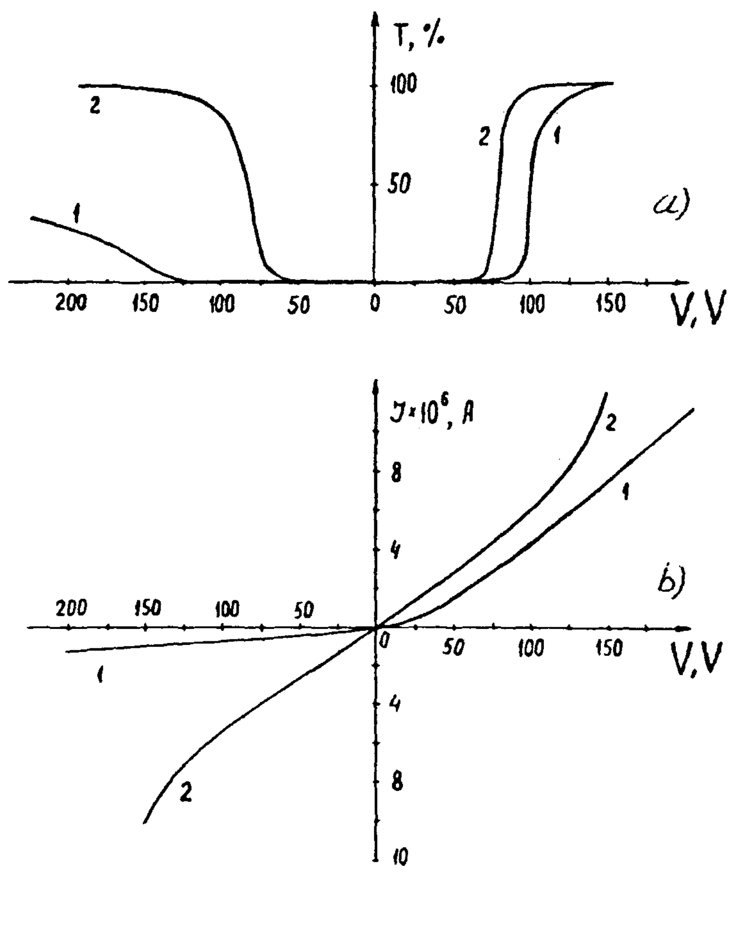

Figure 5 Volt-contrast (a) and current-voltage (b) characteristics for polyimide modulator with cholesteric-nematic phase transition without (1,1′) and with switch (2,2′) of the read on light. Constant regime.

The transparency versus voltages was measured without polarized optics. The structure scatters the light in the initial stage due to the confocal texture of the LC. The spiral unwinding at high voltages leads to a transparent state because of the electrically induced homeotropic orientation of the LC molecules.

One can see that both characteristics in Figure 5 at the initial stage are unsymmetrical versus polarity bias. This confirms that there is a barrier at the interface between Ph and LC. The barrier height depends on the distribution of charges at the Ph surface, Helmgolz layer,and diffuse part of the double layer in LC. Switching on the light leads to the elimination of the assymetry. The same result was obtained with the deposition of a SiO film on the Ph surface to induce aligment of the LC molecules for BR type modulator. This indicates that switching on the light or SiO deposition lead to a polymer surface sign change from positive to neutral or negative because with hole polyimide semiconductor, the rectifying effect can be produced only by excess positive carriers on the polymer surface[4]. This was also confirmed by photovoltaic measurements on the same structure. The height of the barrier was estimated as 0.3 eV. The main parameters of the SLM with PT are: sensitivity 10^{-4} W cm^{-2}, τ_{on}- 0,2 s, τ_{off}- 0,75 s, controlled voltages 60-140 V. Similar results were obtained for SLM with BR and OA.

Maximum diffraction efficiency up to 13% has been reached with negative bias on Ph (Figure 6) in the holographic regime.

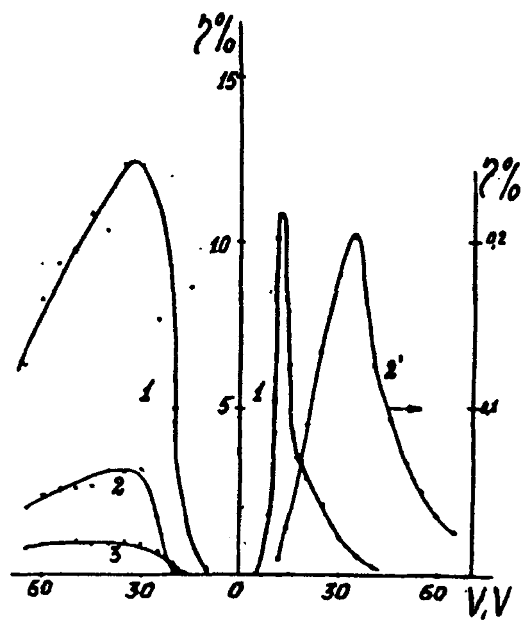

Figure 6 Diffraction efficiency versus bias polarity on polymer photoconductor. Positive (right), negative (left) for spatial frequencies in mm^{-1} 20(1), 100(2,2'), 150(3). The intensity of the read on light $-8\cdot10^{-5}$ Wcm^{-2} . Constant regime.

The values of the diffraction efficiency versus controlled voltages can be seen in the same Figure 6.

The period of the formed in SLM grating was much less of the LC thickness. This corresponds to a thin flat hologram. The limiting value of the diffraction efficiency for such hologram is equal to 33.9% in case of normal light incidence if the condition $2\pi\Delta nd/\lambda \rightarrow 2$ is fulfilled. Here Δn is the birefringence, d is the LC thickness, and λ is the wavelength of light. The LC thickness is really much less than d due to the interaction with electrodes and the nonideal planar alignment of the LC molecules. The value of the halfwidth of the η for positive bias voltages on the Ph was less than for negative bias voltages for equal frequencies and increases with spatial frequency increase. The maximum η value for positive bias on the Ph was reached with different voltages in case of different frequencies. All these phenomena are due to a Schottky barrier at the LC-Ph interface. This was confirmed by current-voltage characteristics and photo-emf measurement. The complicated dependencies of the η, τ on and τoff upon the intensity of the read on light were established for this type of the SLM.

The results obtained show that negative bias on the Ph is more advantageous for SLM functioning. For example, the diffraction efficiency was 3% for positive and only 0,2% for negative bias in the case of equal intensity of the read on light and a frequency of 100 mm^{-1}. Half values of η were reached at 85 mm^{-1} for negative and 35 mm^{-1} for positive bias. One may conclude that SLM modulation is really controlled by the driving voltage and intensity of the read on light. It is clear that these conditions play the key role for resolution increase but not only the anisotropy of the LC dielectric permittivity as it was shown in [1]. Maximum holographic sensitivity was 10^{-6} J/cm^2 %.

The long-time memory regime of the SLM can be obtained by the positive bias on the Ph. The reasons of it will be the subject of future investigations.

Commutational regime[5-7].

Diffraction efficiency, τ on and τoff versus voltages for an electrically controlled birefringence type modulator with laser light intensity $5x10^{-4}$ W cm^{-2} is shown in Figure 7. An efficiency value of 23% at 10 V was obtained and it has not been practically changed up to the frequency of 60 mm^{-1}. The resolution value at 0,8 level of the η was 45 mm^{-1} at 40 V. On times at different frequencies were ca 15 msec at voltages more than 40 V (Figure 7) and had different values

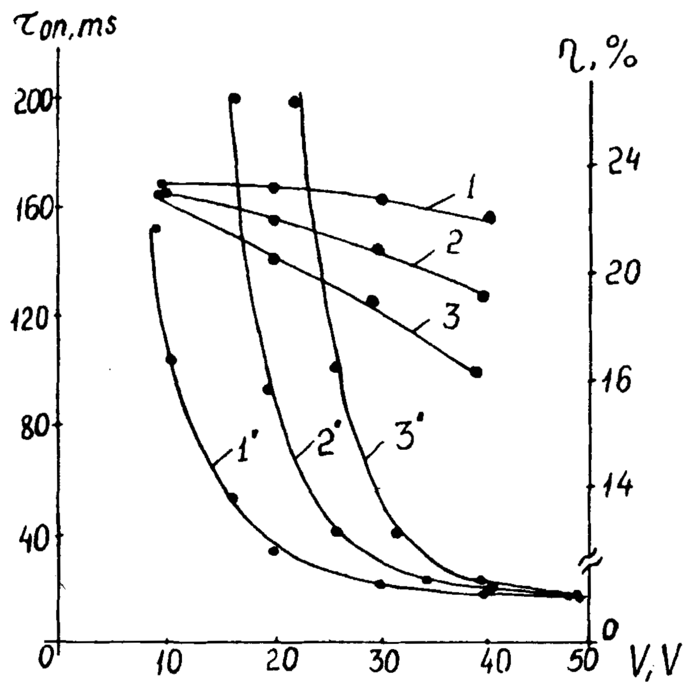

Figure 7 Diffraction efficiency versus voltages for spatial frequencies in mm^{-1} 10 (1); 28 (2); 51 (3). Switch on time versus voltages at space frequencies in mm^{-1} 10(1);36(2);60(3). Commutational regime.

for lower voltages. This allows the space time selection of the images. The low limiting Λ obtained was 125 mm^{-1} due to the restrictions of the experimental set up[6]. The holographic sensitivity was 3×10^{-7} J/cm^2 % at 40 V and $\Lambda=28$ mm^{-1}. Storage times of a image varied from seconds to hours for different regimes. The maximum modulator parameters were obtained when the direction of the electrical vector of the read out light coincided with the planar LC director orientation and was normal to the wave vector of the read on light.

Pulse regime[8-10,12,13,15]

The best results were achieved in this regime, where laser light pulses lasted 20 nsec with energy densities up to 2 mJ cm^{-2}. The above mentioned conditions may be expected to result in resolution gains due to a decrease of the transverse spreading of charges at the Ph-LC interface and elimination of the set up vibrations. Moreover, the switch on times have to be decreased due to a photocurrent increase if the exciting light energy is high. These expectations were firmly confirmed, as seen in Figure 8, where the diffraction efficiency versus spatial frequency is shown. A maximum value of 36% at 100 mm^{-1} was obtained for a polyimide modulator with controlled voltages of 30-50V and a light on energy of 10^{-4} J cm^{-2}. The limiting resolution value obtained was 1500 mm^{-1} with a diffraction efficiency of 0,1%. As far as we know, the above-mentioned value is the highest one reported for the LC SLM. If Λ is equal to 400 mm^{-1}, η = 20%; for Λ = 650 mm^{-1} η = 12%. The maximum holographic sensitivity was 10^{-8} J/cm^2 %, the minimum τ on time-100μsec, τ off time - from 0,3 to 1 sec.

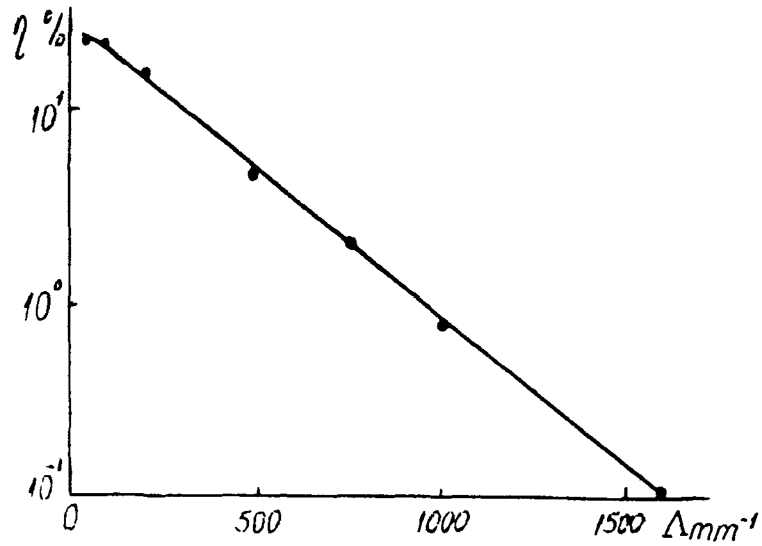

Figure 8 Diffraction efficiency versus spatial frequency for electrically controlled birefringence modulator. Pulse regime.

Earlier we pointed out that the SLM might be considered as a thin phase hologram, the diffraction efficiency of which was determined by the formula

$$\eta = J_1^2(\Delta\varphi) = J_1^2(2\pi \Delta n d/\lambda)$$

The maximum η value is reached at φ = 2 radian. The charge distribution can be concentrated on the Ph-LC interface or in the photosensitive material itself[11]. The latter took place in our case as it was shown. The upper limit for a thin phase hologram in 33,9% in the case of low frequencies was exceeded up to 36%. It was probably the result of more rectangular but not sinusoidal charge distribution in the grating at the high light intensities.

SLM modulation function with different values of the LC thickness is presented in Figure 9. One can see a

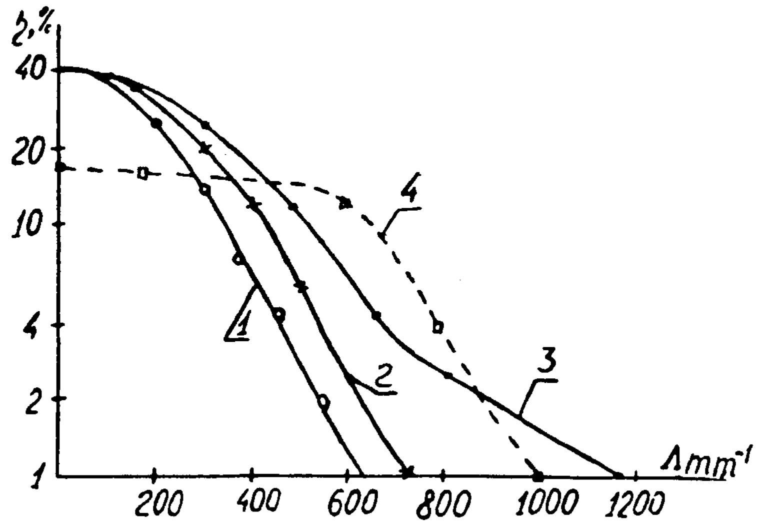

Figure 9 Modulation function of the polyimide modulator with LC thickness in μm 20(1), 15(2), 10(3), 3(4). Anisotropy of the dielectric permittivity 12(1,2,3), 0,05(4). Polyimide thickness - 2μm. Pulse regime.

considerable increase of the resolution, especially in the region of high frequencies, with decreasing the LC thickness. The same tendency was observed for decreasing the anisotropy of the dielectric permittivity (curve 4). Thus, we experimentally demonstrate a metod for substantial improvement of the modulation characteristics for a given spatial frequency.

Besides polyimides,various types of other photoconductive polymers were used. As an example, the dependencies of the diffraction efficiency, τon,and τoff times versus spatial frequency for a modulator containing a polymer with conjugated bonds was used as a photosensitive layer are presented in Figure 10. The main parameters obtained are:

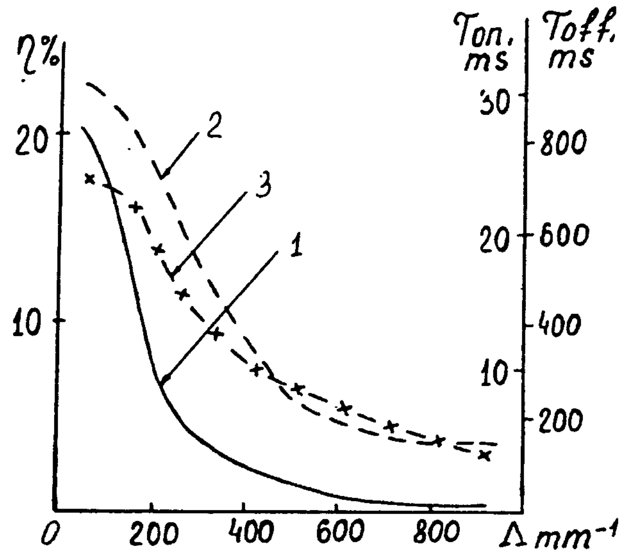

Figure 10 The dependence of the diffraction efficiency (1), switch on (2) and switch off (3) times versus spatial frequency for modulator with photosensitive conjugated polymer. Birefringence effect. Pulse regime.

sensitivity 10^{-6} J/cm^{2} %; diffraction efficiency - 20% at spatial frequency of 60 mm^{-1} and 0,3% at 800 mm^{-1}; rise time $\geq$ 5 ms, decay time $\geq$ 100 ms. It should be noted that the homogeneous orientation of the LC was achieved by direct rubbing of the photoconductive polymer film. This allows a simplification in modulator production due to the exclusion of the alignment film deposition.

All the previous results were concerned for SLM's with clear aperture of 30 mm. A considerable increase of the aperture may be reached due to the simplicity of producing thin photoconductor polymer films compared with monocrystals and polycrystalline films of inorganic photoconductors. SLM's with more than 100 mm aperture were constructed and investigated with BR, PT, DS electrooptical effects[12,13]

A maximum contrast ratio of 18:1, and minimum τ_{on} and τ_{off} of 2 ms and 250 ms were obtained for the DS and PT mode SLM. For the BR valve the corresponding times were 6 ms and 70 ms, while a contrast ratio of 90:1, resolution of 140 mm^{-1} and a diffraction efficiency 10% were achieved. The controlled voltages were higher, ca 100-400 V for the SLM's with large aperture. Polyimide photoconductive films were used in such SLM. It should be noted that the SLMs with large aperture have increased information capacity and may be used in the corresponding optical devices.

The SLM's mentioned above all contained molecules in LC state. An entirely solid state device is naturally preferable for practical use. The polymer dispersed LC materials may be beneficial as a counterpart of the SLM as a modulating medium. The nematic LC dispersed as submicron size droplets in polymers may find a use as the electrically switched shutters with a wide range of applications[14]

Films created by us had contrast ratios up to 400:1, $\tau_{on} \geqslant 0{,}4$ ms, $\tau_{off} \geqslant 2$ ms, and maximum transparency at 25 V. The sandwiched structure from the polyimide photosensitive film and the polymer dispersed LC film - i.e. the optically controlled solid state SLM - had the characteristics[15] presented in Figure 11. For pulse regime with the exposure up to 5×10^{-5} Jcm^{-2} such SLM had the contrast ratio 35:1, $\tau_{on} \geqslant 400$ s, $\tau_{off} \geqslant 80$ ms. The reversibility up to 10 hertz was reached. Thus, the polymer dispersed LC films are very perspective for the SLM.

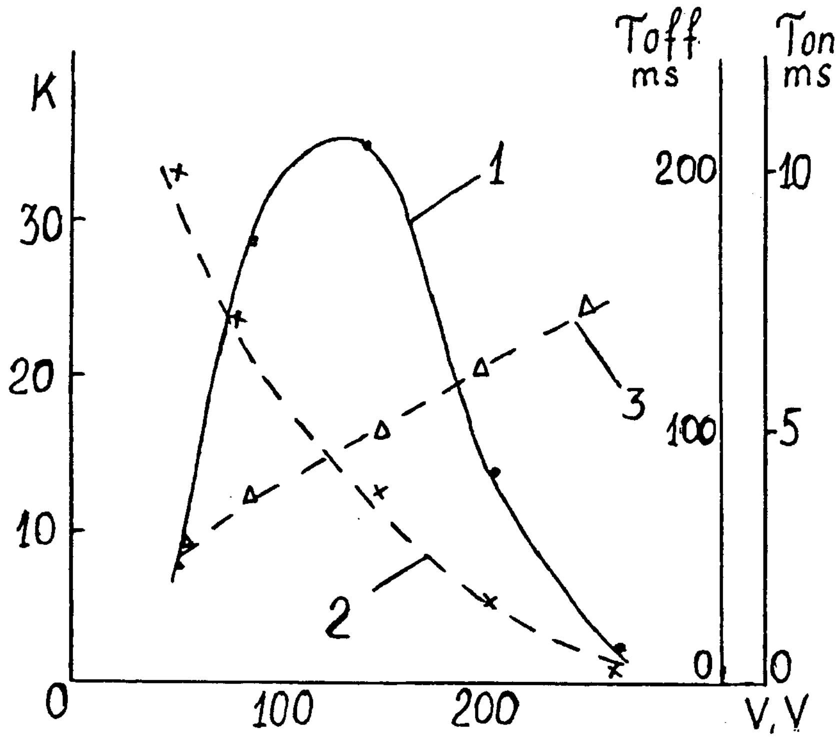

Figure 11 Contrast ratio (1), switch on (2) and switch off (3) times versus voltages for modulator with polymer dispersed LC. Photoconductor - polyimide film. Pulse regime.

CONCLUSION

1. High efficiencies of the LC SLM with photoconductive polymer were established. The maximum diffraction efficiency up to 36% and the limiting resolution 1500 mm^{-1} were obtained.

2. Record regimes optimization is necessary for the solution of concrete problems. The pulse regimes allow to obtain the highest holographic parameters compared with the constant ones.

3. The interface LC-Ph barrier determines the peculiarities of the main SLM characteristics.

4. The usage of the organic polymer photoconductor itself for the LC alignment allows to simplify the SLM production.

5. The information capacity of the SLM can be easily increased due to the manufacture of the devices with high apertures.

6. The polymer dispersed LC as a modulating medium connected with the photoconductive polymers allow to obtain the entirely solid state SLM with high parameters.

7. High resolution, mechanical and electrical stability, possibility for sensitization of the photosensitivity make the use of the organic polymer photoconductors in the SLM very perspective. Such SLM, as the phase reversible material, can be used for input and output of the optical information, in optoelectronics, holography, for in-and out-resonance of laser light modulation, associative memory, optical inter-connections and so on.

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