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Surface-assisted optical storage in a nematic liquid crystal cell via photoinduced charge-density modulation

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Abstract

This study reports the observation of quasi-permanent recording of holographic gratings in a twisted nematic liquid crystal cell by He–Ne 632.8 nm laser light. The cell is equipped with blocking SiO_2 layers and polyimide insulating/alignment layers and filled with liquid crystalline 6CHBT, doped with an anthraquinone derivative dye. Photogeneration in the bulk of the liquid crystal, photocurrent flow, and loading of the surface with spatially modulated charge density are the major mechanisms which lead to surface electric field induced director reorientation. The novelty of the system described by us is the possibility of ac frequency tuning of the intensity of light diffracted at the so formed refractive index grating. © 2001 Elsevier Science B.V. All rights reserved.

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1. Introduction

Nematic liquid crystals (LCs), because of their unusual anisotropic optical properties, were frequently used in a variety of electro-optic applications [1,2]. Among them liquid crystalline optically addressed spatial light modulators (OA SLM) form a class of devices with growing interest in the area of optical data processing and storage. In most 3-D or 2-D optical information processing systems holographic techniques based on photorefractive material properties are widely used [3]. To make a photorefractive material from a LC (e.g. a photosensitive material useful for dynamic or quasi-permanent reversible hologram recording) a controllable process of photo-induced molecular rearrangement is needed. This was already demonstrated for numerous systems: guest–host dye-doped LCs [4–6], photoconducting LCs containing sensitizers [7–11], LCs containing photochromic compounds [12–15], LCs in intimate contact with photoresponsive substrate materials (semiconducting inorganic [16–19] or photoconducting organic [19–21] layers), polymer-dispersed LC composites [22,23], surface-command LC panels [24–26] and many other even more complex

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structures [27,28]. Most of the general solutions mentioned above have their limitations and drawbacks with respect to other known materials such as photorefractive inorganic crystals [29] and photorefractive polymers [30]. However, interest in this group of materials continues because of several properties typical of LCs, such as high nonlinear optical and electro-optic coefficients, large and easily achievable modulation of refractive indices ($\delta n \sim 0.2$) at a low operating voltage (switching fields typically are ~0.5 V/μm), and low light intensities required ($I \sim 10$ mW/cm^2). Moreover, LC panels can be fabricated at relatively low costs.

During our studies of dye-doped LCs [31] and hybrid photoconducting-polymer – LC structures [32] we examined systems exhibiting relatively fast relaxation of photoinduced gratings with decay times of holograms typically between 1 and 100 ms. These are not suitable for a prolonged optical memory on the time scale of minutes or hours. In this paper we would like to present a system showing long-lived surface-activated photorefractivity in a twisted nematic liquid crystal (TNLC) cell equipped with SiO_2 and polyimide insulating and alignment layers. We describe a mechanism which allows a quasi-permanent hologram recording in this system with the unique possibility of ac frequency tuning of the intensity of light diffracted at the generated refractive index grating. This opens up new ways of electronic control of light in OASLM.

2. Experimental

For the purpose of the present studies we prepared a LC cell with LC layer thickness of $L = 6.65$ μm. For the cell assembly we used 1 mm thick glass plates covered with ITO. The novelty is the use of a 50–100 Å thick SiO_2 blocking layer deposited onto the ITO covered glass. Over this layer we spin coated a polyimide layer of ≈100 nm thickness. After polymerization at 180 °C the polyimide layers were mechanically rubbed. We assembled a so called TNLC structure by orienting the rubbing directions of the opposite sides at a twist angle of 40°. Then we filled the cell with the nematic LC 4-(*trans*-4′-*n* -hexylcyclohexyl)isothiocyanatobenzene 6CHBT (Merck) which exhibits a positive dielectric anisotropy ($\Delta\varepsilon > 0$). The physicochemical properties of 6CHBT are the following: $\mu = 3.5$ D, $\Delta\varepsilon = +8.0$, $\varepsilon_{\parallel} = 12$, $\varepsilon_{\perp} = 4$, optical anisotropy $n_e = 1.6737$, $n_o = 1.5205$, $\delta n = 0.15$, elastic constants $K_{11} = 7.5 \times 10^{-12}$ N, $K_{33} = 9.5 \times 10^{-12}$ N, viscosity at room temperature (20 °C) $\gamma = 13.3 \times 10^{-3}$ N s/m^2, nematic to isotropic phase transition temperature 43.1 °C. The decay time of the orientational mode of 6CHBT in the LC layer can be estimated from the relationship $\tau_o = \gamma L^2/\pi^2 K_{av}$ and amounts to $\tau_o = 7$ ms, where the average value of the elastic constant $K_{av} = 8.5 \times 10^{-12}$ N has been used. 6CHBT was doped with 1.0% w/w of the dye molecule 1,4-dibutylamino-9,10-anthraquinone. As reported earlier this LC-dye solution shows photoconductive properties [8] when illuminated by light within an absorption band of the dye.

The used two-wave mixing technique (allowing for holographic recording of an image) and the sample configuration are schematically depicted in Fig. 1. The TNLC sample was illuminated by two coherent s-polarized beams ($\lambda = 532$ nm, $I_1 = 6.6$ mW and $I_2 = 4.65$ mW, beam diameter: 2 mm) from a 50 mW frequency doubled cw Nd:YAG laser. The bisectrix of the angle between the beams was precisely orthogonal to the sample surface. The power-meter head (Labmaster Ultima, Coherent) was placed at an angle corresponding to the first order diffraction direction to monitor an eventual appearance of self-diffraction of light. The optical intensity incident on the LC panel forms a sinusoidal grating $I(x) = I_o(1 + m\cos(qx))$ where $q = 2\pi/\Lambda$ is the wave vector of the interference pattern and Λ is the spacing of the grating, $I_o = I_1 + I_2$, and $m = 2\sqrt{I_1 I_2}/(I_1 + I_2)$ is a modulation factor (fringe visibility). In our experiment the important parameters were the following: $I_o = 358$ mW/cm^2, $m = 0.989$ and $\Lambda = 4.7$ μm.

3. Hologram recording

In contrast to systems studied earlier [8,31,32] no diffraction of light could be observed under dc voltage applied to the sample, except for momen-

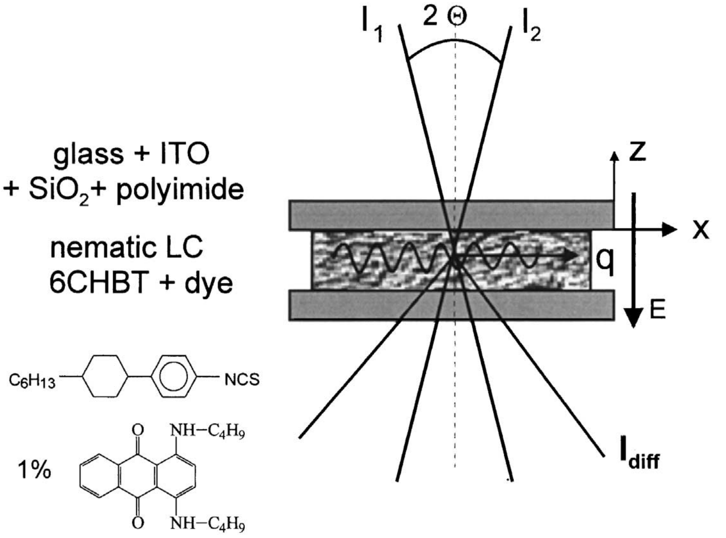

Fig. 1. Schematic view of the two-wave mixing experimental set-up and the TNLC sample structure – with the chemical formulae of the LC 6CHBT and the anthraquinone derivative dye (used in 1% (w/w) solution) given.

tarily appearing flashes of diffracted light when the dc voltage was switched between on and off states. We associate this major difference with the presence of the SiO_2 insulating layer which distinguishes this cell design from previously studied ones. The systematic studies of the properties of this system have lead us to the discovery of a unique mechanism/procedure by which holograms could be stored and read. The hologram recording, reading and erasing procedures are schematically shown in Fig. 2. The steps marked in the Fig. 2 are (i) *Neutral state:* Application of a sinusoidal wave ($f = 30$ Hz) of electric voltage with amplitude $U = \pm 3$ V to the sample under two-beam illumination does not lead to light self-diffraction. (ii) *Hologram recording procedure:* An offset voltage $U_o = -30$ V is added to the sinusoidal wave and the combined signal is applied during 1 min. Again no self-diffraction is visible during this step. (iii) *Hologram reconstruction by incident light:* Upon return to the conditions of (i) i.e. to sinusoidal wave of 30 Hz frequency and amplitude ±3 V strong self-diffraction is observed. No appreciable decrease of first order diffracted light intensity can be observed during several minutes. (iv) *Hologram erasing procedure:* An opposite offset

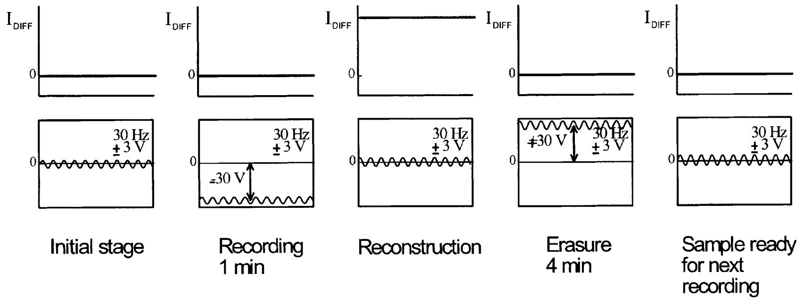

Fig. 2. Scheme of the full procedure leading to quasi-permanent hologram recording, its reconstruction, and its erasure in the TNLC cell.

voltage $U_o = +30$ V is added to the sinusoidal wave during 4 min. No self-diffraction is visible after this step; the hologram is erased. (v) The sample is ready for the next recording procedure.

4. Microscopic mechanism of grating recording

It is quite easy to propose a major mechanism leading to a quasi-permanent diffraction grating formation in the described LC cell. First of all we assume that incoming light is absorbed by the dye molecules which by interaction with LC molecules generate (in addition to the impurity ions) charge carriers (i.e. molecular ions resulting from the photoinduced charge transfer between a photoexcited dye and LC molecules [7]). Under relatively small alternative voltage applied to the sample no efficient redistribution of charge carriers takes place. However, in the presence of much larger dc voltage (here ±30 V) a considerable photocurrent flows through the LC layer. As follows from the Onsager theory [33], the probability that ion pairs escape from geminate recombination largely increases with an increase of the applied static electric field. Therefore we assume that under the influence of the external electric field E_o applied to the sample (here $\sim$5 V/μm), a certain fraction of the ion pairs formed by the light can dissociate.

During approximately $\sim$1 min exposure time a current of ions flows through the volume of the LC layer and reaches the surface. This flow is spatially nonuniform; it is appreciably more intense in the bright than in the dark regions of a light interference pattern. In a first approximation we neglect the lateral ion diffusion (along the grating wave vector $\boldsymbol{q}$) which influences the optical resolution of the grating. Lateral ion diffusion when compared with ion drift along the electric field lines can be neglected because in nematics the ratio of ionic mobilities along the director and perpendicular to it $\mu_{\|}/\mu_{\perp}$ is usually $\sim$3. Concerning the creation of a refractive index grating one may conclude from the experimental results that the ions of one sign (cations), after arriving at the electrode, are quasi-permanently adsorbed at the LC–polyimide interface or trapped within the polyimide–SiO_2 interface to form a spatially modulated surface charge density $\rho(x) = \rho_m \cos(qx)$. The crucial point in obtaining a charge distribution with considerable amplitude ρ_m is to achieve a very subtle balance between the amount of charge carriers trapped at the electrode and that being able to flow through the near electrode area to the ITO, either by tunneling of electrons through the SiO_2 layer or by photo-assisted current flow through a manifold of surface states of the defective SiO_2 layer [34]. A lack of the SiO_2 layer, for exactly the same cell construction and filling, resulted in structures which, although allowing for grating recording under dc field, lacked any memory effect. Apparently in these samples the polyimide layer was partially conducting, so a dc current could flow in the external circuit [31]. One can realize that the deposition of a too thick SiO_2 layer would completely prevent current flow in a circuit and quickly lead to wide spread of charged ions. This process usually causes a serious deterioration of the periodic modulation of the surface charge and consequently of the refractive index within the LC layer. However, no systematic studies of LC cells with different thicknesses of SiO_2 layers were performed by us.

We assume here that the spatially modulated electric field resulting from charge density modulation at the surface is the reason of molecular reorientation and the associated local refractive index change. The amplitude of the refractive index modulation is directly observed experimentally as a first-order diffraction intensity. Too long (above 1 min) as well as too short hologram recording leads to a decrease of the intensity of the diffracted light during the readout period. Prolonged illumination of a photoconductive LC system from one side also leads to the build-up of a photovoltaic electric field across the LC layer [35] as a consequence of nonuniform light absorption, $I = I_0 \exp(-\alpha z)$. Lowering of the dc field from 5 to 0 V/μm stops the ionic current flow almost completely. Then under appropriate conditions e.g. when the externally applied ac field acting on the nematic LC layer exceeds the threshold field of the Freédericksz effect ($E_{LC} \geqslant E_{th}$) the molecules are pretilted and, hence, susceptible to interactions of subtle electric fields, also of that coming from the surface charge distribution $\rho = \rho_m \cos(qx)$. Then the spatially modulated electric field $\boldsymbol{E}_{\mathrm{S}}(x)$ introduces a noticeable modulation of the director $\boldsymbol{d}(x)$ of the NLC; one should notice that two components of this field are present, E_{s}^{x} and E_{s}^{z}. The director is then modulated along the x-axis as well as along the z-axis (perpendicular to the sample surface) as the surface charge induced electric field is attenuated as $E_{\mathrm{s}}^{z}(z) = E_{\mathrm{s0}}^{z} \exp(-z/\lambda_{\mathrm{D}})$, where λ_{D} is the Debye length. The refractive index grating arising as a result of director reorientations and seen by a polarized laser beam has a rather complicated form, but it can be expressed as $n(x,z) = n_0(z) - \Delta n_{\mathrm{G}}(z)\cos(qx + \varphi)$. Due to the nonuniformity of the grating amplitude along the z-axis the integral over the sample thickness will correspond to the total phase modulation seen by an incoming plane wave. The amplitude of this grating Δn_{G} depends on the value of total optical birefringence $\delta n = n_{\parallel} - n_{\perp}$ of NLC, the value of $|\partial n/\partial E|$ at the operation electric field (E_{LC}), and the attenuation constant of the field $\boldsymbol{E}_{\mathrm{S}}(x)$ along the NLC layer depth (z). As a consequence of the phase grating Δn_{G} both the self-diffraction as well as the diffraction of light coming from a separate laser can be observed. Occasionally we used a He–Ne laser ($\lambda = 632.8$ nm) for measurements of the diffraction efficiency (defined as $\eta = I_{\mathrm{diff}}/I_{\mathrm{in}}$, where I_{diff} is the light intensity diffracted on the grating into the first order diffraction direction and I_{in} is the incident light intensity). The diffraction efficiency η in the case of a thin transmission phase grating is given by the square of the first-order Bessel function $\eta = |J_1(\phi)|^2$ with ϕ describing an effective phase change,

$$\phi(E) = \frac{1}{L}\int_0^L \frac{2\pi\,\Delta n_{\mathrm{G}}(z,E)}{\lambda}\,\mathrm{d}z, \tag{1}$$

where L is the thickness of the NLC layer and λ is the wavelength of the diffracted light. In the studied system the diffraction efficiency attained a maximum value $\eta = 0.004$. Within a Raman-Nath diffraction regime approximation [36] this means that the effective refractive index grating amplitude induced by light in the described TNLC structure amounts to $\Delta n_{\mathrm{G}} \cong 1.6 \times 10^{-3}$ which constitutes around 1% of the 6CHBT optical birefringence $\delta n = 0.15$ in the LC cell.

5. Experiments and discussion

We proved that the origin of the refractive index grating was due to surface charge modulation by performing an experiment in which we recorded a hologram, reconstructed it by one beam only, obtaining $I_{\mathrm{diff}} = 205$ μW, and next repeated the hologram reconstruction procedure 48 h after (during this period of time the laser was switched off as well as the voltage supply). Under usual hologram readout conditions (sinusoidal wave of 30 Hz frequency and amplitude ±3 V) we were able to see diffracted light coming from the previously written hologram. Despite the fact that the intensity of the diffracted beam dropped from 205 μW to the value of 80 μW, which constituted about 40% of the initially diffracted power, we so obtained strong confirmation that the grating originated from the trapped surface charge. This also means that the adsorbed or trapped charge at the surface can last more than two days in the darkness.

In Fig. 3 we show results of diffraction efficiency, η, measurements as a function of the dc field applied to the sample. In this case diffracted light appeared just for less than 1 s after field application. The points shown in Fig. 3 reflect the maximum of the signal picked up during field application. The explanation of signal disappearance could be the efficient formation close to the electrode area of a charge density so huge that any

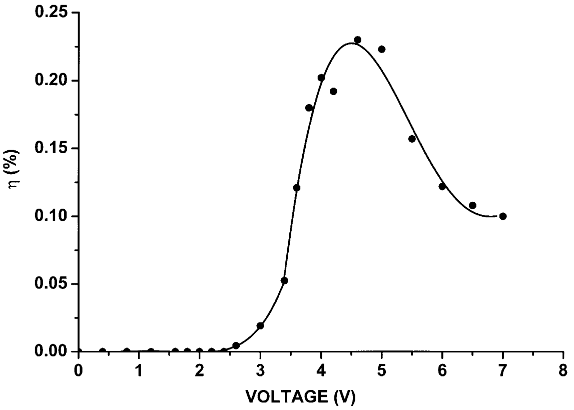

Fig. 3. First order diffraction efficiency η versus bias voltage applied to the sample. (The line is a guide to the eye.)

periodic modulation was smeared out. The blocking electrodes obviously help to effectively diminish the potential drop across the LC layer and disable the optimal conditions for molecular reorientations. These optimal conditions, however, are met in a relatively small range of the applied electric field. The maximum value of diffraction efficiency measured in the sample amounted to 0.4% and occurred at $\approx$4.8 V. It should be mentioned, however, that the sample history played a certain role during this type of measurements.

We noted that the diffraction efficiency of the hologram written according to procedure (ii) depended on the frequency of the alternating electric field used in the readout procedure (iii). An example of the diffraction efficiency dependence of η on the frequency f of the electric field is presented in Fig. 4. Measurements were done during the hologram reconstruction process with a sinusoidal wave of amplitude $\pm$3 V applied to the sample. This amplitude was chosen on the basis of studies of the dependence of light diffraction on the value of the electric field amplitude: The diffraction signal shows a maximum at $\approx$3 V and drops to unmeasurable values for amplitudes lower than 2 V and higher than 5 V. From Fig. 4 it follows that the highest response was observed at around 30 Hz, whereas at 400 Hz almost no diffracted light could be detected anymore. This is an interesting result as it allows to tune the diffracted light intensity by merely tuning the frequency of electric signal applied to the sample. The data presented in Fig. 4 were highly reproducible, although with some hysteresis when frequency was increased and then decreased (cf. arrows in Fig. 4). Such a behavior might be connected with some specific charge relaxation process taking place in the studied NLC sample. As was shown in Section 2, the reorientation relaxation time was estimated to amount to $\tau_o \approx 7$ ms, which means that for ac frequencies above 300 Hz, molecules practically cannot follow the field changes. Moreover, under these conditions the LC is so stiff that its molecules cannot effectively reorient according the weak spatial electric field modulation caused by the charge density modulation at the surface.

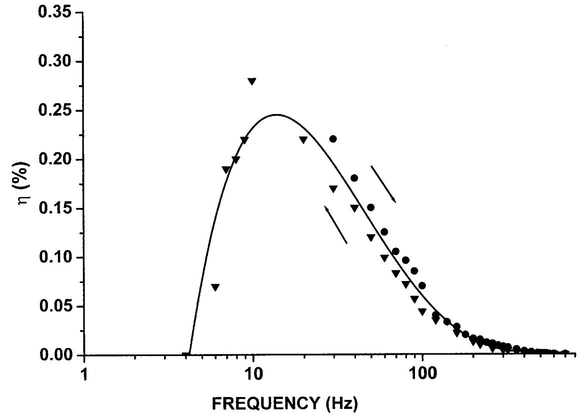

Fig. 4. Dependence of the diffraction efficiency η on the logarithm of the ac electric field frequency f as observed for the TNLC sample. Arrows point to the frequency change cycle and the line is a guide to the eye.

During the hologram reconstruction procedure the diffraction grating slowly disappeared; we call this process *hologram self-erasure*. An application of an electric field of opposite sign to that used during recording led to fast hologram erasure. From the practical point of view it was interesting to study the time constantτ_{s-e} of self-erasure of the hologram as well as the conditions and the time constant τ_e of its erasure. The self-erasure time constant was measured by monitoring the light power diffracted into first order just after completing a hologram recording procedure ($\pm$3 V, 30 Hz, offset +40 V, 1 min) by blocking one of the writing beams and setting the offset voltage to 0 V. The kinetics of decay of the diffraction efficiency under the one beam illumination condition is shown in Fig. 5. The diffraction efficiency decay $\eta(t)$ is well approximated by the exponential decay function:

$$\eta = \eta(0) + A\exp\left(-\frac{t}{\tau}\right), \qquad (2)$$

where τ is the time constant of the relaxation process and $\eta(0)$ and A are constants that depend on characteristic parameters of an experiment, in particular on the hologram reading beam intensity I_i. In the case presented in Fig. 5 the time constant τ_{s-e} amounts to 16.3 min, $\eta(0) = 1$ (normalized to 6.3×10^{-5}) and $A_{s-e} = 3.8 \times 10^{-4}$. In addition we show in Fig. 5 the grating decay kinetics under erasure conditions i.e. under application of an

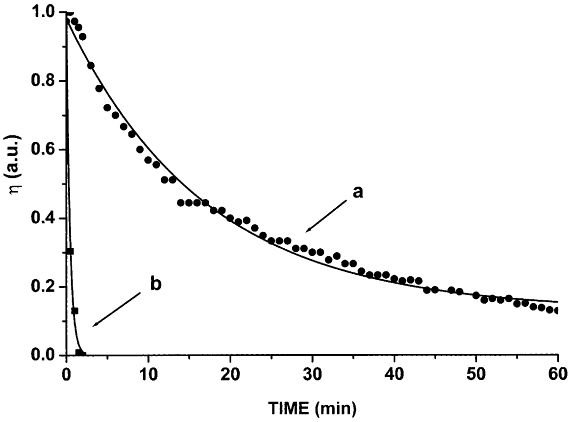

Fig. 5. Kinetics of decay of diffraction efficiency under the one beam illumination condition and at an offset voltage of 0 V (curve a); kinetics of grating erasure measured under the one beam illumination condition at an offset voltage equal to that applied during hologram recording but opposite in sign (curve b). Both curves represent a fit to the experimental points according to Eq. (2).

electric field of a sign opposite to that used during recording under one-beam illumination. In this case the time constant for the erasure procedure was only $\tau_e = 0.4$ min. This may be regarded as a confirmation that only one charge carrier sign is effectively trapped at the surface. It is worth mentioning here that by using a UV lamp the holograms written in the TNLC system could be erased within a few seconds, because UV light can more efficiently than 532 nm green light depopulate deep traps and release trapped charge carriers.

The twisted nematic structure is unique also in its polarization properties [37]. When the grating is recorded in TNLC it can be read out for any linear polarization of the incoming reading beam. The diffracted power, however, depends on the front layer alignment direction with respect to the beam polarization plane. These changes of η are shown in Fig. 6, where two minima and maxima are observed for full angle (360°) rotation of the polarization plane of the inciding beam. Diffraction efficiency maxima (~0.04%) occur for the beam polarization plane parallel to the alignment direction and minima (~0.008%) for the perpendicular configuration.

An observation of the TNLC cell described so far, under a polarization microscope with dc

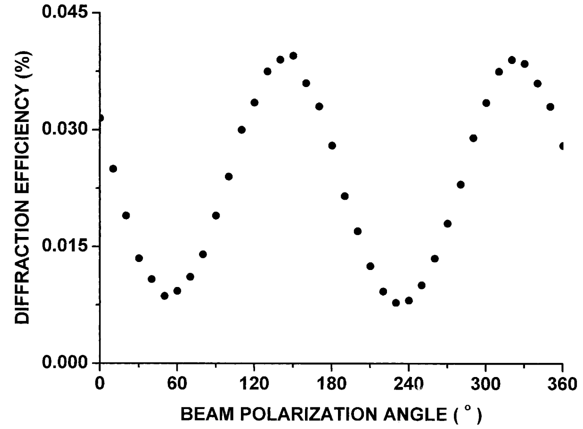

Fig. 6. Changes of diffraction efficiency η as a function of the orientation of the plane of polarization of the reading laser beam with respect to the alignment direction of the surface polyimide layer.

voltage applied revealed the position of the electric Freedericksz transition at 1.9 V (0.286 V/μm) as shown in Fig. 7. We analyzed the mean pixel intensity of the photographs of TNLC taken under crossed polarizers as a function of the dc voltage applied to the sample. The small peak around 2.6 V followed by a minimum at 3.2 V could be understood on the basis of the theory of light transmission through a twist cell as discussed in [38].

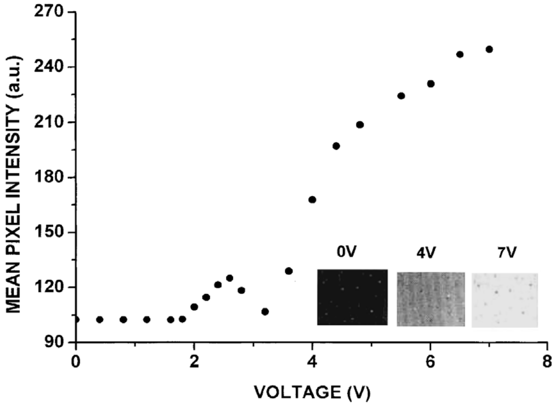

Fig. 7. Polarizing optical microscope studies of the TNLC cell as a function of the bias voltage. The inset shows examples of microscope images taken at a magnification of 100 at 0, 4 and 7 V, respectively. The mean pixel intensity of the photographs (taken under crossed polarizers conditions) indicates the amount of light emerging from the sample.

The inset of Fig. 7 shows examples of observed images at 0, 4 and 7 V. For voltages higher than 7 V no further changes of the microscopic image were observed.

It is worth mentioning that successive repetitions of the grating recording procedure in the same spot of the sample, while rotated in its plane by a small angle allowed multiple grating recording. All gratings could subsequently be read simultaneously by a single beam during the hologram reconstruction procedure.

6. Conclusions

In conclusion, we have demonstrated a new concept of a TNLC cell equipped with a blocking SiO_2 layer and a polyimide orienting layer, which can be optically addressed and which allows to record quasi-permanent transmission type phase gratings or holograms. We described the procedures used for hologram recording, erasure and reconstruction, and the control of these processes by the frequency of an ac electric field. Under illumination the surface is loaded with charge which is photogenerated in the bulk of the LC layer. We believe that structures similar to the one described in the present work will soon lead to the development of valuable new photonic real-time holographic LC devices.

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