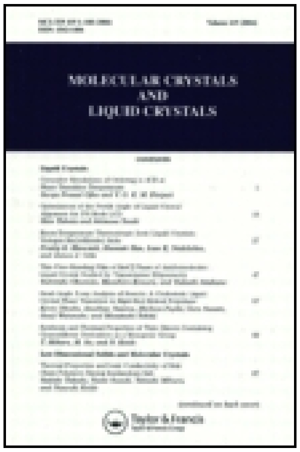

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Storage of erasable laser induced holographic gratings in low molar mass cholesteric liquid crystals

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Storage of erasable laser induced holographic gratings in low molar mass cholesteric liquid crystals

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Abstract. We succeeded in storing optical information with low-molecular-weight cholesteric liquid crystals. Nematic mixtures of glassy compounds which are chiralized by the addition of small amounts CB15 (BDH) have been used. Reversible optically induced holographic gratings were stored both below the glass transition temperature and above the glass transition in the temperature region of high viscosity. Erasure can be achieved by local heating of the sample making possible to use the material for multiple storage processes. A mechanism to explain the formation of the grating is discussed.

Introduction.

At the present time optical information storage systems are under strong investigation to fulfill the challenging demands of today's and future optical information and data processing techniques. Liquid-crystalline polymers have been extensivly studied in recent years owing to their suitability for optical write-once as well as reversible optical information storages. The information is e.g. written in the polymer above the glass transition and can be frozen-in by cooling the polymer below the glass transition temperature. Erasure is achieved by heating up the sample above the glassy state or the clearing point of the polymer.[1,2]

However, many practical applications of polymers are somewhat limited because of their high viscosity, which can be several orders of magnitude larger than their low molar mass analogues.[3] Furthermore, low molar mass liquid crystals are easier to synthesize in comparison to many polymeric materials and well-known prealignment techniques together with other established liquid-crystal technologies would eventually allow the mass-production of optical storages at the present state of the art. As a consequence glassy low-molecular-weight liquid crystals seem to be very promising candidates for the realization of reversible optical information storage media.

Different techniques have been reported to achieve optical data storaging.[4,5] We used the well-known optical holographic grating technique, realized by interference of two coherent laser beams in the sample.[6] The resultant intensity grating is converted into a temperature grating by opto-thermal heating. Opto-optical or electro-optical switching in the areas of high temperature leads to a periodical deformation or unwinding of the cholesteric helix thus containing the optical information of the two writing beams. After switching off the writing laser beam the grating remains until the liquid crystal is heated up once more above the glass transition temperature.

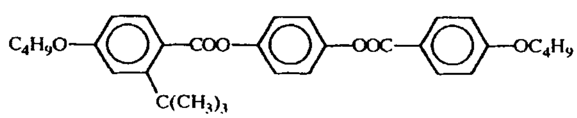
Experimental part.

Substances.

For our investigations we have choosen two eutectic mixtures. Each of the liquid-crystalline substances has a glassy nematic state.[7] Molar fractions of the pure compounds (A-E), onset glass transition temperatures (lower limit of the glass transition interval) and phase sequences (temperatures in °C) of the mixtures (I, II) are given in the following:

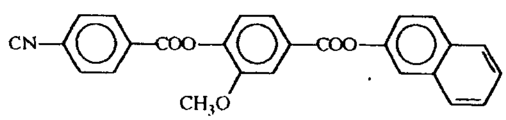
I

A



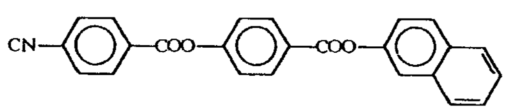
$x_A = 0.7$

B



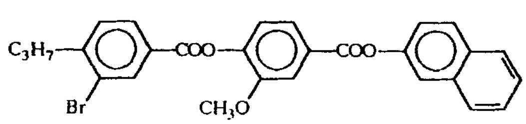
$x_B = 0.09$

C



$x_C = 0.12$

D

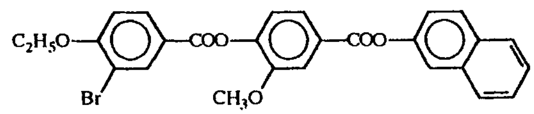


$x_D = 0.09$

N 122 I; $T_g^{on} = 10$ °C.

II

E



$x_E = 0.8$

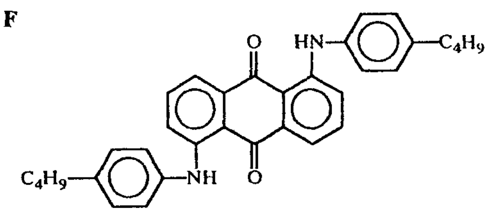
and compound C $x_C = 0.2$

N 140 I; $T_g^{on} = 34$ °C.

Substance A has been synthesized by Weissflog.[8,9] The others were synthesized by Schäfer *et al.*.[10] The phase sequences are measured with a DSC-7 calorimeter (Perkin-Elmer). Both mixtures were chiralized by the addition of 10 to 40 wt.-% CB15 (BDH) in order to obtain a better orientation of the sample and a positive dielectric anisotropy for the electro-optical switching.

Additionally, we investigated cholesteric samples with small amounts (< 0.5 wt.-%) of an dichroic anthraquinone dye (BDH) to improve the orientation and to enhance the absorption of the writing laser wavelength:

F



Sample preparation.

In our studies we used cells of 10 μm thickness consisting of two conductive glass plates (ITO-coatings) with planar alignment. To improve the alignment the filled cell was kept some time at elevated temperature a few degrees below the clearing point.

Holographic measurements.

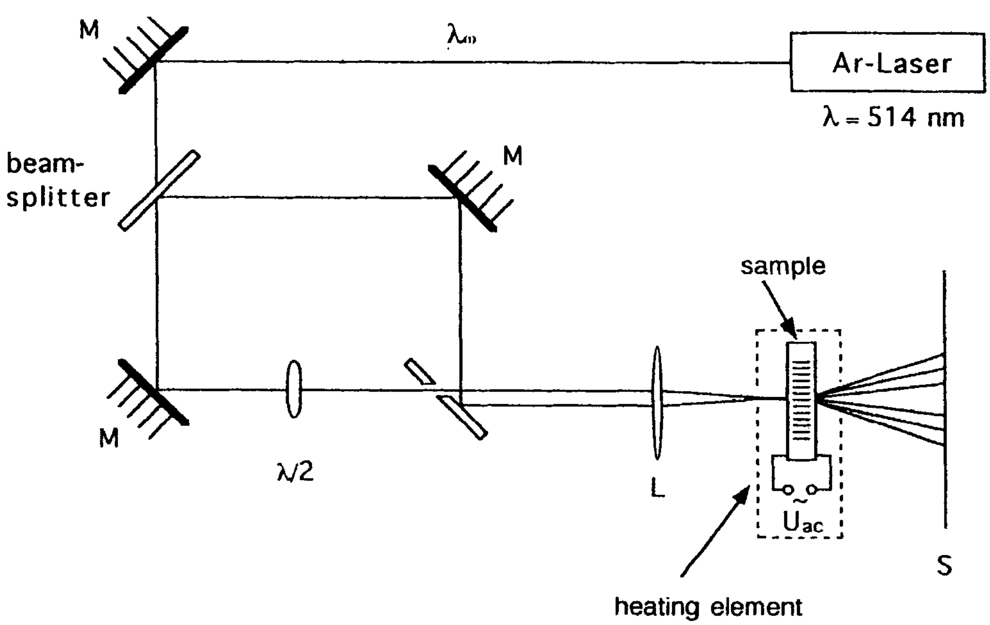
The experimental set up is shown in Fig. 1a. Two coherent writing beams obtained from an argon-ion laser ($\lambda_W = 514$ nm) in a symmetrically Mach-Zehnder beamsplitter are crossed at the sample at an intersection angle of 2Θ. The induced optical grating can be observed as a selfdiffraction pattern on a screen or is detected by diffraction of a weak probe beam and a calibrated photodiode. A $\lambda/2$-plate is placed in one arm of the interferometer to adjust the relative polarizations of the writing beams. Intensity gratings are obtained with parallel, and polarization gratings with perpendicular polarized beams.[11] The intensity of the writing beams was varied between 0.1 and 0.6 W. The beam radius w_0 was about 90 μm. Additionally we applied an AC voltage of 100 V with a frequency of 1 kHz across the samples without the dye.

The grating period Λ is characterized by the wavelength of the writing beams and the intersection angle:[12]

$$\Lambda = \frac{\lambda_W}{2 \sin\Theta}. \tag{1}$$

The typical grating period in our experiments was 50 μm. All experiments were performed with samples which are already cooled down below the glass transition temperature or somewhat above the glass transition in the temperature region of high viscosity. The grating was read out with one weak single beam obtained from the same argon-ion laser (Fig. 1b).

(a)



(b)

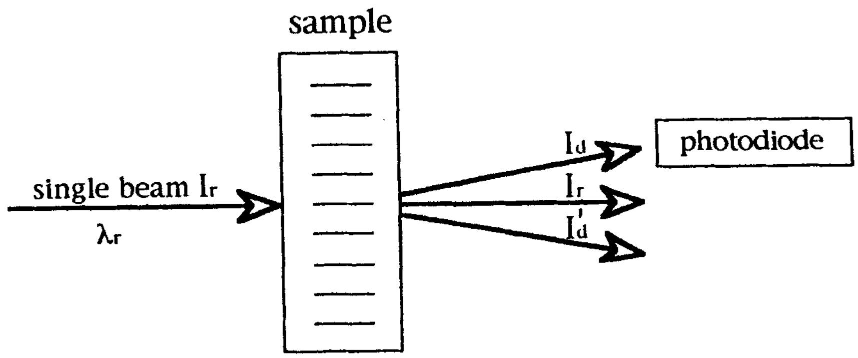


Fig. 1. Experimental set up for holographic measurements.

λ_w and λ_r denote the wavelengths of the writing and reading laser beam,;I_r, I_d and I_d' are the intensities of the diffracted reading beams (M = mirror, L = lens, S = screen).

(a) Arrangement for the writing process.

(b) Arrangement for the reading process.

The diffraction efficiency η of the hologram is determined by the ratio of the intensity of the diffracted beam I_d to the input intensity of the reading beam I_r:

$$\eta = \frac{I_d}{I_r}. \quad (2)$$

The stored information is erased by locally heating the sample with a single laser beam of higher power or by heating up the whole device. The temperature which is necessary to erase the information must be considerably higher (at least more than ten degrees) than the glass transition temperature.

Results and discussion.

A typical temporal development of the diffraction efficiency during a writing and reading cyclus is shown in Fig. 2a. The applied voltage is U_{ac} = 40 V during the writing process. The grating appeared in a few minutes after switching on the intensity grating. The optical information was fixed by switching off the voltage and blocking one of the writing beams whereas the remaining beam was weakened at the same time and used for reading now. After switching off the writing beam the diffraction efficiency of the hologram decreases by about 2 % but then stabilizes at about 8 % for times as long as two hours and more. The cholesteric liquid crystal used in Fig. 2a is not in the glassy state, but above the glass transition in the region of high viscosity. As depicted in Fig. 2b a similar result is obtained for the glassy state.

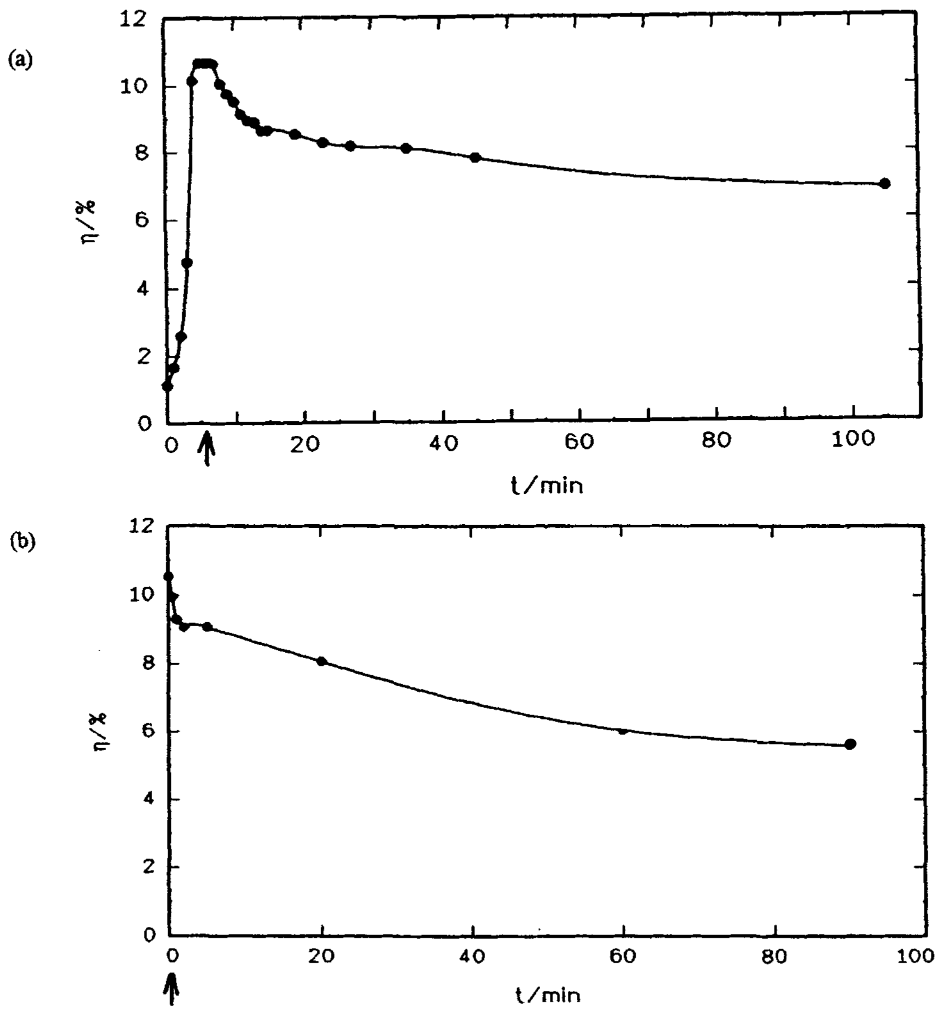


Fig. 2. *Diffraction efficiency η as a function of time*

a) in a mixture of 36.9 wt.-% CB15 in I (T_g^{on} = -20 °C). After the holographic grating appeared, the writing beam and the applied voltage were switched off. The temperature on the surface of the cell was 8 °C (writing: I_W = 0.6 W, U_{ac} = 40 V; reading: I_r = 0.2 W, U_{ac} = 0 V).

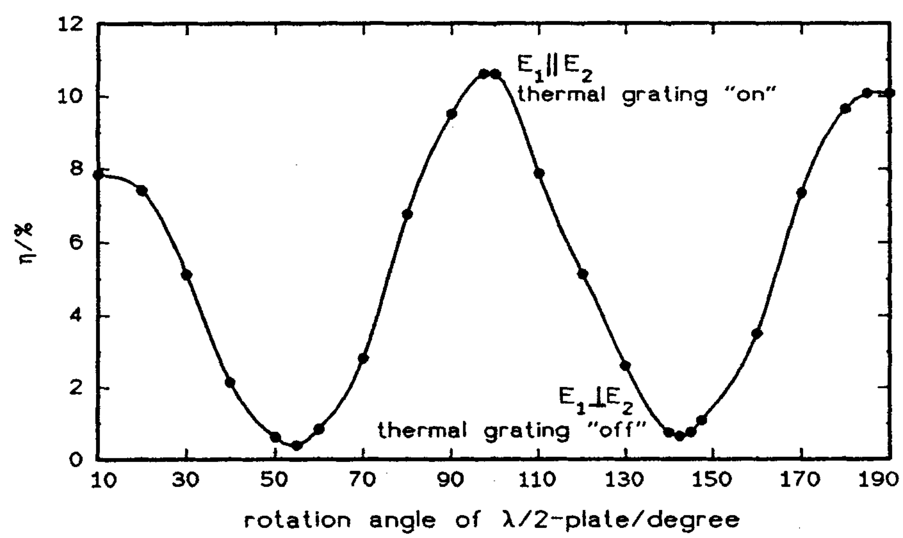
b) in a mixture of 27.13 % CB15 in II (T_g^{on} = 8°C; writing: I_W = 0.2 W, U_{ac} = 100 V; reading: I_r = 0.16 W, U_{ac} = 100 V).

The arrow denotes the time, where the writing beam was switched off.

To prove that the writing of the grating is of thermal origin, we changed the relative polarizations of the laser beams from parallel to perpendicular polarizations. As shown in Fig. 3a, a maximum diffraction efficiency is obtained if the electric field vectors are parallel and a minimum if they are perpendicular to each other. In the parallel orientation we induce a thermal grating via absorption of the input intensity, whereas in the perpendicular orientation the two linearly polarized plane waves do not interfere and are not able to induce a thermal grating.

For an intensity grating in the sample the temperature profil in the cell corresponds to the variation of the laser intensity, as indicated in Fig. 3b. Absorption and heat production is provided in the ITO-coatings or the dye, respectively. As a consequence, a periodically unwinding or distortion of the cholesteric helix is obtained in the high temperature regions of the grating by means of the optical field itself or under the influence of the electric field applied additionally. It turns out that the latter process is mainly responsible for the grating formation in non-dyed samples, whereas no electric field is necessary if dyes are added (e.g. with compound F). This may indicate that we have different mechanisms for the two kinds of materials under investigation. It should be noted, however, that the exact role of the dye is not clear in at the present stage of our investigations. Beside simply enhancing the absorption, there we observed that the alignment of dyed samples is improved and scattering losses are much weaker. To understand the whole influence on the grating formation further investigation is still needed.

(a)



(b)

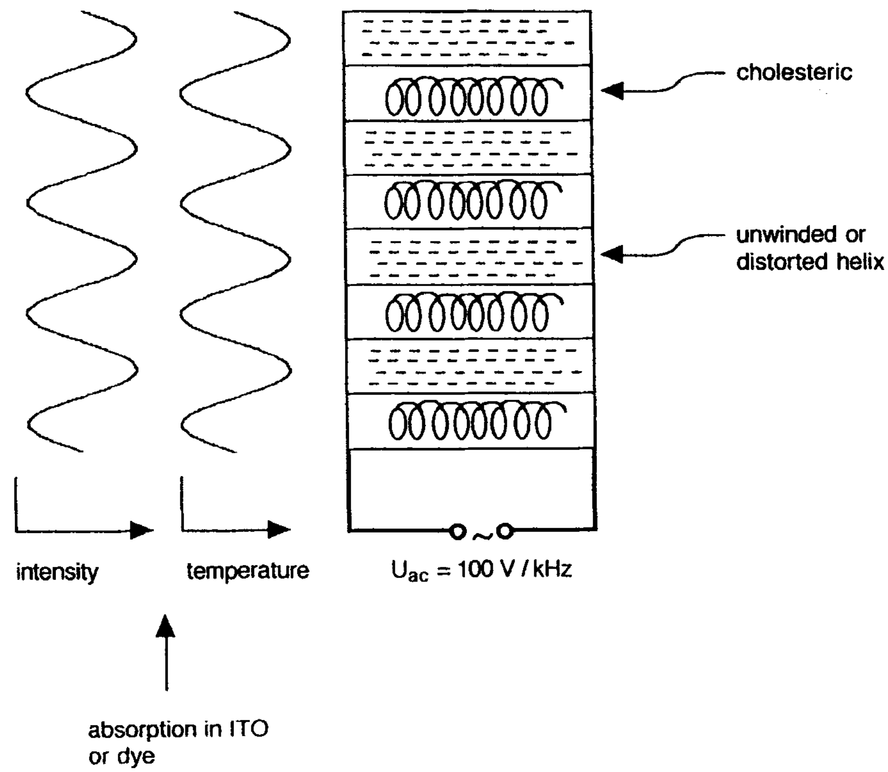


Fig. 3. a) Diffraction efficiency for intensity and polarisation gratings.

If the electric field vectors of the two linearly polarized plane waves are parallel ($E_1 \| E_2$), we observed a maximum in the diffraction efficiency and if they are perpendicular ($E_1 \perp E_2$) there is a minimum in the diffraction efficiency. Composition of the mixture is 36.9 wt.-% CB15 in I (writing: I_w = 0.6 W, U_{ac} = 40 V; reading: I_r = 0.17 W, U_{ac} = 0 V).

b) Mechanism of laser adressed electro-optical switching of grating. Additionally applied electric field is only required in the samples without dye.

Fig. 4 shows exemplary the dependence of the first order diffraction efficiency on the intensity of the writing laser beams in a mixture of 9.58 wt.-% CB15 and 0.1 wt.-% dye (compound F) in II. The diffraction efficiency in the pure cholesteric mixtures is in the same order of magnitude. With increasing intensity the diffraction efficiency increases first until a maximum of η = 12 % - 20 % is reached and than decreases.

This result shows that the induced grating in a slightly absorbing cholesteric liquid crystal can not be described in terms of a pure thin phase-grating, because the maximum diffraction efficiency should reach about 30 % in this case.[12] The experimental result can be explained in principle, however, if we assume that the distorted cholesteric liquid crystal leads to phase- as well as amplitude-modulations. This seems likely to be the case since any pitch dilatation or distortion of the cholesteric helix usually results in a modulation of the birefrigence and the selective reflectivity.

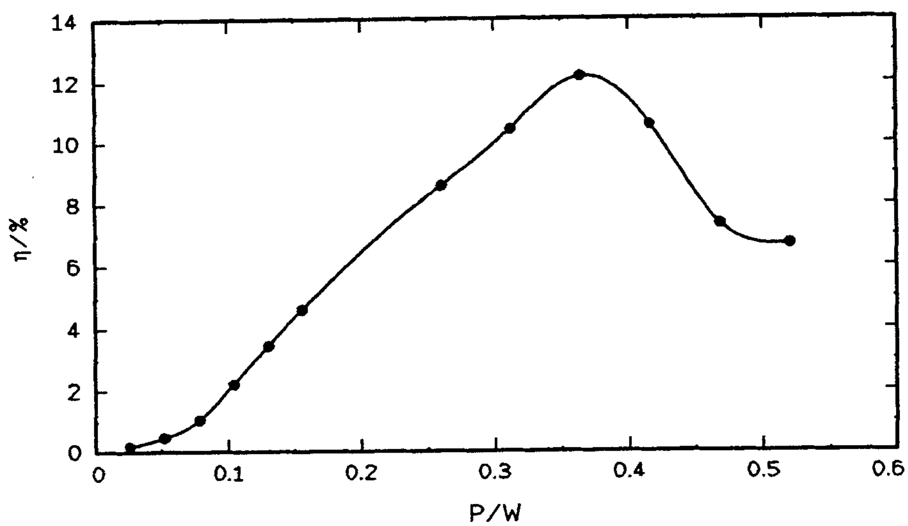


Fig. 4. Dependance of the first diffraction order efficiency on the intensity of the writing laser beams in a mixture of 9.58 wt.-% CB15 and 0.1 wt.-% dye (F) in II (T_g^{on} = 23°C; $I_W = I_r$; $U_{ac} = 0$ V).

Conclusion.

Storage of erasable holographic gratings in low molar mass cholesteric liquid crystals has been demonstrated. The storage-mode with grating lifetimes of two and more hours has been obtained in the glassy phase of these materials or in the vicinity of the glass transition on the high temperature side, where the viscosity of the mesophase is extremly high. The laser-induced grating formation is explained by local opto-thermal heating up at temperatures above the transition and a simultaneously opto-optical or electro-optical deformation of the cholesteric helix in the region of enhanced temperature. The frozen-in optical information persists until the next heating cyclus starts, which may be either locally in the spot of an erasing laser or by heating up the whole sample.

The maximum observed diffraction efficiency in the thin grating limit has been about 20 %, whereas typical storage-mode efficiencies range around 8 %.

Acknowledgments.

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