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WRITING AND ERASURE OF HOLOGRAPHIC GRATINGS IN DYE-DOPED POLYMER DISPERSED LIQUID CRYSTALS

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Abstract We Demonstrate that is possible to write and erase holographic gratings in dye-doped polymer dispersed liquid crystals (PDLC). PDLC samples were irradiated after complete curing using two beams in an interferometric geometry. In this way permanent holographic gratings were written with a resolution up to 100 lines/mm. The erasure was obtained by a single beam, while successive writing were possible again by two beams interference, without any degradation after many cycles. The origin of the effect can be the onset of periodic distribution of droplets of different shape and spatial diffusion of the dye molecules.

INTRODUCTION

In the last years considerable interest has been devoted to holographic and diffractive technology owing to possible applications for optical processing and storage[1,5]. Many applications require the control of the diffraction efficiency and writing/erasing procedure in order to get reprogrammable interconnects. Electrically switchable gratings with high diffraction efficiency have been already realised using PDLC[6,7]: in that case writing was performed during the polymerisation of the matrix to get a grating distribution of liquid crystal droplets embedded in the polymer. Then gratings are permanently written in the material but their diffraction can be modulated by an electric field applied to the sample. In fact the droplets are realigned by the field and their refractive index can match the one of the matrix, thus optically erasing the grating. Switching-off the field results as a reset of the optical grating.

Recently Kitzerow[8] have reported the possibility of writing gratings in dye-doped PDLC after the matrix polymerisation. He suggests that the temperature modulation induced by the interfering beams change locally the stability condition of the liquid crystal-polymer mixture leading to formation of droplets of different size which would realise the grating structure. However erasure of these gratings was not reported and the electrical modulation was only partially possible.

In this paper we demonstrate the 100% writing/erasure capability in dye doped PDLC under many writing/erasure cycles, moreover we show that diffraction efficiency can be totally modulated by an applied electric field. The impinging energy required for the writing/erasure procedure is about 10^3 J/cm^2.

EXPERIMENTS

The experimental set-up (fig.1) is the standard one for investigation of nonlinear gratings in liquid crystals. An Ar^+ laser beam (λ=514nm) is spliced in two beams of intensity I_1 and I_2 having an intensity ratio $I_1/I_2 \approx 1$, which are recombined at a crossing angle θ on the PDLC sample. Variation of this angle allows to change the pitch Λ of the induced gratings ($\Lambda=\lambda/[2\sin(\theta/2)]$). The beams are weakly focused (spot diameter≈0.5 mm) to reach the intensity necessary to write permanent gratings, in fact at low intensities only dynamic gratings are possible, as already reported by Simoni et al.[9]. The PDLC samples were obtained by polymerisation induced phase separation (PIPS), controlled by temperature, starting from an homogeneous solution of i) nematic liquid crystal E7 by BDH, ii) epoxy fluid prepolymer (mixture of Epon 815 by Shell Chemical, MK107 by Wilmington Chemical and Bostik B by Boston, iii) Capcure 3-800 as curing agent and iv) the azo-dye D2 by BDH as liquid crystal dopant. The mixture was placed between ITO coated conductive glasses with 36mm thick Mylar spacers. The intensity distribution of the interfering beams gave rise to gratings structure in the samples, which could be easily observed only after the exposition of the exciting beams due to the diffraction effect on a linearly polarised He-Ne laser used as probe to study the phenomenon.

However in order to get a meaningful diffraction efficiency η value we have not used the usual definition $\eta_i = I_i/I_{inc}$, being i the order of diffraction and I_{inc} the probe beam incident intensity. In fact the sample is opaque and many losses due scattering are present, thus we have chosen an alternative definition as $\eta_i = I_i/I_T$, being I_T the total transmitted intensity (summed over all the order of diffraction). Measurements were also taken for different polarisation state of the probe beam, namely parallel and orthogonal to the exciting beam.

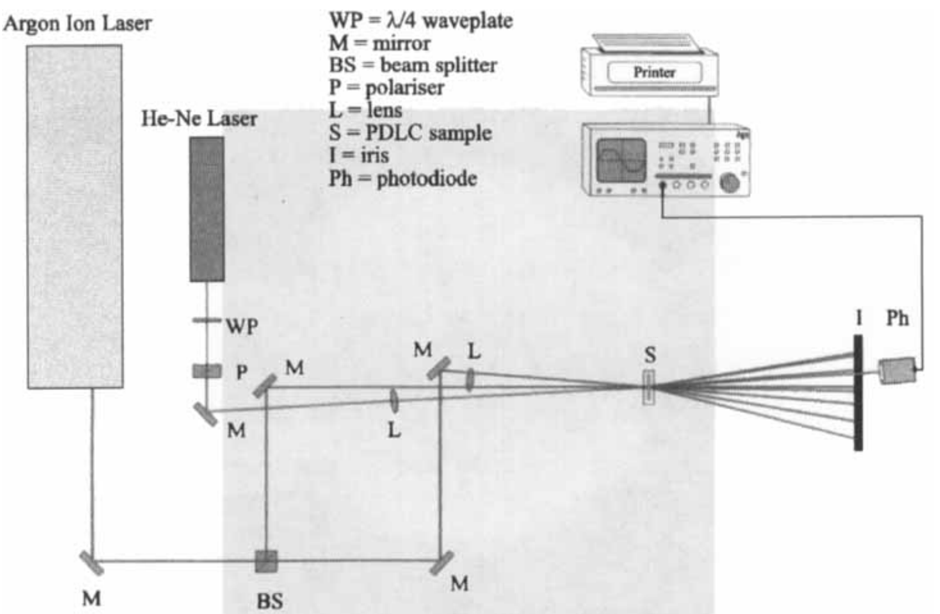

FIGURE 1 The experimental apparatus. (See Colour Plate V).

After studying the characteristics of the optical grating generated by two beams irradiation, one beam was blocked and the intensity of one single beam increased. Under this condition the diffraction effect disappeared indicating the erasure of the grating. A new irradiation by two interfering beam led to a new grating formation, and this writing/erasure procedure could be repeated several times on the same spot of the sample. All these results were confirmed by observation under a polarising microscope, which could easily show the written and erased gratings.

RESULTS AND DISCUSSION

The permanent grating formation in the sample needs few seconds of irradiation at 200-300 W/cm^2 light intensity; the probe diffraction appears only when the exciting beams are removed. An example of such a grating is shown in fig.2, which reports the observation by optical microscope. No degradation of the grating was observed after several months.

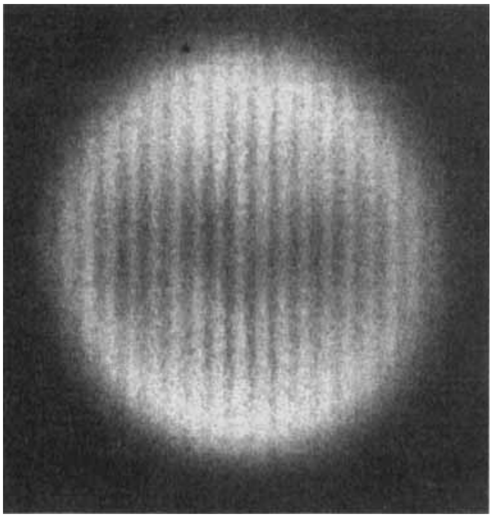

FIGURE 2 Optic microscopy observation of a grating with Λ=25 μm.

The efficiency of the grating was studied for different values of the grating constant Λ in order to get information on the spatial resolution which can be achieved by this method. The results are reported in fig.3, where η_{par} and η_{ort} refer to probe beam polarisation parallel and perpendicular with respect to the exciting beam. A clear anisotropic behaviour is observed since η_{par} is always lower than η_{ort}, which has still a high value at about 40 lines/mm. This observation suggests that these gratings could be constituted by a spatially periodic distribution of elongated droplets. Such droplets deformation may depend on a combination of thermal and mechanical effects induced by the temperature gradient over the gratings pattern, which leads to a liquid crystal droplets shape distribution which in turn may explain the observed anisotropy of the diffraction efficiency.

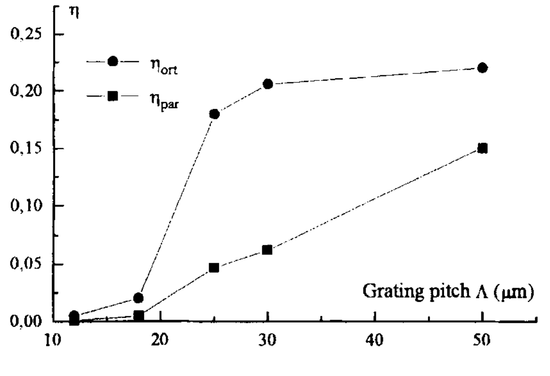

FIGURE 3 First order diffraction efficiency versus grating pitch Λ

On the other hand the experimental observation that probe diffraction is detected only after removal of the exciting beams says that the main contribution to the grating comes from the liquid crystal droplets, which become isotropic under irradiation matching the refractive index of the polymeric matrix and making invisible the recorded grating. Moreover the role of the dye is also not negligible in fact after the first irradiation all the excited spots appeared more transparent than the surrounding coloured area showing that a diffusion process of the dye molecules occurred. Because of the anisotropy of the dye this process might be more efficient for the excited molecules (with long axis parallel to the exciting beam) leaving in the excited area a majority of molecules with their long axis perpendicular to the light beam polarisation molecules and this effect should follow the intensity modulation of the interfering beams. This effect might be responsible of the formation of an amplitude grating due to a non uniform distribution of dye molecules which would be superimposed to the phase grating due to droplets' shape distribution described above. The presence of mixed phase/amplitude gratings seems evident from the relative efficiency analysis of the different orders of diffraction.

A very important feature of the created gratings is the possibility of erasing them by using a single laser beam with the same energy as the one used to write the permanent grating. After the erasure it is possible again to write the grating in the same spot and to repeat the cycle several times. One example of observations made under optical microscope for the writing/erasing process is reported in fig.4. It was always detected a reduction of the anisotropy effect (η_{ort}/η_{par}) after the second recording, anisotropy which kept constant for all the successive writing.

a) b) c)

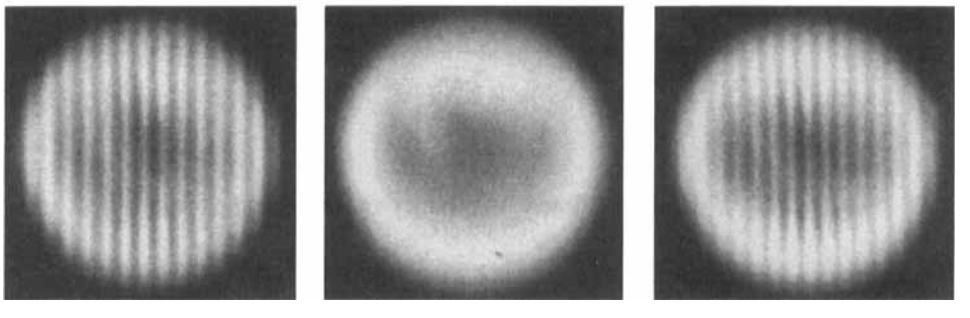

FIGURE 4 Optic microscopy pictures of permanent gratings: a) first writing, the diffraction efficiencies are η_{par}=6.4%, η_{ort}=21% ; b) erasing, the normalised transmitted intensity values are I_{par}/I_{inc}=16%, I_{ort}/I_{inc}=8% (the initial values are I_{par}/I_{inc}=0.01%, I_{ort}/I_{inc}=0.02% ; c) second writing, the diffraction efficiencies are η_{par}=7%, η_{ort}=10%.

A more systematic study of these effects together with a Scanning Electron Microscope (SEM) investigation is currently under way in order to understand the origin of the grating formation and erasure which can be also useful to optimise the process in view of possible applications.

The possibility of modulating the grating efficiency by an applied voltage has been also checked. After creating the grating a single square pulse (V=100V) was applied and the intensity of the first diffracted beam detected. The voltage application leads to a full signal switch off due to the induced index matching between the liquid crystal droplets and the polymeric matrix (see fig.5).

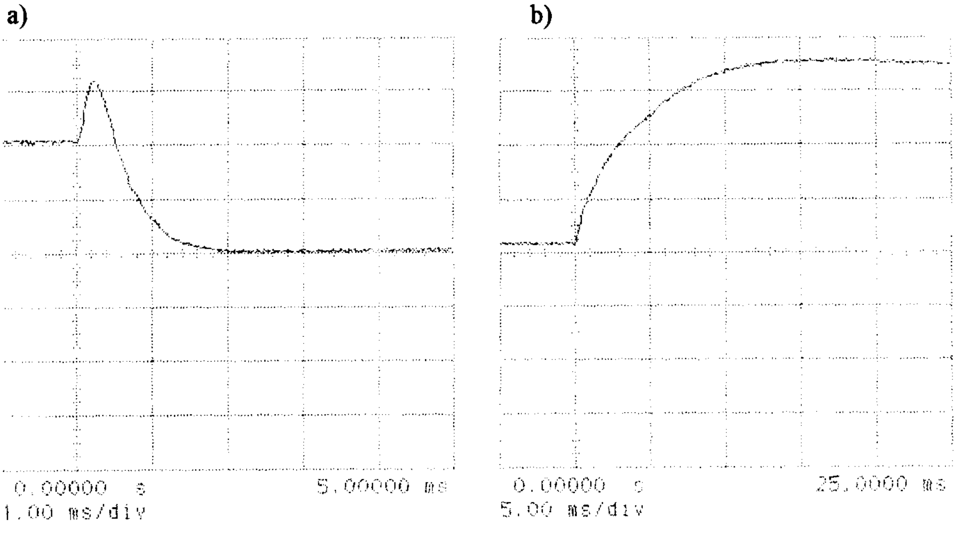

FIGURE 5 First order diffraction intensity: a) Field on, the extinction time is 1.5 ms; b) Field off, the rise time is 8 ms.

This observation confirm that the liquid crystal droplets are important in determining the characteristics of the gratings.

In conclusion we have demonstrated that in dye-doped PDLC it is possible to write and to erase holographic gratings several times in the same spot. At the moment high diffraction efficiency has been reached for spatial resolution higher than 40 lines/mm with writing energy density of about 10^3 J/cm^2. The investigation of the origin, not yet clear, of the writing/erasure process can easily lead to optimisation of spatial resolution and of sensitivity (lowering the writing energy density) making these physical systems attractive has high density storage media.

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