Molecular Crystals and Liquid Crystals Science and Technology. Section A. Molecular Crystals and Liquid Crystals

Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl19

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Version of record first published: 24 Sep 2006

To cite this article: L. Lucchetti, F. Simoni & Y. Reznikov (2001): Director Reorientation Induced by Short Pulsed Irradiation in Dye Doped Liquid Crystals, Molecular Crystals and Liquid Crystals Science and Technology. Section A. Molecular Crystals and Liquid Crystals, 359:1, 131-136

To link to this article: http://dx.doi.org/10.1080/10587250108035574

Mol. Cryst. and Liq. Cryst., 2001, Vol. 359, pp. 131-136
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Director Reorientation Induced by Short Pulsed Irradiation in Dye Doped Liquid Crystals

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We report the effect of light-induced anchoring in dye-doped liquid crystals using a single 4 ns pulse as excitation source. The kinetic of director reorientation has been recorded by a pump-probe technique. Fast recording of high resolution holographic gratings (more than 500 lines/mm) has also been obtained.

Keywords: Dye doped liquid crystals; Director reorientation; Short pulsed irradiation

INTRODUCTION

The research of new materials for optical storage is at present a field of intense study. The investigation of media with optical properties that can be varied locally by light permanently or in a reversible way, is in fact an area of intense research activity. In particular, permanent recording, high storage density, high sensitivity (that is low writing intensity) and fast response are some of the main goals to be achieved. While photorefractive inorganic and organic materials have been considered for a long time the most promising for optical storage[1,2], dye-doped liquid crystals have recently shown to be very interesting for these kind of applications[3,4]. In the recent few years, it has been claimed in fact that the illumination of an azo-containing liquid crystal bulk with linearly polarised light induces a stable alignment of the director on an isotropic surface[5]. Light absorption into the liquid crystal bulk leads to a permanent reorientation of the director on the surface toward the incident polarisation direction. The effect seems to be caused by the adsorption on the isotropic surface of the photo-exited dye molecules, which gives rise to a stable easy axis on the surface[6]. Recent experiments have shown that the light-induced director reorientation can be successfully exploited to write intensity[3] and polarisation[4] gratings with high sensitivity and spatial resolution.

We have recently shown[7] that a single laser pulse (4 ns) is able to induce a stable anchoring in dye-doped liquid crystals. Here we present data on the kinetic of the effect pointing out the strong dependence of the anchoring stability on the exciting polarisation angle.

EXPERIMENTAL

Experiments have been carried out on sandwich combined cells filled with a mixture of the nematic liquid crystal 5CB and the azo-dye Methyl Red (MR). The Methyl Red weight concentration is about 1%. Combined cells substrates are coated with different layers. One, the isotropic surface, is covered with an isotropic layer of Poly(vinyl)-cinnamate fluoride (PVCN-F) deposited by spin coating on the ITO glass, while the other is treated in order to obtain strong planar anchoring of the liquid crystal director. Cells are filled under the influence of a strong magnetic field (0,8 T) with the axis parallel to the rubbing direction. In this way, the planar surface determines the initial planar alignment of the cell. Cells thickness varies from 10 to 50 μm.

Pump and probe measurements have been performed in order to study the response of our samples to short pulsed excitation. A linearly polarised beam from the second harmonic of a Nd:YAG laser (λ = 532 nm) impinges on the isotropic surface of the cell at normal incidence. The optical energy density on the sample is in the range (10^{-3} + $5x10^{-1}$) J/cm^2. The irradiated region is probed by a linearly polarised He-Ne laser that passes through the sample and then through an analyser. The probe beam polarisation is parallel to the initial planar orientation of the cell, so that the light intensity behind the analyser is determined by the Malus law:

$$I^{out}_{He\text{-}Ne} \approx I^{in}_{He\text{-}Ne} \, sen^2\theta_{is}$$

being θ_{is} the angle between the liquid crystal director on the isotropic surface (n_{is}) and the one on the treated surface (n_{plan}). Before irradiating the cell, n_{is} is parallel to n_{plan} and $I^{out}_{He\text{-}Ne} = 0$. The appereance of probe light behind the analyser indicates that θ_{is} is changing and since n_{plan} is fixed by the strong planar anchoring, variations of θ_{is} are to be ascribed to director reorientation on the isotropic surface. Therefore analysis of $I^{out}_{He\text{-}Ne}$ as a function of time allows to study the kinetic of director reorientation on this surface.

RESULTS AND DISCUSSION

Cell irradiation with a single pulse of the pump beam results in the appearance of light behind the analyser, that is in director reorientation on the isotropic surface.

The first obvious condition necessary to get director reorientation is a pump beam polarised along a direction different from the initial planar orientation. Defining α as the angle between these directions we have observed that the value of

α affects the kinetic of the process. As expected from data obtained under c.w. excitation[3] the best conditions occur for $\alpha = 45°$. Figure 1 shows the detected signal vs time after irradiation (t = 0 s corresponds to the 4 ns excitation spike). It is clearly observed a rise of the signal to a maximum in few minutes and a subsequent stabilisation after about 30 minutes. This stable value keeps constant for a long time (the correspondent spot on the sample has been observed for months).

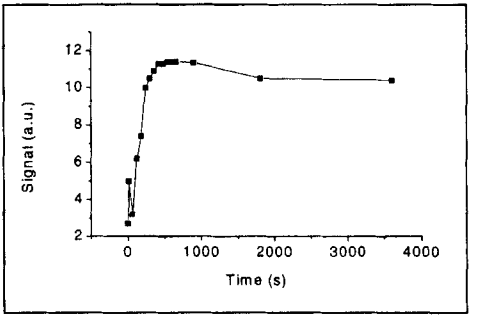

FIGURE 1 Probe signal versus time for $\alpha = 45°$.

The detected signal can be set to zero by rotating the analyser and rotation angle measures the rotation of the probe-beam polarisation through the cell. Since the rotation of the director occurs over a distance that is much greater than the light wavelength, the probe light polarisation follows the director orientation adiabatically so that the measured rotation coincides with θ_{is}. Referring to fig. 1, the measured reorientation angle is $\theta_{is} = 34°$.

By increasing α the effect becomes less and less effective with a permanent reorientation corresponding to a lower value of θ_{is}.

In fig. 2 we show data obtained with $\alpha = 90°$. In this case the induced surface reorientation is not stable and after few hours it cannot be observed any more (i.e. the initial planar orientation is restored). This behaviour can be expected since it was demonstrated that when $\alpha = 90°$ it is necessary to overcome a threshold intensity in order to get a stable anchoring. The energy density used in the present experiments was thus below the threshold value, which was confirmed by the observation of a stable reorientation in the same conditions using additional excitation pulses with a delay of few seconds. The existence of a threshold intensity in the case $\alpha = 90°$ can be easily accounted for taking into account that the absorption of the incident light by the dye molecules strongly depends on the incident polarisation direction. In particular, if the incident light is polarised perpendicularly to the dye molecules dipole axis, the absorption coefficient is minimum. In our experimental conditions it is likely to suppose that, before irradiation, the spatial distribution of the dye molecules dipole axis is on the average parallel to the planar orientation of the cell.

In this case, when $\alpha = 90°$, the number of photo-excited dye molecules results to be minimum and the required energy density to achieve permanent reorientation is thus much higher than in the other cases.

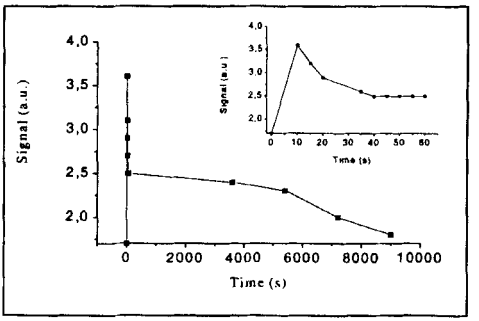

FIGURE 2 Probe signal detected behind the analyser versus time for $\alpha = 90°$. The inset shows the initial peak in details.

Preliminary observations suggest that the initial peak observed in fig. 2 with a decay time of the order of 10÷20 s, can be due to direct optical reorientation of the liquid crystal in the azimuthal plane. Further investigations are needed in order to clarify this point.

The stable anchoring induced by choosing $\alpha = 45°$ allows to investigate the capability of the described effect for optical storage applications.

Polarised optical microscope observations of the irradiated regions in these conditions, show the formation of a twisted structure, which is generated because the liquid crystal director is fixed on the treated surface and follows the incident polarisation direction on the isotropic one. The possibility of using a single 4 ns pulse for recording binary images is shown in fig. 3. The pump beam passes through a mask reproducing the "clubs" shape before impinging the cell. The formation of the twisted structure in correspondence of the irradiated area is evident by changing the mutual orientation of the two polarisers (they are parallel in fig. 3a and crossed in fig. 3b).

The described reorientation effect has also been exploited to write holographic gratings in the 5CB+MR combined cells. The experimental set up was a standard Mach-Zender geometry for holographic recording. One of the obtained gratings is shown in fig. 4. Gratings are stable (they didn't change for more than one year) and the total energy density required for permanent recording was just 7×10^{-3} J/cm^2. The origin of the grating is the light-induced molecular director reorientation, which occurs only in correspondence of the maxima of the incident interference pattern.

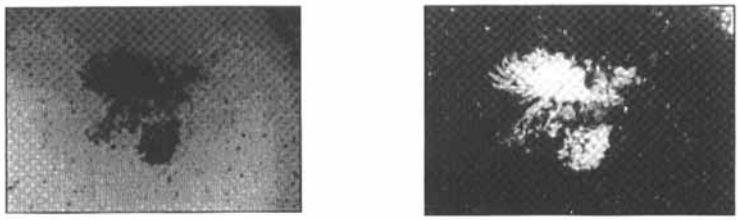

FIGURE 3 Optical microscope view of a binary image written with a single 4 ns pulse. Polarisers are parallel (a) and crossed (b). Cell thickness is 50 μm.

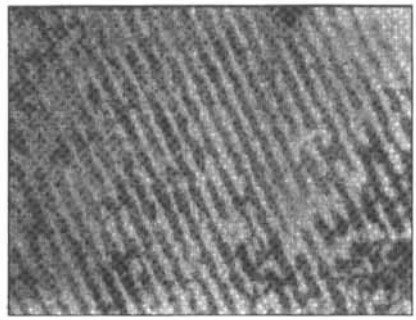

FIGURE 4 Permanent grating written with a single pump pulse observed under a polarised optical microscope. The grating period is $\Lambda = 10$ μm.

Gratings characteristics were probed even in this case by a low intensity He-Ne laser. In particular, the diffraction efficiency η was measured as a function of the grating period Λ for both the polarisations of the probe beam (parallel, η_p, and perpendicular, η_s, to the polarisation of the writing beams). η is defined as $\eta = I_1/I_0$, where I_1 is the intensity of the first-order diffracted beam and I_0 is the intensity of the incident probe beam. η_p versus Λ is reported in fig. 5. As can be seen the diffraction efficiency of our gratings is 20% for a grating period of 8 μm, but an efficiency of 0.5% is still detectable for a period of 1.8 μm, which corresponds to more than 500 lines/mm.

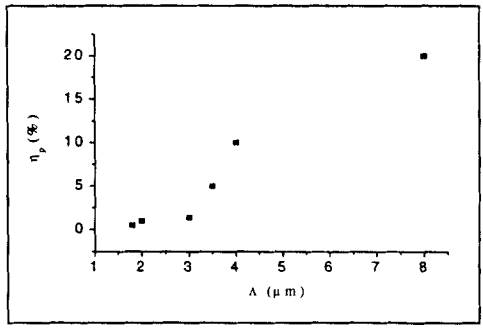

FIGURE 5 Diffraction efficiency vs grating period. The probe beam polarisation is parallel to the polarisation of the writing beams.

CONCLUSIONS

The response of dye-doped liquid crystals to short pulsed irradiation and the dependence of the light-induced reorientation effect on the incident polarisation direction have been studied. Moreover, the effect of permanent director reorientation, obtained under specific conditions, has been exploited to write binary images and high resolution holographic gratings in our cells.

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