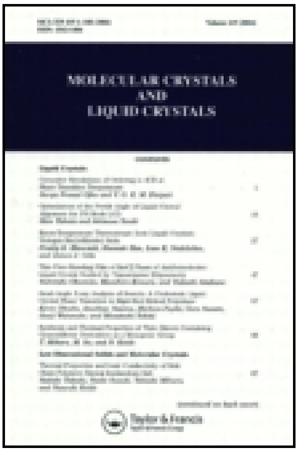

Molecular Crystals and Liquid Crystals
Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl20

Resonant Optical Torque in Dichroic Azo Dye Doped Nematic Liquid Crystals

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Version of record first published: 18 Oct 2010

To cite this article: T. V. Galstian, E. Brasselet & D. Dumont (2002): Resonant Optical Torque in Dichroic Azo Dye Doped Nematic Liquid Crystals, Molecular Crystals and Liquid Crystals, 375:1, 593-606

To link to this article: http://dx.doi.org/10.1080/10587250210595

Mol. Cryst. Liq. Cryst., Vol. 375, pp. 593-606
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1058-725X/02 $12.00 + .00

Resonant Optical Torque in Dichroic Azo Dye Doped Nematic Liquid Crystals

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The optical torque is experimentally studied in Disperse Red dichroic dye doped E7 nematic liquid crystal. Creation of a transitory out-of-plane (3D) anisotropy axis is observed in a geometry were in-plane reorientation is observed in pure nematics. The non-monotone behavior of dye absorption during the excitation and relaxation is correlated with the 3D reorientation. This shows that the photo-isomerization of the dye via transient excited states (particularly of *Cis* form) should be the mechanism, which gives a "pulse" for the creation of the 3D anisotropy axis.

Keywords resonant optical torque, nematic liquid crystal, dichroic azo dye.

INTRODUCTION

Dye doped nematic liquid crystal (NLC) compounds have been studied during last few decades [1,2]. The contribution of the dye in the optical behavior of these guest-host compounds was rather quantitative,

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changing mainly the absorption value of the system, but keeping the character of light interaction with NLC almost the same. It was the same also in the particular case of nonlinear light-matter interactions, where the dye was often used as a dichroic absorbing agent, with or without a heat release [3,4].

The situation was changed when Janossy and collaborators have demonstrated experimentally a significant amplification of the optical dielectric torque (ODT) by adding a small quantity of anthraquinone dye in the NLC host [5]. The threshold of light induced Freederisckz transition (LIFT, [6]) was decreased almost by two orders of magnitude [5]. That was rather surprising and has shown a qualitatively different behavior, in spite the fact that at the first view, the character of the LIFT was not significantly changed.

First attempts were made to describe this phenomenon using a coefficient of amplification [7a] of the "traditional" ODT. It was discovered that other, e.g. azoic [8-11] dyes also, may significantly change the light-NLC interaction. It was shown by number of groups that the phenomenon is very complex, depending dramatically upon various experimental parameters as for example, the angle of incidence [9], the frequency of excitation light [11,12], etc. A model that takes into account the opposite contributions of the *Trans* and *Cis* isomers to the optical torque was proposed to explain the change of its sign [7b]. Among other interesting results, was the fact that, in some cases, the molecular reorientation was opposed to the direction expected for the traditional ODT in pure NLC. That is, instead of being attracted by the electric field of light, the director of NLC was reoriented towards the wavevector of the excitation beam [8,9]. Our group has observed another intriguing phenomenon of three-dimensional (3D) reorientation in an experimental geometry, where an in-plane rotation of the director is expected if the traditional ODT is the driving force for reorientation [11].

In what will follow, we shall try to describe and analyze these phenomena using our recent experimental results obtained with Disperse Red (DR1) dye doped E7 NLC. The choice of these components was based on the fact that the E7 matrix is more stable having much higher clearing temperature compared to 5CB (used in Ref. [11]). Also the dye DR1 undergoes photo-isomerization reaction as the Bisazobenzene dye (used in Ref. [11]), but seams to be a simpler system compared to Bisazobenzene, which contains two Nitrogen double bonds and three Phenyl groups.

MATERIAL SYSTEM AND EXPERIMENTAL CONDITIONS

Our experiments were carried out at room temperature (22°C) without thermo-isolation of cells. Both homeotropically and planarly oriented cells were used. The homeotropic cells were fabricated by means of CTAB surfactant. A standard rubbing technique was used to get planarly oriented samples (with PMMA coated glass substrates). The thickness of cells was controlled by a Mylar spacer of 37.5 μm thick. E7 nematic was volume doped by DR1 dye (typically 0.4 wt% concentration). We checked that the azo dye is well mixed with the nematic host by measuring the transmission spectra of planar oriented samples at normal incidence. The analyzing beam was polarized parallel (to measure the optical density $D_{//}$) and then perpendicular (to measure the $D_\perp$) with respect to the director of the NLC (figure 1). This spectra exhibit strongly dichroic absorption near to 0.5 μm (figure 1). Such an absorption is known to be the combination of absorption of *Trans* and *Cis* molecular forms of azo dye used (see next section). The typical maximum absorption coefficient is at the order of $\sigma_\perp \approx 150\ \mathrm{cm}^{-1}$.

PHOTO-EXCITATION DYNAMICS OF *TRANS* AND *CIS* ISOMERS

It is well known that azoic dyes undergo *Trans-Cis* and *Cis-Trans* isomerization reaction absorbing light in the visible and near UV spectral band [13]. We shall come back to the details of this reaction later. Note for the moment that usually the used azo dye is known to

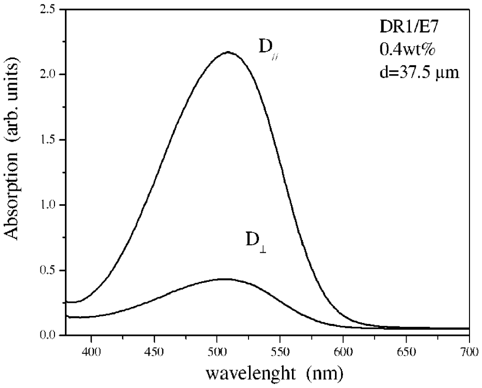

FIGURE 1 Absorption spectra of the planarly oriented E7 NLC that was doped by dichroic azo dye DR1.

have absorption of the *Cis* molecular form in the blue band and absorption of the *Trans* form in the green band [13,14]. In spite of that, enhanced absorption of the *Cis* molecular form in the red spectral band has been detected and studied by several groups [15,16]. We shall use this spectral information to monitor the photo-excitation dynamics at two probe wavelengths.

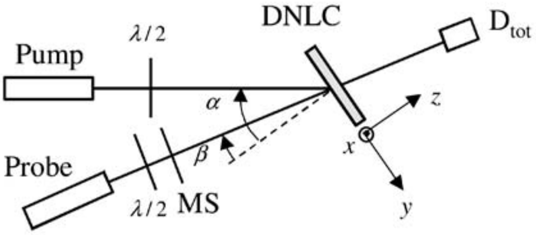

FIGURE 2 Cross modulation set-up. DNLC: DR1-doped nematic liquid crystal (0.4% wt), $\lambda/2$: half-wave plate, MS: mechanical shutter, D_{tot}: detector for the total intensity of the probe beam. Dashed line is the normal of the cell (*z*-axis), α and β are the angle of incidence of the pump beam and the probe beam, respectively.

Thus, we carried out experiments (figure 2) where a DR1-doped NLC E7 cell was pumped by a Verdi laser operating at 532 nm. The transmission of the probe beam at 514.5 nm (Ar^+ laser) or at 632.8 nm (He-Ne laser) was monitored. Note that the probe wavelengths are chosen to monitor the absorption of the non-excited *Trans* molecular form (at 514.5 nm) and the absorption of the *Cis* form (at 632.8 nm). We used a square-like excitation by means of the mechanical shutter MS (figure 2). Thus, in the figures 3a, three time domains can be distinguished in the transmitted signal $I(t)$: before the pump was switched on ($t < 2s$), during the irradiation (approximately $2s < t < 11s$) and after the pump was switched off ($t > 11s$).

In these cross-modulation experiments we used co-propagating pump and probe beams ($\alpha = \beta$, see figure 2). In this case, the probe detection is achieved using an interferential filter when $\lambda_{probe} = 632.8$ nm. However, the spectral separation is difficult when $\lambda_{probe} = 514.5$ nm, then the lens L (placed between the cell and the detector) is slightly misaligned in order to separate the beams in the focal plane and a sharp metallic sheet is used to block the pump beam without affecting the probe beam. The excitation beam diameter at the plane of the NLC film is ≈1.5 mm, while the probe diameter is ≈0.5 mm. All incident polarizations (pump and probe) are linear and their direction is adjusted by half-wave plates $\lambda/2$ (figure 2). We shall call $I_s(t)$ the signal obtained from D_{tot} when the polarization of the incident probe beam is perpendicular to the plane of incidence $\{y, z\}$ (figure 2) and $I_p(t)$ when the polarization of the probe beam is parallel to the plane of incidence.

We have observed director reorientation in a homeotropically aligned cell when $\alpha = \beta \neq 0$ (see later, figure 4). That is why, we decided first to obtain some "reference" spectral information in the absence of director reorientation. This was done when $\alpha = \beta = 0$, using a planarly oriented cell. We shall further report separately the results for the *Trans* and *Cis* isomer monitoring.

Dynamics of the Trans isomer excitation (probe beam at 514.5 nm)

In a first experiment, we used a planarly aligned cell fixed in a holder so that the rubbing direction is parallel to the x-axis (vertical). All beams impinge on the cell at normal incidence ($\alpha = \beta = 0°$) and the incident pump electric field is chosen to be parallel to the rubbing direction (in other words, it is parallel to the director in the absence of reorientation). The experiment was performed with $\lambda_{probe} = 514.5$ nm. The figure 3a shows $I_s(t)$ for $P_{pump} = 70$ mW (curve 1) and $P_{pump} = 170$ mW (curve 2). In this case, the photo-excitation leads to an abrupt increase in the probe transmission that stabilizes within 0.1 sec, for all pump power values without oscillations. Indeed, the excitation of *Trans* and *Cis* molecules should tend to decrease the probe absorption at the very beginning of the illumination. This decrease saturates in a time of the order of the longest lifetime of those excited molecules. The corresponding probe transmission saturation value depends on the pump power, leading to higher values for higher pump power while the initial value of transmission depends upon the probe polarization. As expected, the transmitted signal intensity is higher when the electric field of the probe beam is perpendicular to the rubbing direction, and thus, to the longer molecular axis of the dye before illumination. The relaxation is also abrupt and monotonic with a duration less than 0.1s.

The same experiments, but performed for $\alpha = \beta \neq 0$ in a *homeotropically* aligned cell, have shown that the green probe transmission for various pump power values behaves similarly to the described above experiment. Namely, there is always an abrupt increase at the excitation switch-on, then the steady state is reached and an abrupt decrease occurs at the excitation switch off.

In all probe polarization cases, the transmission is increased upon excitation, showing the absence of reorientation of the *Trans* form in the experimental range of intensities where 3D reorientation is observed (see later). This may be due to the strong angular mobility of the dye as it is often the case in liquids. The strong anisotropy of the angular potential created by the NLC would enhance this “angular relaxation”.

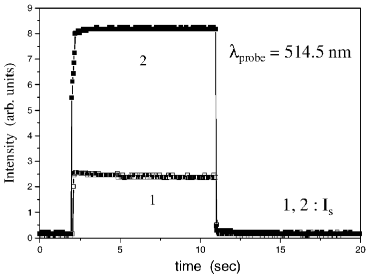

FIGURE 3(a) s-polarized probe transmission through the planarly aligned DR1-doped E7 cell. The wavelength of the probe is 514.5 nm. The pump polarization is parallel to the rubbing direction. P_{pump} = 70 mW (curve 1) and P_{pump} = 170mW (curve 2).

Dynamics of the Cis isomer excitation (probe beam at 632.8 nm)

The figure 3b shows behaviors of $I_s(t)$ (curves 1 and 3) and $I_p(t)$ (curves 2 and 4) for different pump powers when λ_{probe} = 632.8 nm (the used cell is still planarly oriented). At low excitation power (P_{pump} = 70 mW, curves 1 and 2), both transmission of s- and p- polarized probes are monotonically decreased to a stationary state when the excitation beam is switched on. When the excitation is switched off, the system relaxes monotonically towards the unperturbed state in few seconds. In this regime of excitation both probe polarizations detect similar dynamics, however the variation of I_s is much (3-4 times) larger than that of I_p.

This indicates a significant increase of the anisotropy of the red absorption (and consequently of the *Cis* molecular population), which is parallel with the rubbing direction (s-polarized probe). This kind of excitation (via, e.g., reorientation or angularly selective excitation) is possible also in polymers when anisotropic species, such as azo dyes, are present in the media [14-17].

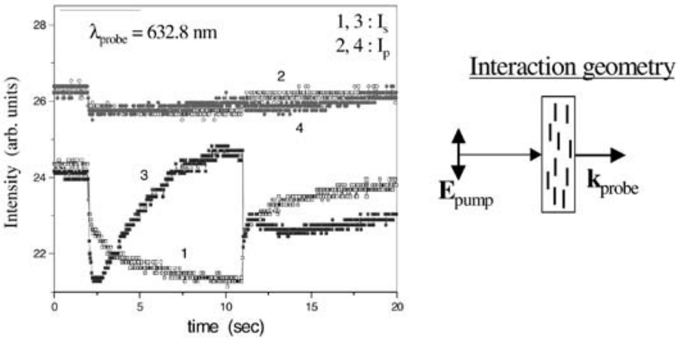

FIGURE 3(b) Total intensity transmitted through the planarly aligned DR1-doped E7 cell by s- (curves 1&3) and p- (curves 2&4) polarized probe beams at 632.8 nm when $\alpha = \beta = 0°$. The photo-excitation takes place in the time interval $2\text{s} < t < 11\text{s}$. The pump beam (at 532 nm) polarization is parallel to the rubbing direction (the x-axis). $P_{pump} = 70$ mW for the curves 1 and 2, and $P_{pump} = 170$ mW for the curves 3 and 4.

At higher excitation regime ($P_{pump} = 170$ mW, curves 3 and 4), the behavior of $I_p(t)$ is almost the same while the behavior of $I_s(t)$ is qualitatively different. Indeed, $I_s(t)$ exhibits clear non-monotonic behavior, namely at the first stage of excitation there is a rapid decrease followed by a slower monotonic increase up to a value that is even higher than the initial one. It is important to note that the minimum value of $I_s(t)$ is the same (nearly 10% of the initial value) from different pump powers but it is reached more rapidly for higher power excitation. Then, in the second stage of excitation, the anisotropy of the red absorption (*Cis* population) decreases, but there is no a correlated increase of the perpendicular polarized probe absorption. Thus, the angular reorientation could not be responsible for such a decrease, which very likely should be related to the angularly selective photo-excitation (hole burning) mechanism.

The relaxation to the unperturbed state is also non-monotonic, showing an abrupt decrease, at less than 0.1s, followed by a non-monotonic relaxation. The variations of $I_p(t)$ show also some non-monotone signature while remaining as small as in the case of lower excitation power.

We believe that the first stage of the relaxation should be related to the rapid relaxation from transient excited molecular states to the *Cis* form that increases the absorption in the red-band. Moreover, the fact that only $I_s(t)$ exhibits this (abrupt) behavior suggests that this relaxation goes through strongly anisotropic states. The last stage of relaxation may be explained as back *Cis*→*Trans* relaxation.

Note that during the above mentioned experiments (where the exposition time was ≈ 10s) no significant photo-induced scattering, self-focusing or defocusing (rings) were observed. However, these phenomena were observed for much longer exposition (approximately 40s and more) indicating the presence of some significant accumulative effects.

Similar red-probe experiments were carried out using a *homeotropically* aligned cell, the only difference being that the co propagating pump and probe beams hereafter make 30° of angle of incidence ($\alpha = \beta = 30^o$). The figure 3c corresponds to the case when the pump is p-polarized (as in our polarimetric studies, see later). The light-induced behavior of the homeotropically oriented cell appears to be quite different compared to the case of the planarly aligned cell at normal incidence (compare figure 3b with figure 3c). Indeed, at low power ($P_{pump} = 70$ mW, curves 1 and 2) both $I_s(t)$ and $I_p(t)$ have non-monotonic behavior with two different time scales. Namely, there is a slight decrease of transmission in the first stage of the excitation followed by an increase up to a steady state value until the pump is switched off. At higher excitation power ($P_{pump} = 210$ mW, curves 3 & 4), these changes are significantly amplified. Indeed $I_s(t)$ and $I_p(t)$ seems to reproduce the excitation scenario corresponding to the low pump power case, but more rapidly and with more pronounced steady state value. After an initial decrease, a significant monotonic increase of both $I_s(t)$ and $I_p(t)$ takes place until the pump is switched off. This decrease of the red absorption takes place without significant anisotropy (compared to the figure 3(b).

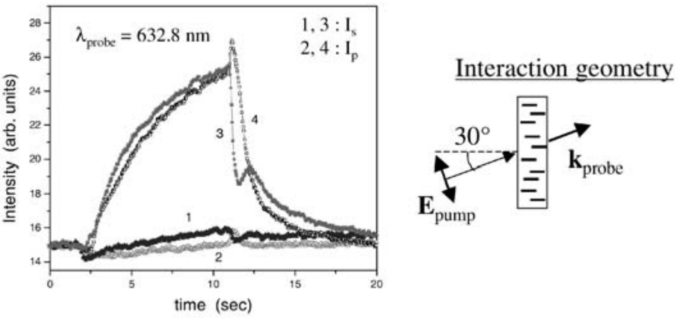

FIGURE 3(c) Probe (at 623.8 nm) transmission through the homeotropic aligned DR1-doped E7 cell. The pump beam has p-polarization. P_{pump} = 70 mW for the curves 1 & 2, and P_{pump} = 210mW for the curves 3&4. Curves 1 & 3 correspond to the s-polarized probe and curve 2&4 to the p-polarized probe.

The relaxation for both low and high power excitation regimes is strongly anisotropic with $I_s(t)$ and $I_p(t)$ having opposed time variations. Moreover, at higher excitation regime, the non-monotonic relaxation may be divided into several sub-regimes. First, there is an anisotropic process where the variation of $I_s(t)$ and $I_p(t)$ are rather the same in absolute value but with opposite signs. Second, both signals decrease, third opposite variation occurs once again. Finally, all signals relax monotonically to their unperturbed value.

Apparently, the *Cis* isomer exhibit a complex excitation picture including angular hole burning and probably some reorientation process, which is difficult to model at this stage of our study. The complexity of the situation is in part related to the absence of the values of molecular susceptibilities (angular dependences) for different excited states. However, the dynamics of *Cis* absorption changes appears rather well correlated to our polarimetric studies, which helps, at least, to relate the observed 3D director reorientations (see later) to the transitions to the *Cis* molecular form.

POLARIMETRIC STUDIES

The reorientation of the director in a *homeotropically*-aligned cell under the resonant photoexcitation of dye molecules was analyzed using a polarimetric set-up. A Wollaston prism was added at the output of the cell (as in Ref.[18]) using the same experimental set-up as the one of figure 2. We have performed similar to the previous section experiments with co-propagating excitation and probe beams ($\alpha = \beta = 30°$). However, in this experiment, the probe beam ($\lambda_{probe} = 632.8$ nm) is linearly polarized at 45° to the horizontal plane. Two detectors, placed behind the Wollaston prism, detect simultaneously the intensity of the vertical (I_s) and horizontal (I_p) components of the emerging probe beam. The polarization of the pump beam is in the horizontal plane. Let us emphasize that the unperturbed director and one of axis of the Wollaston prism are in the same horizontal plane. That shows unambiguously that the energy *exchange* between probe's vertical and horizontal components will be possible *only* if an *out* of the horizontal plane optical axis is created. Corresponding results are presented in the figure 4.

In the absence of excitation both signals have approximately the same intensity (figure 4), since the output probe has an elliptical polarization with its main axis at 45° from the plane of incidence, due to the linear birefringence of the guest-host compound.

When the excitation beam is switched-on and off, we observe an almost perfect symmetry of opposed variations of both polarization components ($P_{pump} = 70$ mW, curves 1 and 2). Thus, there are initially rapid and opposed variations of the intensities of these components, then, the system reaches slowly a steady state value. The symmetry of these changes clearly indicates the energy exchange between I_s and I_p and consequently, the creation of an out-of plane (3D) optical axis. Our cross-modulation experiments (figure 3c) have shown no dye reorientation. Thus, these opposed and symmetric changes indicate a director's 3D trajectory.

Note also, that at higher excitation power ($P_{pump} = 210$ mW, curves 3 and 4) the symmetry of these changes is slightly broken, which could be related to the photo-induced absorption anisotropy in the red-band (figure 3c).

Surprisingly, when the excitation is switched-off, the transient relaxation has again a 3D character. Moreover, this relaxation demonstrates a rotation of the output probe polarization ellipse in the opposite direction compared with the excitation stage. Additionally, it is interesting to note that the transient 3D trajectories are not the same when the excitation beam is switched on and off.

Some preliminary experiments were conducted also with s-polarized pump beam. Surprisingly, there are no significant differences between the cases of p-and s-polarized pump beams in spite the fact of different absorptions for those pumps. This fact is in opposition also with the traditional ODT, where the reorientation is expected to be more significant for the p-polarized pump. These phenomena are under study and will be reported soon.

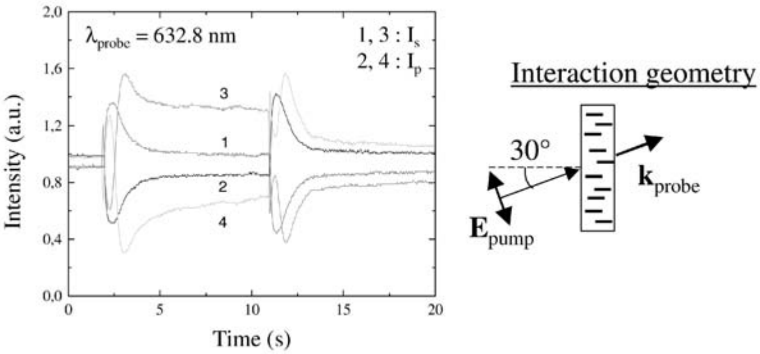

FIGURE 4 Intensity of the vertical (s) and horizontal (p) components of the probe beam polarization, detected behind the Wollaston prism, when $\alpha = \beta = 30°$. The DR1-doped E7 cell is homeotropically aligned and the incident He-Ne probe beam is linearly polarized at 45°. The photo-excitation takes place in the time interval $2s < t < 11s$. The curves 1&3 correspond to I_s and the curves 2&4 correspond to I_p. $P_{pump} = 70$ mW for the curves 1 and 2 and $P_{pump} = 210$ mW for the curves 3 and 4. The pump beam is p-polarized.

DISCUSSION

We believe that the absence of correlation between cross modulation experiments for the probe at 514.5 nm (which is particularly sensitive to the *Trans* form) and polarimetric data shows that the only excitation of the *Trans* molecular form is not sufficient for the generation of the 3D optical axis. In addition, according to the figure 3a, there is no significant reorientation of the *Trans* form.

In contrast, the intensity variations (in polarimetric studies, figure 4) are well correlated with cross modulation experiments at red wavelength probe (632.8 nm), which is particularly sensitive to the *Cis* molecular form. Thus, the non-monotone behavior of the corresponding absorption during the excitation and relaxation (figures 3b-c) shows that the *Trans-Cis* photo-isomerisation via transitory states should be the mechanism, which gives the anisotropic "pulse" for the creation of the 3D anisotropy axis (figure 4). Indeed, the time scale of observed quick modifications suggests the possible involvement of the dye triplet states (particularly of the *Cis* form) in this process.

Particularly interesting is the transitory opposed (bipolar) change in the red probe absorption during the relaxation (figure 3c), which should be at the origin of the "strange" 3D orientational relaxation via a trajectory different than the excitation one.

At a phenomenological level, we can imagine the existence of two mechanisms, which generate opposed torques trying to bring the director out of plane, but with different characteristic times. Thus, the faster mechanism *F* develops first as soon as the excitation is switched on, forcing the optical axis to be reoriented out of the horizontal plane. Then the second (slower) mechanism *S* develops trying to compensate the first one and bring the optical axis back to the horizontal plane. The equilibrium is established at a certain out-of-plane orientation of the axis, which is closer to the horizontal plane compared to the transitory regime. This equilibrium may be broken down when the excitation is switched off. Namely, the faster mechanism *F* relaxes quickly and then the slower mechanism *S* attracts the axis towards the opposed (compared to the excitation regime) direction. This continues up to the moment when the relaxation (e.g., elastic) forces become stronger than the second mechanism (due to the relaxation of the last one). The system then monotonically relaxes to its initial state.

We believe that the break-down of the symmetry, which defines the direction of the out-of-plane reorientation, should be related to a possible very small pretilt angle defined by the surface alignment imperfections that are difficult to detect in a linear polarimetric experiment. In some (relatively rare) cases, we indeed have observed an initial out-of-plane reorientation in the opposed direction.

CONCLUSION

In conclusion, an out of plan transitory director reorientation is identified in DR1 azo dye doped E7 NLC in a geometry where an in plane reorientation is observed in the pure NLC. We believe that this reorientation is related to the photo-induced transitions towards (during excitation) and from (during relaxation) the excited *Cis* molecular state of the dye.

We have to admit also that our suggestion that the DR1 doped E7 would be a simpler guest-host system than the Bisazobenzene doped 5CB was wrong. Indeed, in both cases we observe almost the same behavior [18]. This takes place in spite the fact that there is an additional Nitrogen double bond and phenyl group in the Bisazobenzene dye. This could means that we deal with a "single" photo-isomerization in those two cases. The behavior of the NLC matrix also seams to remain the same in spite the fact that the clearing temperature of E7 is almost twice higher compared to the 5CB. The later fact indicates that the interaction of the excited dye is not a near-to-phase transition effect.

Finally, we would like to emphasize that the discussed out-of plane reorientation has a transient character (both growth and decrease) in contrast to the case of strongly focused light excitation in pure NLC, where a very small, slowly growing and stabilizing (achieving a steady-state) 3D reorientation may be obtained in some particular cases [19].

Acknowledgments

We would like to thank National Sciences and Engineering Research Council of Canada (NSERC), le Fond pour la Formation des Chercheurs et l'Aide à la Recherche (FCAR) for their financial support to this work.

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