

GIANT OPTICAL RESPONSE OF AZO-DYE DOPED LIQUID CRYSTALS: SURFACE AND BULK EFFECTS

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We report an experimental study on the origin of the giant optical response in azo-dye doped nematic liquid crystals. We show that a very large response is obtained for extremely low input light intensity, of the order of a few tens of $\mu\text{W}/\text{cm}^2$, and that it is mediated by the action of the photo-induced voltage on the specific surface coupling agent which is an ionic surfactant. We report a characterization of the nonlinear optical response as a function of different azo-dye concentrations in the liquid crystal host. Then, by performing circular polarization experiments, we show that, during the reorientation, the nematic director follows a three-dimensional trajectory composed by in and out-of-plane motions with respect to the incidence plane of the light, indicating that the bulk contribution is dominated by the photoisomerization of azo-dye molecules

also by other recent papers [9–12] on the effect of ion-controlled anchoring transitions in NLC. In another recent paper [13], the effect of photo-alignment which is related to the presence of dye-doped anchoring layers has also been demonstrated.

In general, surface and bulk contributions may be simultaneously present, and it is the interplay between these two effects that finally leads to a stable grating formation. Bulk reorientation has been reported as a consequence of the *trans-cis* photoisomerization of azo-dye molecules under irradiation in the absorption band of dopants [14, 15]. Surface and bulk driven reorientations may also be combined for technological applications, such as the recording of high resolution permanent gratings [16].

Recently, we have shown that the large optical nonlinearity of dye-doped NLC is indeed mediated by a specific surface alignment agent [17, 18]. By testing a large part of the known alignment agents that give homeotropic orientation, we have obtained an extremely high sensitivity only for the cells treated with an ionic surfactant. However, the origin of such an extremely high sensitivity is still a matter of debate and many questions remain open, such as the ones related to the contributions of the surface and bulk effects to the whole reorientation process. Indeed, once the reorientation has started at the surface as a consequence of the light-induced weakening of the anchoring strength, the director motion propagates in the bulk of a cell, where it couples with other mechanisms which are usually resumed under a mean-field description in terms of guest-host interaction [19]. In the details, this interaction strongly depends on a particular type of liquid crystals, especially on their polar or non-polar nature, on the molecular conformation of azo-dye dopants and on the incident light polarization, wavelength, and propagation direction.

As the first step in the characterization of the bulk response, we have performed several experiments at

Introduction

Light-induced reorientational phenomena in nematic liquid crystals (NLC) have been the subject of intense research interest during the last 20 years [1]. Giant optical nonlinearities [2] have been observed for a suitable choice of the incident light polarization and the nematic liquid crystal director \vec{n} . More recently, Janossy [3] has shown that NLC doped with dye may give even a stronger nonlinear response. In addition to their large optical reorientational nonlinearities, several research groups have found that they exhibit different properties, like surface mediated reorientation [4] connected with the *trans-cis* photoisomerization of azo-dye molecules [5] and photorefractive effects due to space charge modulation [6]. Further studies by Khoo [7] showed the extremely large sensitivity of a particular mixture: a small amount ($< 1\%$) of Methyl Red (MR) in 5CB. This was interpreted as a large photorefractive effect arising from an optically induced space charge field in combination with a bias dc field. Another possible interpretation is the one given by Simoni and coworkers [8], who have shown that the director reorientation originates from light-induced modifications of the anchoring conditions. This conjecture is supported

different concentrations of the azo-dye in the liquid crystal host. The results show that, for increasing dye concentration, a lower input intensity is needed in order to observe a nonlinear optical response. However, as the concentration increases, memory effects become more important and eventually the gratings become permanent at moderate values of the input pump intensity.

Then, we have tested our homeotropic cells for circular polarization of the pumps and we have shown that the surface and bulk contributions combine in such a way that the resulting motion of the director is a three-dimensional trajectory leaving the plane of incidence of the incoming laser beams [20]. This motion starts with a fast in-plane reorientation, due to the surface effect. Then, a much slower contribution brings the director out-of-plane, leading to a continuous increase of the light absorption because of the dye dichroism. The subsequent photoisomerization of dye molecules induces, in turn, a further rotation of the director. Finally, in the stationary state, the director lies in a plane almost parallel to the bounding plates, that is, the reoriented cell is very similar to a planar one. Once written, the grating is self-sustained, that is, it is still present and stable even when we switch off one of the two pump beams.

In the first part of the article we present the methods for the preparation of homeotropic cells. Then, we describe the experimental setup and we present the results obtained in the diffraction experiments for the linear polarization of pumps. We report the behavior of the nonlinear coefficient for different concentrations of the dye and we present circular polarization experiments. Finally, we discuss the experimental results and we outline the memory effects related to the photoisomerization of the azo-dye dopants.

Preparation of Liquid Crystal Cells

All our cells are made by ITO coated glass plates (Asahi Glass). The size of each glass plate is 20×30 mm. The plates are cleaned in an ultrasound bath at 25°C for 15–20 min with a Micro 90 cleaning solution (Bioblock Scientific). After cleaning, the slides are rinsed 5 times, 5 min each time, in a highly purified water ($R \simeq 18\text{ M}\Omega$) and then dried with filtered compressed air. The clean glass plates are then treated for deposition of the alignment agent.

Homeotropic alignment of nematic liquid crystals can be obtained by different methods and by the use of different anchoring agents [21]. We have tested most of the standard techniques such as the spin-coating and

deposition of a Langmuir film. All the technological procedures were performed in a clean bench. Spin-coating of the glass plates was used to treat the surfaces with Lecithin or with a specific poly-imide, provided by Nissan Chemicals. Lecithin was just spin-coated over the plates whereas Nissan Chemicals poly-imide was prepared by two-step curing, at 80°C during 30 min after the spin-coating, then at 180°C during about 60 min.

We have also prepared cells by treating the surfaces with chlorosilanes which are well known to provide a good homeotropic alignment. Deposition of N,N-dimethyl-N-octadecyl-3-amino-propyl-trimethoxysilyl chloride (DMOAP) was done according to the protocol described by Khan [22]. Clean glass plates were dipped into a 0.1% wt DMOAP water solution and kept about 20–30 min. Then, the plates were rinsed, dried and cured at 110°C during 1 h. We have also tested commercial cells from Linkam Scientific Instruments, treated to provide homeotropic alignment.

Another well-known method to obtain homeotropic alignment is to depose a surfactant monolayer over the glass substrates [21, 23]. We have followed this procedure with two different surfactants, hexadecyl-trimethyl-ammonium-bromide (HTAB) and hexadecyl-trimethyl-ammonium-chloride (HTAC). Both HTAC and HTAB belong to the group of ion pair reagents. The deposition of the HTAB and HTAC mono-layer film was obtained by withdrawing the glass plates very slowly out of a water solution. The withdrawing speed was about 5 mm/min and the typical concentration of surfactant was $\simeq 10^{-5}$ M.

All the cells are filled with a mixture of MR (Aldrich Chemicals) in 5CB (Merck). The MR concentration has been varied from 0.1 to 1.0% in weight. Typical cell thickness is $d = 10$ or $14\ \mu\text{m}$. Before filling the cells, the azo-dye doped liquid crystals are filtered, by centrifuging with Millipore filters of $0.22\ \mu\text{m}$ pore size, as well as degassed in a vacuum oven. For different MR concentrations, the cell absorption has been measured by using a commercial spectrometer (Ocean Optics UV-VIS 2000). The results are plotted in Fig.1, where the transmission coefficients T (at $\lambda = 514\text{ nm}$) are normalized to 1 with respect to the transmission of a cell filled with a pure liquid crystal and of the same thickness ($d = 14\ \mu\text{m}$).

For all the cells, the final quality of the liquid crystal alignment was tested by observing the sample under a polarizing microscope. All the differently prepared cells have shown perfect homeotropic orientation. However,

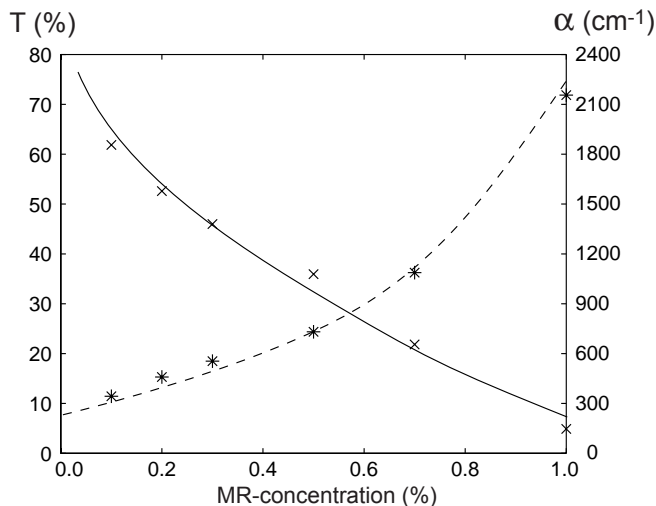


Fig. 1. Cell transmission $T(\%)$ and absorption coefficient $\alpha(\text{cm}^{-1})$ at $\lambda = 514 \text{ nm}$ and for different dye concentrations

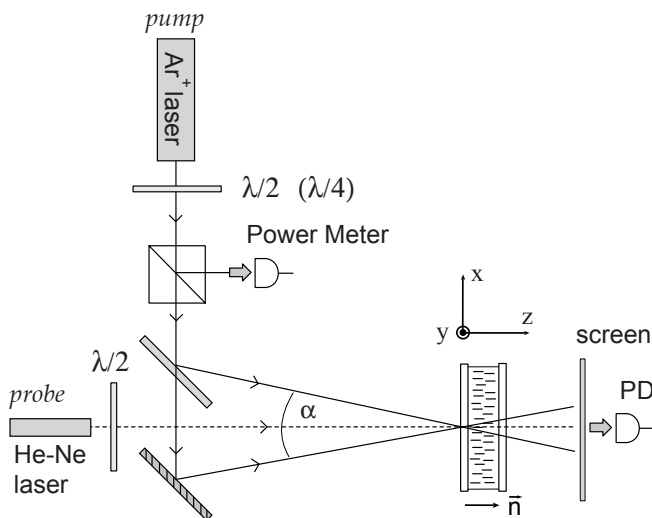


Fig. 2. Experimental setup. PD – photodiode, \vec{n} – liquid crystal nematic director, $\lambda/2$ ($\lambda/4$) – half-wave (quarter-wave) plate

only the cells treated with an ionic surfactant have displayed a giant optical response, that is, an extremely large nonlinear coefficient and this even without any externally applied d.c. field. Moreover, in our experiments, we have seen that the cells treated with HTAC display the largest response, that is, give high diffraction efficiencies at light intensities lower than the ones required for HTAB-treated cells. The larger surface mobility of the HTAC cells with respect to the HTAB ones can be easily explained if we consider that Br^- ions have a five times greater binding affinity for cationic hexadecyltrimethylammonium HTA^+ surfactant micelles than Cl^- ions have [24].

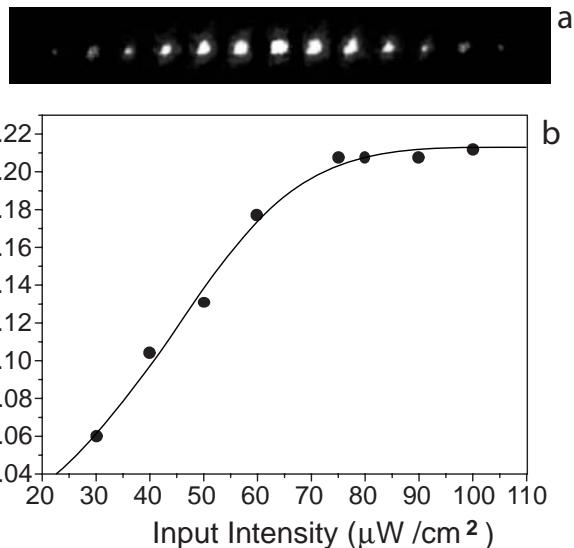


Fig. 3. Diffraction pattern (a) and diffraction efficiency as a function of the input pump intensity (b)

Experimental Setup

The experimental setup is shown in Fig. 2. The liquid crystal cell is placed at the intersection of two linearly polarized Ar^+ laser beams. The angle between the two beams is $\alpha \simeq 1^\circ$, corresponding to a fringe spacing of $\Lambda \simeq 18 \mu\text{m}$. The two Ar^+ beams propagate in the $x-z$ plane and are at normal incidence, that is, are symmetrically placed with respect to the normal of the cell. A linearly polarized 0.8 mW He-Ne laser is used to probe the grating.

Diffraction Efficiency

We measure the first-order diffraction efficiency $\eta = I_{+1}/I_0$, where I_{+1} is the intensity of the first diffracted order and I_0 is the total intensity of the probe beam. For the cells prepared with HTAB or HTAC, the diffraction efficiency is very high ($\eta \simeq 0.20$) even at normal incidence and without any bias voltage applied. Neither Linkam S.I. cell nor the cells prepared with Lecithin, Nissan Chemicals or DMOAP alignment agents give such a large reorientational effect.

A typical diffraction pattern observed for a HTAB cell is shown in Fig. 3,a. This pattern has been obtained for a total input light intensity of $I_{\text{in}} = 250 \mu\text{W}/\text{cm}^2$ and for the pump and the probe orthogonally polarized (p -polarized pump and s -polarized probe). In Fig. 3,b, the diffraction efficiency η is plotted as a function of the input pump intensity I_{in} . Similar results can be obtained

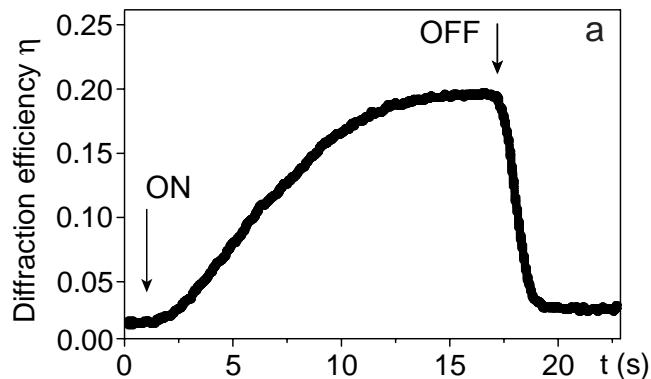


Fig. 4. Temporal response of the HTAB coated cell for switching on and off of the writing beams

for *s*-polarized pump and *p*-polarized probe. However, maximum diffraction was obtained for horizontal pump polarization.

As usual [7], we can express the refractive index change as $\Delta n = n_2 I_{in}$ and estimate the nonlinear coefficient n_2 from the first-order diffraction efficiency in the Raman–Nath regime, $\eta \simeq (\pi \Delta n d / \lambda)^2$. By substituting $\eta \sim 0.20$ at $I_{in} \sim 100 \mu\text{W}/\text{cm}^2$, $\lambda = 0.632$ and $d = 10 \mu\text{m}$, we get $\Delta n \simeq 1.0 \cdot 10^{-2}$ and thus $n_2 \simeq 75 \text{cm}^2/\text{W}$, which is an extremely large value of the nonlinear coefficient [18].

Typical response time is about 20 s, as can be seen in Fig. 4, where the diffraction efficiency is reported as a function of time for a total pump intensity $I_{in} = 100 \mu\text{W}/\text{cm}^2$.

Self-diffraction is not observed in the whole range of very low pump intensities in which we operate. We note, however, that self-diffraction appears at higher intensities, as well as in the transient regime induced by the switching of the pump polarization from one direction to the orthogonal one. This transient behavior does indeed prove that the LC reorientation is first established in the plane orthogonal to the pump polarization.

The photo-induced voltage, measured for $I_{in} = 2 \text{mW}/\text{cm}^2$, has a typical value of the order of 10 mV. As was already noticed [8], such a field is much lower than the one usually required for director reorientation, especially at the very low total input power, at which we observe the light-induced reorientation. However, close to the ITO layer, an interface is formed between the ionic surfactant and the cyano-heads of 5CB and the photo-induced voltage may change the mobility and hence the concentration of ions and counterions at this interface. As a result, at the incidence surface the anchoring

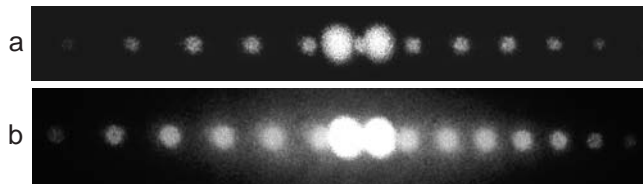


Fig. 5. Probe diffraction for $c = 0.5 \%$: a – $I_{in} = 150 \mu\text{W}/\text{cm}^2$ and b – $I_{in} = 1.5 \text{mW}/\text{cm}^2$

strength is weakened so that liquid crystals close to this surface start to reorient.

Nonlinear Coefficient at Different Dye Concentrations

We have repeated the diffraction experiments by changing the dye concentration. At low concentrations, low intensities would show nothing and patterns would not start to appear until around $600 \mu\text{W}/\text{cm}^2$. However, after getting a signal with the higher intensities, the same cell showed a response even at lower intensities and a diffraction pattern would start to appear. The origin of this memory effect has to be related to the induced photovoltaic effect. At the early times of illumination, only a few MR molecules are excited and, at the same time, those molecules are uniformly distributed in volume. Because of the photovoltaic effect, the MR molecules start to move towards the surface of the confining ITO coated plates. By keeping the laser onto the cell, the more and more MR molecules get excited and go close to the surface. Eventually, at high laser intensity, MR molecules can bind chemically to HTAC molecules, leading to the formation of permanent gratings [15].

As known, excited azo-dye molecules try to orient themselves normally to the light polarization. The conformational changes of the dye molecules induce first the reorientation of liquid crystal molecules close to the surface of the cell. Then, the reorientation propagates in the bulk and a diffraction pattern appears, as shown in Fig. 5, a.

When the laser is shone onto the cell for a second time, a lesser energy is needed to stir the molecules as some have already moved, and so already being excited, start to create an easy axis at the surface. For higher concentrations of the dye, such as $c = 0.5 \%$, responses could be obtained at lower intensities such like, $56 \mu\text{W}/\text{cm}^2$. However, this had the consequence of starting to diffuse at lower input intensities. Indeed, when the concentration increases, or the pump intensity is high, the molecular reorientation becomes irregular and is characterized by large fluctuations. This is a

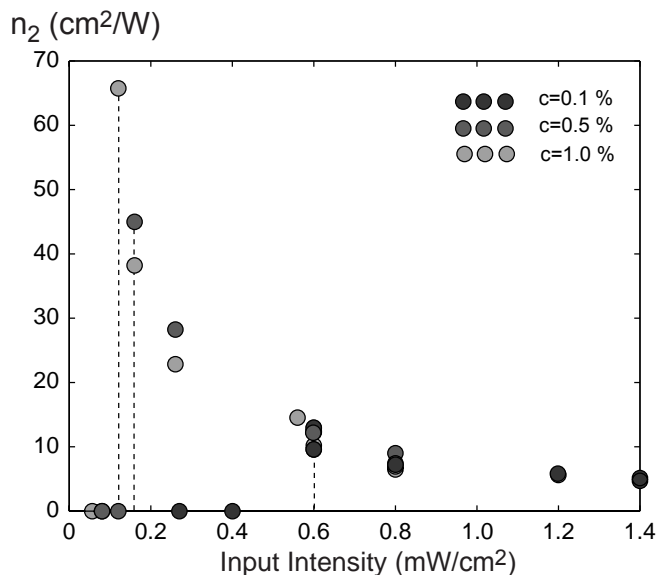


Fig. 6. Nonlinear coefficient as a function of the input pump intensity. For each concentration, a dashed line marks the values at which a diffraction signal starts to be distinguishable

consequence of out-of-axis components of the reorientation, which leads to the self-diffraction and increased light absorption. In Fig. 5,*b*, we show a diffraction pattern observed for the horizontal pump polarization at $I_{in} = 1.5 \text{ mW/cm}^2$ and $c = 0.5 \%$. For these values of I_{in} and c , the index grating becomes unstable and a large light scattering appears in all directions.

For a fixed concentration, $c = 0.1, 0.5$ and 1.0% , we varied the pump intensity and we measured the diffraction efficiency. Then, we have evaluated the nonlinear coefficient n_2 . In Fig. 6, the values of n_2 are plotted against the pump intensity. For each different dye concentration, a dashed line marks the value of the input intensity, at which a diffraction pattern is distinguishable at the first stage of the cell illumination. However, it can be noted that all the measured values of n_2 fit on the same curve, independently of the concentration. If we plot the inverse of n_2 as a function of I_{in} , we can see that all the points scale linearly with the input intensity. This means that the sensitivity of the cell does indeed not depend on the input pump intensity, since this linear dependence is a consequence of the definition of n_2 ,

$$n_2 = \frac{\lambda}{\pi d} \frac{\sqrt{\eta}}{I_{in}}.$$

The light-induced reorientation results as a combination of the surface effect with the bulk features

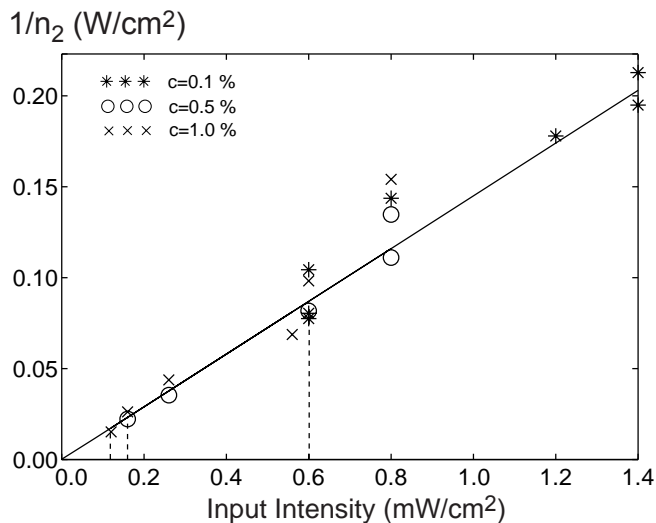


Fig. 7. Inverse of the nonlinear coefficient as a function of the input pump intensity. The straight line is a linear fit with a slope of 0.145. Dashed lines mark the values at which a diffraction signal is distinguishable

of the MR molecules. The reorientation starts at the surface, because of the migration and excitation of the MR molecules close to the confining plate, and then propagates in the bulk. As the surface induced reorientation sets in, dye-doped liquid crystals begin to absorb more and more light because of the dye dichroism and the diffraction signal grows with a long time scale. This slow process is accompanied by the *trans* – *cis* photoisomerization of dye molecules and saturates after a time that decreases with increasing the input light intensity [17].

The *cis* – *trans* relaxation process may be either a thermal one either a photoinduced transition. At relatively high values of MR concentrations, the absorption of the probe beam ($\lambda = 633 \text{ nm}$) may become sufficiently large to induce a significant role of the *cis* – *trans* photoinduced transition [25]. A transmission spectrum of a $c = 0.5 \%$ dye-doped cell is shown in Fig. 8, where a small dip may be distinguished around the wavelength of the probe beam.

Circular Polarization Experiments

In order to get information about the trajectory of a nematic director during the light induced reorientation, we have checked the response of liquid crystal cells under the action of circularly polarized laser beams [20]. In the experimental setup, as shown in Fig. 2, we have

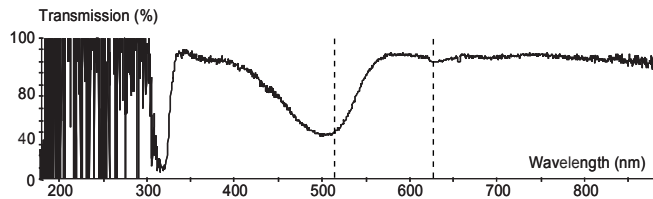


Fig. 8. Transmission spectrum of a $c = 0.5\%$ dye-doped cell. Dashed lines mark the wavelengths of the pump (514 nm) and of the probe (633 nm)

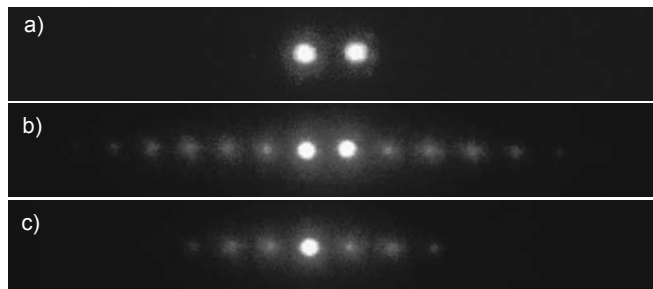


Fig. 9. Self-diffraction of two circularly polarized pump beams: *a* — start-up signal and *b* — stationary diffraction pattern. *c* — stationary pattern observed once one of two pump beams has been switched off

inserted a $\lambda/4$ wave-plate at the exit of an Ar^+ laser, so that two pump beams are circularly polarized, both in the same direction.

In Fig. 9, we show a typical self-diffraction pattern observed after a HTAC coated cell ($d = 10\ \mu\text{m}$, $c = 0.3\%$) for a total input light intensity $I_{\text{in}} = 2.0\ \text{m W/cm}^2$ and for the right-handed circular polarization of two pump beams (left-handed polarization leads to the same results). The response time is very slow, and in particular it takes quite a long time (several tens of seconds) to see the appearance of the first diffraction order.

In Fig. 10, the self-diffraction efficiency $\eta_s \equiv 2I_{+1}/I_{\text{in}}$ is plotted as a function of time. I_{+1} is the intensity measured for the first self-diffracted order of one pump and $I_{\text{in}}/2$ is the intensity of one pump. It can be noticed that once the diffraction grating is established, the signal is characterized by large fluctuations. Indeed, since the pump beams are circularly polarized, the self-diffracted signal is the sum of the projections of the nematic director in the transverse plane (plane $x - y$ in Fig. 2), thus very sensitive to fluctuations, both along the radial and azimuthal directions. Once we switch off one of the pumps, as marked by the arrow in the plot, the amplitude of fluctuations decreases drastically and the signal stabilizes in long time onto a lower value but does not go to zero. The corresponding diffraction

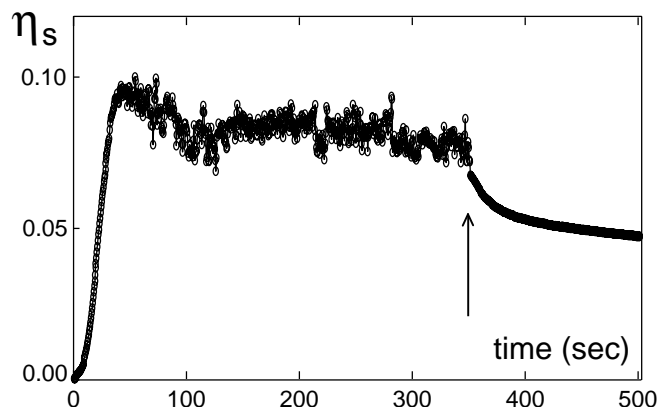


Fig. 10. Self-diffraction efficiency η_s as a function of time. The arrow indicates the point where one of the pump beams was switched off

pattern is shown in Fig. 9, *c*. Similar memory effects have also been observed in other liquid crystal cells [26] coated with photoconductive polymer and filled with the same mixture of MR in 5CB. In that case, it was shown that the bulk photoisomerization of the dye can be controlled by pre-illumination of the sample.

Pump-probe experiments are performed for a linearly polarized probe, both s and p components, analyzed by a crossed polarizer after the liquid crystal cell. For p -polarized probe, the intensity I_p transmitted after the analyzer is given by the usual formula [27]

$$I_p = I_0 \sin^2(2\theta) \sin^2(\Delta\Phi/2), \quad (1)$$

where I_0 is the probe input intensity, $\Delta\Phi = (2\pi/\lambda)\langle\Delta n(z)\rangle d$ is the total phase shift integrated over the cell length, and $\Delta n(z) = n_{\text{eff}}(\theta, \beta, z) - n_0$, n_{eff} and n_0 being the refractive indices for the extraordinary and ordinary waves, respectively, and the brackets indicating the integration along z . $0 < \theta < \pi/2$ and $0 < \beta < \pi/2$ are the in-plane and out-of-plane reorientation angles, respectively, where the plane of reference is the incidence plane of the light, which is also the plane containing the nematic director \vec{n} before the onset of the reorientation (plane $x - z$ in Fig. 2). For s -polarized probe, the intensity I_s passing through the analyzer is given by the same formula, Eq. (1), with β replacing θ .

As shown in Fig. 11, when we switch on the pumps, for both s and p polarizations of the probe, we observe a signal on the photodiode. The diffraction efficiencies $\eta_\theta \equiv I_{p+1}/I_0$ and $\eta_\beta \equiv I_{s+1}/I_0$ are evaluated by normalizing the intensities I_{p+1} and I_{s+1} of the first orders to the total probe intensity I_0 . During the build-up of the grating, there is first a signal for the p -polarized

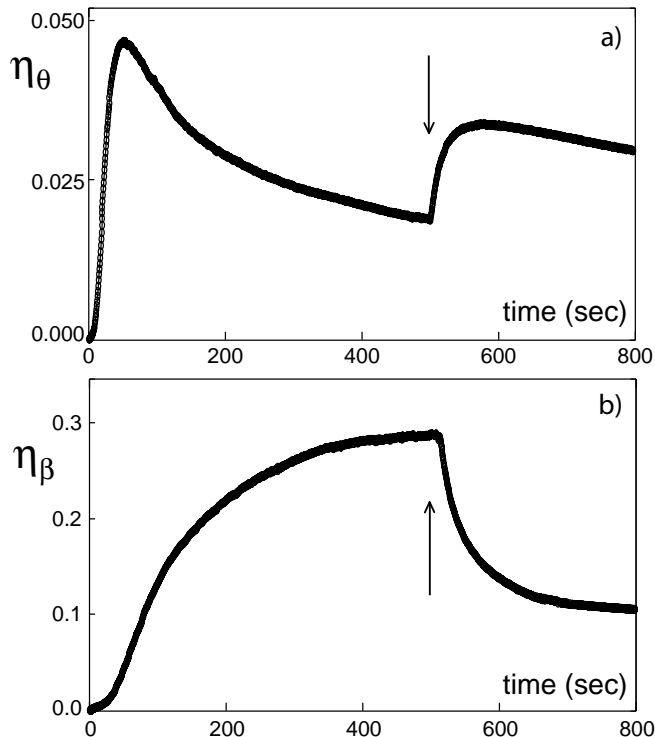


Fig. 11. Diffraction efficiency $\eta_\theta = I_{p+1}/I_0$ for p -polarized probe (a) and $\eta_\beta = I_{s+1}/I_0$ for s -polarized probe (b). Arrows correspond to switching off one of the pumps

probe (Fig. 11,a), whereas there is no signal for the s -polarized component (Fig. 11,b). This indicates that a relatively fast reorientation takes place in the plane of incidence of light. Then, the in-plane motion stops (correspondingly the signal in Fig. 11,a decreases) and a slow out-of-plane motion starts, as indicated by the rise of the signal in Fig. 11, b. In the stationary state, η_θ has considerably decreased, which means that the final configuration of an initially homeotropic cell is very similar to that of a planar one, with an almost vertical director. By switching off one of the two pumps (arrows in Fig. 11), we observe a decrease of η_β accompanied by an increase of η_θ , reflecting the reversed motion of the director along its original trajectory. Then, the two signals stabilize at intermediate values, but they do not go to zero, that is, the grating is still present, according to the self-diffracted signal.

The fact that the first component of the director motion is the in-plane one is related to the presence of a small symmetry breaking at the surface of the cell. Indeed, this can be associated to the direction of withdrawing the glass plates during the deposition of a surfactant layer. If we turn the cell by 90° , we observe the same behavior for the probe diffraction, but the role

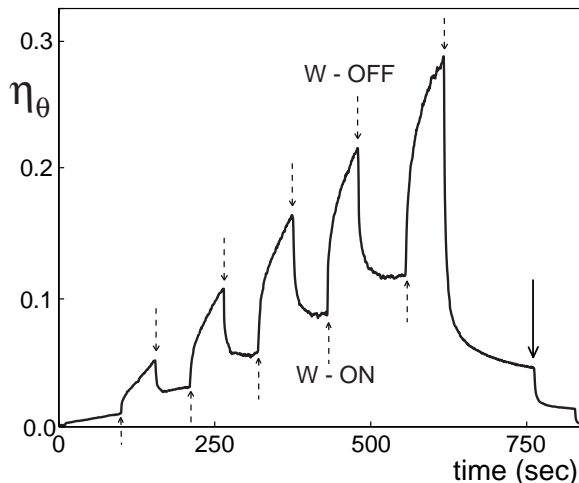


Fig. 12. White light response for p -polarized probe. Dashed arrows indicate switching on and off the white light. Solid arrow indicates the switch off of one pump

of s and p polarizations is exchanged, that is, the first and fast component of the director motion is the out-of-plane one, whereas the successive slow component is the in-plane one. These observations support the conjecture that the first and relatively fast component of the director motion is the one resulting from the surface-induced reorientation, whereas the successive slow component is related to the *trans* – *cis* photoisomerization of the dye dopants.

White-light Experiments

Finally, we have illuminated the cell with two circularly polarized pumps at the normal incidence and the total intensity $I_{in} = 1 \text{ mW/cm}^2$ well below the threshold for the liquid crystal reorientation, and we have sent onto the cell an additional white light illumination. The white light source is supplied by an halogen lamp with an optical fiber output and with a large emission spectrum in the visible range of wavelengths (450 – 750 nm).

The total white light intensity on the cell is $\sim 10 \text{ mW/cm}^2$. The white light illumination is sent on the back side of the cell, that is, counterpropagates to the laser light. It must be noted that the white light source does not contain UV wavelengths and, in any case, the glass windows of our LC cell are filtering out any spectral component with a wavelength smaller than 300 nm, as we have verified by spectrometric measurements.

When we shine the cell with white light, for both s and p -polarized probes, we observe an increase of the diffracted signal, as indicated by the dashed arrows (*w - on*) in Fig. 12,b. The signal decreases as white

light is switched off (*w - off*). Further illumination cycles lead to a continuous increase of the cell response, confirming that the light-driven motion of the director is mainly maintained by the process of photon injection, independently of the properties of coherence of the light source.

Discussion of the Results

The cells were filled with azo-dye doped liquid crystals at different concentrations, and a light beam shone through each one to obtain results on how different concentrations and intensities affect the overall value of the non-linear coefficient n_2 . From the experimental observations, it can be concluded that there must be a compromise between the value of the concentration and the intensity in order to get the best results. Low concentrations can give good diffraction patterns with intensities up to 1.4 mW/cm^2 , however they don't seem to have reproducible results using intensities below $600 \mu\text{W/cm}^2$, since giant responses are observed only after pre-illumination of the sample. Higher concentrations give an extremely high sensitivity from the early times of illumination, and this with intensities as low as $56 \mu\text{W/cm}^2$. However, the drawback of high concentrations is that a huge light diffusion occurs at moderately higher intensities, as a consequence of the grating degradation. All these observations indicate that the *trans - cis* photoisomerization of dyes is an important mechanism participating to the enhanced reorientation in dye-doped liquid crystals. Recent experiments with circularly polarized pumps have also shown the important role of dyes in sustaining the liquid crystal reorientation [20].

All our observations demonstrate that the motion of the nematic director is composed of two contributions, parallel and orthogonal to the light incidence plane, respectively. The two contributions follow two different time scales, the first one being related to a relatively fast (tens of seconds) motion and to the surface contribution, the second one being characterized by the slow (hundreds of seconds) features of the *trans-cis* conformation of azo-dye dopants in the bulk of a cell.

The extremely high sensitivity of the cell treated with ionic surfactants is a strong indication of the surface effect. Indeed, the action of the photo-induced voltage on the ionic layers may be at the basis of the huge nonlinear response observed for this kind of cells. Note that this huge nonlinearity cannot be obtained with another kind of the known alignment agent.

Conclusions

We have presented a large number of measurements of the nonlinear optical response in azo-dye doped NLC cells. We have changed the dye concentration and we have tested the cells in different experimental geometries for the linear or circular polarization of pumps. All the results give indications that the high sensitivity of homeotropically aligned azo-dye doped NLC is related to the presence of an ionic surfactant on the side-walls of the cell.

By illuminating NLC cells with two pump beams circularly polarized in the same direction, we have shown that the motion of the nematic director is characterized by a relatively fast in-plane component followed by a much slower and out-of plane component. We suggest that the two contributions are associated to the surface and bulk effects, respectively. Finally, we have shown that the diffraction grating presents memory effects which are related to the photoisomerization of azo-dye dopants in the cell bulk.

This article is dedicated to Prof. Marat Soskin on the occasion of his 75-th birthday in April 2004.

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ГІГАНТСЬКИЙ ОПТИЧНИЙ ВІДГУК РІДКИХ КРИСТАЛІВ З ДОМІШКОЮ АЗО-БАРВНИКА: ЕФЕКТИ НА ПОВЕРХНІ ТА В ОБ'ЄМІ

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Резюме

Наведено результати експериментальних досліджень гігантського оптичного відгуку нематичних рідких кристалів з домішкою азо-барвника. Показано, що дуже сильний відгук можна отримати при гранично низьких потужностях світла в декілька десятків мкВт/см². Причиною нелінійності є дія фотоіндукованого електричного поля на приповерхневий шар, який є іонно-поверхневоактивним. Наведено залежності нелінійного відгуку від концентрації азо-барвника. За допомогою експериментів з циркулярною поляризацією доведено, що реорієнтація директора нематичного рідкого кристала відбувається як в площині, так і поза площиною падіння світлової хвилі. Це вказує на домінуючий вплив фотоізомеризації азо-барвника на об'ємні ефекти.