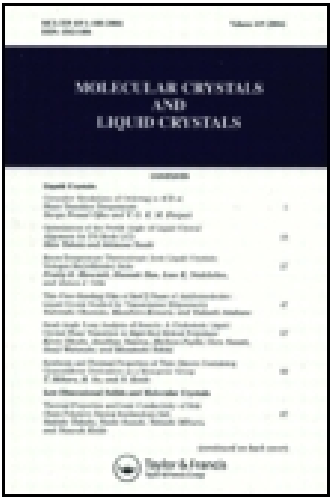

Molecular Crystals and Liquid Crystals Science and Technology. Section A. Molecular Crystals and Liquid Crystals

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

Photoalignment Effect Induced by Angular Momentum of Light in Dye-Doped Liquid Crystals

D. Andrienko [a], D. Fedorenko [a], Yu. Reznikov [a], S. Slussarenko [a b], O. Francescangeli [b] & F. Simoni [b]

[a] Institute of Physics, 46 Prospect Nauki, Kiev, Ukraine

[b] Dipartimento di Scienze dei Materialie e della Terra, Instituto Nazionale per la Fisica della Materia, Universita' di Ancona, Via Brecce Bianche, 60131, Ancona, Italy E-mail:

Published online: 24 Sep 2006.

To cite this article: D. Andrienko, D. Fedorenko, Yu. Reznikov, S. Slussarenko, O. Francescangeli & F. Simoni (1999) Photoalignment Effect Induced by Angular Momentum of Light in Dye-Doped Liquid Crystals, Molecular Crystals and Liquid Crystals Science and Technology. Section A. Molecular Crystals and Liquid Crystals, 329:1, 1-10, DOI: 10.1080/10587259908025919

To link to this article: http://dx.doi.org/10.1080/10587259908025919

Mol. Cryst. Liq. Cryst., 1999, Vol. 329, pp. 1-10
Reprints available directly from the publisher
Photocopying permitted by license only

Photoalignment Effect Induced by Angular Momentum of Light in Dye-Doped Liquid Crystals

D. ANDRIENKO[a], D. FEDORENKO[a], YU. REZNIKOV[a], S. SLUSSARENKO[ab], O. FRANCESCANGELI[b] and F. SIMONI[b]

[a]*Institute of Physics, 46 Prospect Nauki, Kiev, Ukraine and* [b]*Dipartimento di Scienze dei Materialie e della Terra and Instituto Nazionale per la Fisica della Materia, Universita' di Ancona, Via Brecce Bianche 60131, Ancona, Italy; email: denis@iop.kiev.ua*

We studied the effective interaction of the angular momentum of light with a liquid crystal (LC) doped with azo-dye. The experiments were carried out in a combined cell with one substrate providing strong unidirectional planar alignment of LC and the other covered by isotropic polymer layer providing a degenerated planar alignment. The effect manifests itself as azimuth reorientation of the liquid crystal director under the action of circularly polarized light in the absorption band of azo-dye. The new orientation of LC was captured by the command surface because of a light-induced alignment memory effect. To describe the experimental dependencies we solved the self-consistent problem of the propagation of a circularly polarized light in the absorbing nonlinear media using the geometric optics approximation. Comparing the experimental data with numerical evaluation we obtain the basic parameters of the system: the orientation nonlinearly coefficient and the value of anchoring of the director with the polymer surface.

Keywords: liquid crystal; azo-dye; light angular momentum

INTRODUCTION

Laser-beam propagation in nematic liquid crystals (LCs) is a fascinating problem and exhibits unique and promising for applications nonlinear optical effects. The grounds of these effects are anisotropic character of the dielectric properties and large optical nonlinearity of LCs.

It is worth noting that a variety of optically induced reorientation effects are not observed when a liquid crystal is reoriented by a static field. Indeed, liquid crystals display an unusually rich nonlinear dynamical behavior when reoriented by light beam. It is well known that above the transition threshold a linearly polarized light beam distorts the homeotropic alignment of the LC film, reorienting the molecules against the elastic torque. At the same time, a circularly polarized beam can, in addition to the distortion of alignment, cause the molecules to precess uniformly about the surface normal.[1] The latter is the consequence of a constant rate of deposition of angular momentum from the field to the medium. Moreover, the elliptically polarized light can lead the system through various dynamic regimes: torsional oscillation, nonuniform precession, and others.[2,3] These effects should be ascribed to the fact that optical fields, unlike static, do carry angular momentum.

In most materials, the effects of optical torque is hardly detectable even when generated by the intense light of laser beam. On the contrary, in liquid crystals the optical torque induces a large change of the molecule average orientation. Moreover, it is possible to increase the orientational nonlinearity coefficient of pure LC adding some photosensitive dyes. Actually, it was discovered that in dye doped liquid crystals the value of Frederiks threshold is smaller by more than two orders of magnitude than that of the transparent host.[4] Then, using non-threshold geometry and more sensitive z-scan technique Jánossy et.al.[5] proved that the mixture dye + LC possesses strong orientational nonlinearity. Moreover, depending on the specific dye utilized the orientational nonlinearity coefficient can be enhanced by two orders of magnitude, or it can be canceled out, or even sign-inverted. In all cases the birefringence of the material and elastic constants were found approximately unchanged.

The above effects of the director reorientation are due to the interaction of the light field with the entire LC bulk. Alignment surfaces only determine the boundary conditions during the reorientation process. However, the reorientation effect of surface-induced origin was also found in the dye-doped liquid crystals.[7] Under the light illumination the anchoring parameters of the bounding surfaces change as a result of the light action on the bulk of a light-sensitive mixture. In particular, easy orientation axis was induced parallel to the polarization of the laser beam. The effect was explained in the frame of the idea of adsorption of phototransformed molecules onto the aligning surfaces with subsequent capturing of the director along the polarization direction.

Further investigations figured out that this alignment effect is sensitive not only to the polarization direction of the incident light wave but also to the polarization state of it, i.e. ellipticity which is related to the angular momentum carried by the light wave. This is the reason why we studied the force and the torque transferred to the liquid crystal director by circularly

polarized light wave. Such a momentum transfer leads to the reorientation of the director and new orientation can be captured then via the proposed mechanism of adsorption of the azo-dye molecules onto the aligning surface.

THEORY

Consider a NLC cell of thickness d confined between the planes $z = 0$ and $z = L$ of a Cartesian system of coordinates. Let the director $\boldsymbol{n} = (\cos\varphi, \sin\varphi, 0)$ describes the average molecular orientation in the cell. We assume $\varphi = \varphi(z)$ and consider the equilibrium orientation of the director, so that the angle φ does not depend on time.

Let the elliptically polarized monochromatic light wave of frequency ω be normally incident on the cell along z-axis. To obtain the stationary distribution of the director in the cell we use the variational approach which permits us to find conserved quantities. The variational function of system is

$$f = f_{el} + f_E + f_{dye} \tag{1}$$

where $f_{el} = \frac{1}{2} K_2 (\partial\varphi/\partial z)^2$ is the NLC elastic energy, K_2 is twist elastic constant, f_E is the total electromagnetic energy density of the incident light wave, f_{dye} is the contribution to the free energy due to the dye molecules.

Following the works[4-6] we assume that the contribution of the azo-dye assisted nonlinearity to the total free energy density is similar to the usual orientational nonlinearity of NLC with parameter η which characterizes the strength of the interaction between dye and NLC molecules

$$f_{dye} = \eta f_E \tag{2}$$

To obtain the expression for the electromagnetic energy density f_E we assume that NLC is slowly varying uniaxial medium, so that $\varphi(z)$ varies appreciably over a length much greater than the optical wavelength λ. We also assume that the birefringence of the media is low, i.e. $n_e - n_o << 1$. In this case the polarization state of the light is also slowly varying through the medium, and we can use the geometric-optics approximation (GOA) solving the Maxwell's equations for the field in the cell.[9] Again, for the small concentrations of adsorbing impurity we can use expressions valid for non-absorbing medium, i.e. use that the beam intensity I defined as the z component of the average Poynting vector, remains constant.

Following the consideration proposed by Santamato *et. al.*[9] the total electromagnetic energy density f_E of light wave in the non-adsorbing, nonmagnetic medium can be rewritten in terms of the light intensity I,

ellipticity e and major axis angle ψ and in the limit of low birefringence simplifies to

$$f_E = \frac{In_o}{c} + \frac{I}{2c}[n_e - n_o]\left\{1 + (1-e^2)^{1/2}\cos 2[\psi - \varphi]\right\} \quad (3)$$

where $n_{o,e}$ are ordinary and extraordinary indices of the material, respectively, I is the z component of Poynting vector which equals to the total intensity of the light wave. We introduce also the ellipticity e of the polarization ellipse $e = S_3/S_0$ and the angle $\psi = \arctan(S_2/S_1)$ that ellipse's major axis forms with the x axis. Here S_i are the Stokes parameters.

Equations governing the polarization of light can be derived from the electromagnetic energy density f_E provided one considers f_E as Hamiltonian function, having ψ as generalized coordinate and $l_z = -(I/\omega)e$ as conjugate momentum. l_z is the average angular momentum carried by the optical beam along the propagation direction.

$$\frac{\partial \psi}{\partial z} = -\frac{\omega}{2c}[n_e - n_o]\frac{e}{(1-e^2)^{1/2}}\cos 2(\psi - \varphi),$$

$$\frac{\partial e}{\partial z} = \frac{\omega}{c}[n_e - n_o](1-e^2)^{1/2}\sin 2(\psi - \varphi) \quad (4)$$

The first integral can be obtained from the free energy density, since the latter does not depend explicitly on z coordinate and permits, in accordance to Noether theorem, the integral of 'energy'

$$\frac{K_2}{2}\left(\frac{\partial \varphi}{\partial z}\right)^2 + (1+\eta)\frac{I}{2c}(n_e - n_o)(1-e^2)^{1/2}\cos 2(\psi - \varphi) = E \quad (5)$$

Moreover, free energy density also permits the integral of conservation of the total angular momentum flux along the propagation direction - the sum of the elastic and optical angular momentum

$$K_2\frac{\partial \varphi}{\partial z} + (1+\eta)\frac{I}{\omega}e = M \quad (6)$$

Eliminating director angle in (5) with help of (4-6) one can obtain the equation for the polarization ellipse

$$\left(\frac{\partial e}{\partial z}\right)^2 = \left[\frac{\omega(n_e - n_o)}{c}\right]^2(1-e^2) - \left[E - \frac{1}{2K_2}\left(M - \frac{(1+\eta)Ie}{\omega}\right)^2\right]^2\left(\frac{2\omega}{(1+\eta)I}\right)^2 \quad (7)$$

As it was noted by Santamato *et. al.*[19] it can be solved analytically in general case in terms of elliptic integrals. Below we pay attention to the combined cell with one surface providing fixed orientation of the director with strong anchoring and the other giving azimuthaly degenerate orientation of liquid crystal (zero azimuthal anchoring)

$$\varphi\big|_{z=L} = 0,\ \left.\frac{\partial\varphi}{\partial z}\right|_{z=0} = 0. \tag{8}$$

The incident light wave is circularly polarized and the cell is faced to the light wave with plate possessing zero anchoring

$$e\big|_{z=0} = 1. \tag{9}$$

One can see that in this case the constants E and M can be easily determined from the equations (6,7): $M = (1+\eta)I/\omega$, $E = 0$. Then integration of (8) yields

$$e = 1 - \frac{q\alpha_r(1-\mathrm{cn}(u,m))}{p+q+(p-q)\mathrm{cn}(u,m)}, \tag{10}$$

where $\mathrm{cn}(x,m)$ is Jacobi elliptic function, α_r is the real root of the equation

$$2-\alpha-r\alpha^3 = 0,\ r = \left[\frac{c(1+\eta)I}{\omega^2 K_2(n_e-n_o)}\right]^2,\ p^2 = \frac{1}{r}+3\alpha_r^2,\ q^2 = \frac{1}{r}+\alpha_r^2,$$

$$u = \sqrt{rpq}\,\frac{\omega}{c}(n_e-n_o)z,\ m = \sqrt{\frac{\alpha_r^2-(p-q)^2}{pq}}.$$

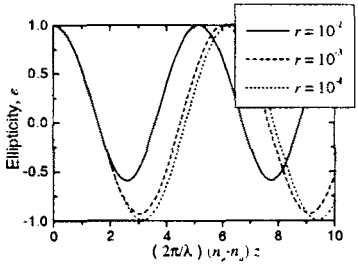

Figure 1 Ellipticity *vs.* dimensionless cell thickness

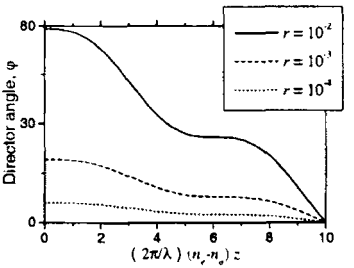

Figure 2 Director deviation *vs.* dimensionless thickness

Corresponding dependencies of the ellipticity and the director angle on z coordinate are presented in Fig. (1,2).

For small director deviations that correspond to small light intensities and, therefore, small values of parameter r, expansion of the director distribution in the power series by r yields the approximate expression for the resulting twist angle

$$\varphi_{z=0} \approx \sqrt{r}(\Delta - \sin\Delta),\ \Delta = 2\pi(n_e-n_o)L/\lambda \tag{11}$$

Let us emphasize the distinguishing features of the light propagation in the planar cell with strong orientational nonlinearity. First we pay attention to the evolution of the light polarization in the cell. For the LCs with typical value of orientational nonlinearity (dimensionless parameter $r \sim 0$) circularly polarized incident light wave becomes elliptically polarized in the cell and sign-inverts its value over the $\lambda/(n_e - n_o)$ distance. The ellipticity is the periodical function of z coordinate and change its value between 1 and -1. However, if we increase intensity of light or get the mixture with larger orientational nonlinearity coefficient the director will be more sensitive to the light field and immediately deviates from the initial planar distribution and the light wave will propagate in the uniaxial media with non-uniform distribution of the optical axis. As a result the ellipticity of the incident light wave does not reach its sign-inverted value, i.e. -1. The bigger is the orientational nonlinearity the lesser is the deviation of the ellipticity from the initial value $e = 1$. Moreover, aside the change in the deviation amplitude, the period of the spatial modulation of ellipticity decreases with increasing of the light intensity.

At the same time, the director deviation depends strongly on the value of orientational nonlinearity. One can easily show from (7) that under the given boundary conditions $\partial\varphi/\partial z \geq 0$, i.e. the director deviation increases monotonically in the cell volume. Since the ellipticity of the polarization ellipse is periodic function of z coordinate and changes the sign in the cell volume the director distribution is superposition of the monotonically growing and oscillating functions. For the small director deviations the resulting twist angle is proportional to the light intensity and the cell thickness (13). Moreover, for large values of the parameter r it can exceed π. This means that for the strongly nonlinear media one can obtain supertwisted distribution of the director or even distribution that realizes in chiral nematics.

EXPERIMENT

The experiments were performed with a standard, sandwich glass cell arrangement filled with a mixture of the LC 4'-n-pentyl-4-cianobiphenyl (5CB) and azo dye methyl red (MR) as a dopant (weight concentration was about 0.5%).

The thickness of the cell was $d = 65\mu m$. The inner surface of the first glass was covered with isotropic layer of a modified poly(vinyl)-cinnamate (PVCN). The second glass was coated with a rubbed polyimide layer providing a strong anchoring of LC molecules with the surface. The polyimide surface determined the planar LC orientation in the cell.

The cell was placed normally to the exiting beam of He-Cd laser (λ = 0.44μm). The beam was focused onto the LC layer from the side of isotropic layer. The circular polarization of the beam was given by Berec polarization compensator. The intensity of the beam can be controlled by means of wedged filters.

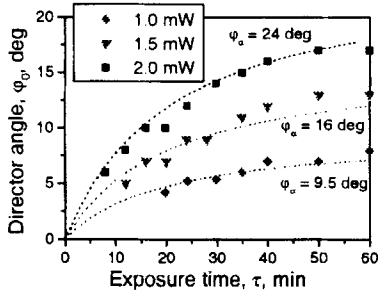

Figure 3 Dependence of the deviation angle on the exposure time for different intensities of laser beam

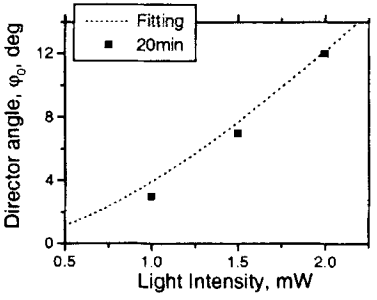

Figure 4 Director deviation as a function of light intensity. Exposure time is fixed (20min).

Observation in a polarizing microscope of the irradiated spots have shown that the twisted structure of the director was induced in the illuminated area. We measured the dependencies of the director deviation φ on the exposure time τ_{exp}, light intensity I_{exp}, and cell thickness d.

We observed that simultaneously with the director reorientation the photobleaching of the nematic mixture in the illuminated area was produced. The speed of the photobleaching increased with increasing I_{exp}. The analogous experiments were carried out with the cell filled with pure 5CB. We did not find director reorientation in this case.

We measured the dependence of the director deviation on the exposure time for several values of light intensity. The character of the dependencies was the same for all chosen intensities. First we observed monotonical growth of the director deviation with further saturation at large exposure. Extra increasing of the light intensity (I_{exp} > 4mW) leads to the fast bleaching of the irradiated spots and decreasing of the saturated value of the director angle.

The obtained results indicate that under the irradiation by circularly polarized light the easy orientation axis e appears on the initially isotropic surface. As a result the director reorients towards e and the twisted director structure is produced in the cell.

Let us discuss the mechanism of the easy axis induction. As it has been already noted the director in the combined cell in the light wave field growths monotonically with the cell thickness and the resultant deviation angle can be large enough for the media with large orientational nonlinearity coefficient. The director deviation is mainly produced by the anisotropic light absorption and reorientation of the MR molecules. Thus, the maximum of the distribution function of the MR molecules also deviates from the initial state along with the director. This MR distribution is captured due to the light-induced adsorption of the MR molecules onto the isotropic polymer surface leading to the light-induced anisotropy of the polymer surface.

Assuming that the maximum of the distribution function of the MR molecules coincides with the director and taking into account that the adsorption takes place under the influence of circularly polarized light we can state that the angle of the induced easy axis φ_a is equal to the director deviation φ induced by light. Moreover, for the small exposure the anchoring energy, which is proportional to the concentration of the adsorbed molecules and surface order parameter, increases linearly with exposure time and light intensity, $W = w\tau I$.

Thus, after the exposure we have an easy axis under the angle φ with the anchoring strength W. Note, that for the large cell thickness $\Delta = 2\pi(n_e - n_o)L/\lambda >> 1$ the easy axis angle (12) is proportional to the cell thickness L

$$\varphi = \frac{\lambda}{2\pi}\frac{(1+\eta)}{cK_2} IL. \tag{12}$$

The actual director angle φ_{exp} can be found from the implicit equation

$$\frac{WL}{K_2} = \frac{2\varphi_{exp}}{\sin 2(\varphi - \varphi_{exp})} \tag{13}$$

and for the small deviations of the director takes the form

$$\varphi_{exp} = \varphi\frac{WL/K_2}{1+WL/K_2}, \tag{14}$$

As we already mentioned, the angle φ can be obtained from eq.(12).

We used the expressions (12),(14) to fit the experimental dependencies $\varphi_{exp}(\tau)$. Both are presented in Fig.3. The varied parameters were photoinduced nonlinearity coefficient η and anchoring energy rate w. For both we got reasonable values: $\eta \sim -200$, $w = 0.45 \cdot 10^{-14}$ min-1. The obtained value of the anchoring rate w gives typical anchoring energy value $W \sim$ 10-12 J/m2 that corresponds to the anchoring parameter $WL/K_2 \sim 0.5$, i.e. the light-induced easy axis possesses comparably small strength.

We also measured the dependence of the director deviation on the intensity of the incident laser beam. Exposure time was fixed τ_{exp} = 20min and intensity varied in the range 2-4mW. The results are presented in fig (4). It is seen, that for the small intensities the director deviation is proportional to the squared light intensity (see Eq.(14)).

Now let us make some final comments. First, we should note that studied mixture MR + LC is adsorbing while the estimations have been done assuming it to be non-adsorbing. However, this only leads to the conclusion that some effective "cell thickness" plays role for the director reorientation, i.e. in eq.(12) we should use L_{eff} instead of the actual cell thickness L. As a result, we will get even higher coefficient of photoinduced orientational nonlinearity. Second, the bleaching of the MR makes the cell more transparent. But we restricted ourselves with dozes less than one need to observe the effect of the MR bleaching. Finally, the refractive index of the mixture also changes during the illumination because of trans-cis isomerization and bleaching. However, this can not influence the value of the surface director deviation: as eq.(12) states, the direction of the light-induced easy orientation axis does not depend on the refractive indexes.

CONCLUSIONS

We studied the interaction of the angular momentum of light with a liquid crystal doped with azo-dye. The dye-doped liquid crystal possesses strong orientational nonlinearuty which makes possible to reach measurable values of the azimuthal director deviations under the action of circularly polarized light. This deviation can be captured by polymer surface due to the light-induced adsorption of dye molecules onto the aligning surface.

We also consider the propagation of circularly polarized light in the strongly nonlinear media using geometric optics approximation and estimate orientation nonlinearly coefficient and the value of anchoring of the director with polymer surface.

Acknowledgments

The research described in this publication was made possible in part by grant No. UE1-315 of CRDF Cooperative Grants Program, grant No.4/358-97-13 of Ukrainian State Fund for Fundamental Studies Support, grant No. B29/13 of the Fund of the Academy of Sciences of Ukraine, INTAS grant No. 96-0359, and grant No. PSU082002 of International Soros Science Education Program.

References

[1] E. Santamato, B. Daino, M. Romagnoli, M. Settembre, Y.R. Shen, *Phys. Rev. Lett.* **61**(1), pp. 113–116 (1988)

[2] E. Santamato, G. Abbate, P. Maddalena, L. Marrucci, Y.R. Shen, *Phys. Rev. Lett.*, **64**(12), pp. 1377–1380 (1990)

[3] G. Abbate, P. Maddalena, L. Marrucci, L. Saetta, E. Santamato, *Physica Scripta,* **T39,** pp. 389–393 (1991)

[4] I. Jánossy, A.D. Lloyd, B.S. Wherret, *Mol. Cryst. Liq. Cryst.* **179,** p.1 (1990)
[5] I. Jánossy, T. Kósa, *Optics Letters* **17**(17), pp. 1183–1185 (1992)
[6] L. Marucci, D. Paparo, *Phys. Rev. E* **56**(2), pp. 1765–1772 (1997)
[7] D. Voloshchenko, A. Khyzhnyak, Yu. Reznikov, V. Reshetnyak, *Jpn. J. Appl. Phys.*, **34,** pp. 566–571 (1995)
[8] S. Slussarenko, O. Francescangeli, F. Simoni, Yu. Reznikov, *Appl. Phys. Lett.* **71**(25), pp. 3613–3615 (1997)
[9] E. Santamato, G. Abbate, P. Maddalena, *Phys. Rev. A* **38**(8), pp. 4323–4329 (1988)