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EXPLANATION OF THE ASYMMETRIC RESPONSE OF A DYE DOPED NEMATIC SAMPLE IN A LASER FIELD

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Abstract. We present a model for the asymmetric optical response of a sandwich type dye doped nematic cell in a pump-probe experiment. The different initial anchoring strength on the two polymer surfaces and the absorption of the Ar^+ laser beam explain the time dependent rotating angle of the nematic director.

Key words: liquid crystals, dyes, molecular reorientation, asymmetric response.

Explanation - asymmetric response of dye doped nematics

INTRODUCTION

Dye doped nematic liquid crystals (DDNLCs) exhibit enhanced orientational nonlinearity, up to three orders of magnitude larger than in pure liquid crystals [1]. For these reason new opportunities of application appear such as optical storage and image processing.

The explanation proposed by Janossy [2] is related to the formation of metastable states in the absorption process: an excited electronic structure (singlet or triplet) or a metastable configuration of the nuclear positions (like the *cis*-configuration of an azo-dye). A dye molecule in the metastable state interacts with the surrounding host NLC molecules differently than a dye molecule in the ground state. The change of interaction modifies the collective behavior of NLC and thus causes large optical effects.

A surface driven reorientation effect as a result of the light action on the bulk of a light-sensitive NLC-azo-dye mixture was reported by Voloschenko and co-workers [3]. The explanation was the adsorption of dye phototransformed molecules onto the aligning photopolymer surface.

Systematic studies about the diffraction efficiency of a LC-dye mixture revealed the basic role of light-induced adsorption and desorption of the dye molecules onto the boundary surfaces (SINE - Surface-Induced Nonlinear Effect) [4-6].

Recently we reported a study of the aligning dye effect on the nematic director for various values of the exciting Ar^+ laser power and we have shown the presence of a threshold effect [7]. In order to clarify the role of surface structure we investigated the dye aligning effect when the exciting Ar^+ laser beam is impinging through the isotropic surface (case 1) or through the rubbed one (case 2). A significant asymmetry has been observed [8].

In this paper we derive the rotating angle of the nematic director on the isotropic surface as function of the irradiation time in these two cases. By taking into account the different initial anchoring strength and the attenuation of the Ar^+ laser beam (due to dye molecules) we explain the asymmetric optical response. We conclude that the irradiation of the sample through the isotropic surface generates a fast rotating of the nematic director.

EXPERIMENTAL RESULTS

The experiments were realized using a standard sandwich glass cell filled with a mixture of NLC 4'-n-pentyl-4-cyanobiphenyl (5CB, clearing point T = 35.2°C) and azo-dye methyl red (MR) as a dopant (weight concentration 1%). Their structural formulas are given in Ref. [7]. Methyl red is a photosensitive dye [9]; its intrinsic photochemical processes are: *trans-cis* isomerization and bleaching under the illumination in the absorption band. The visible absorption spectrum of 5CB+MR is reported in Ref. [3].

The thickness of the cell was approximately 19 μm (obtained by Mylar spacers). The inner surfaces of the glasses composing the cell were covered with a thin polymeric film (for details, see Ref. [7]). One of the surfaces was rubbed in one direction provided a strong uniaxial anchoring of NLC molecules on the surface. This oriented surface (also named passive surface, S_{pas}) imposed the planar orientation of the nematic director for the whole cell in the initial state. The other surface (control surface, S_{con}) is quasi-isotropic.

The experimental setup is shown in Fig. 1. The cell was placed normally to the exciting Ar^+ laser beam (wavelength λ = 476.5 nm; power P_{exc} = 15 mW). The polarization of the excited beam was established by the polaryzer P, the electric vector being horizontal.

The probe He-Ne laser beam (wavelength λ = 633 nm; power P_{test} = 1 mW) was focused by a lens L in the irradiated area.

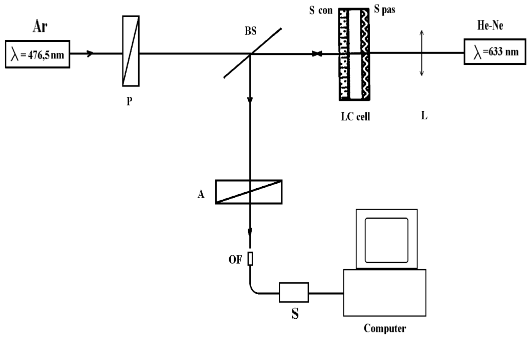

Fig. 1 - Experimental set-up

The two laser beams must be superposed in the same area of the NLC cell but the testing one must be smaller. The polarization of the He-Ne beam was vertical and the cell was placed with the rubbing direction parallel with the electric vector of the probe beam. After having passed through the cell the probe beam was deviated by a beam-splitter (BS) and went through the analyzer A. The transmission direction of the analyzer was horizontal. The probe beam intensity measurement was performed by an optical fiber (OF) connected with an “Ocean Optics” spectrometer S2000 (S) and a computer.

We have recorded the intensity of the transmitted probe beam as a function of the irradiating time in both cases (Fig. 2). As the exciting laser radiation enters the rubbed polymer surface the intensity of transmitted probe beam increases slowly as a function of the irradiation time and its values are smaller than those from case when the exciting laser beam enters the quasi-isotropic surface.

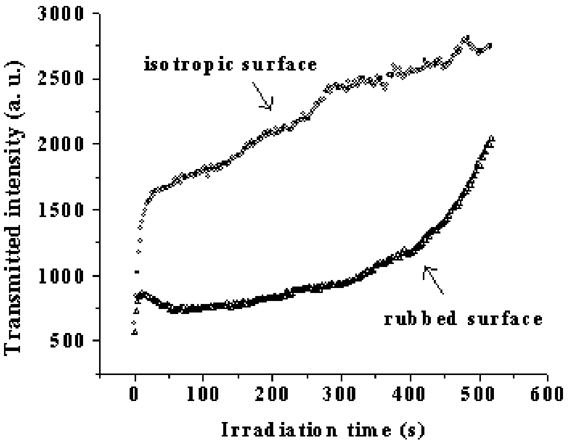

Fig. 2 - Transmitted intensity versus irradiation time.

EXPLANATION OF THE ASYMMETRIC RESPONSE

The asymmetric response, namely the differences between experimental optical signal when the exciting laser radiation is impinging through the quasi-isotropic surface or through the oriented one, could be explained by the different nematic director configurations during irradiation.

In case 1 the reorientation of NLC molecules occurs in the NLC bulk from the quasi-isotropic surface to the oriented one. The result is a twisted configuration of the nematic director from the laser - induced orientation to the orientation imposed by the easy axis of the rubbed polymeric film. The linearly polarized He-Ne probe beam having the electric vector $\vec{E}_{test}$ parallel to the rubbing direction of the oriented surface is impinging through it one. Let I_i the intensity of the He-Ne laser beam. The electric vector follows the nematic director through the whole cell (Mauguin limit [10] is fulfilled in our experiments) and rotates in the plane of the LC cell with an angle θ_1 (passing through the reoriented DDNLC cell). After the reflection on BS this laser beam has a new direction of polarization making the angle $\tilde{\theta}_1$ with the vertical direction. The relation gives this angle:

$$tg\tilde{\theta}_1 = \frac{r_{\parallel}}{r_{\perp}} tg\theta_1 \quad (1)$$

Here $r_{\perp}$ and $r_{\parallel}$ are the Fresnel coefficients corresponding to the transversal electric and transversal magnetic cases.

The intensity of the reflected beam is:

$$I_R(\theta_1) = I_i(r_{\parallel}^{\ 2} \sin^2\theta_1 + r_{\perp}^{\ 2} \cos^2\theta_1) \quad (2)$$

The analyzer A (with a horizontal direction of transmission) allows only the horizontal component of the reflected wave to pass:

$$I(\theta_1) = I_R(\theta_1)\sin^2\tilde{\theta}_1 \quad (3)$$

By using the Eqs. (1-3) we obtain the transmitted intensity by the analyzer at the irradiation moment t:

$$I_{tr1}(t) = I_i r_{\parallel}^{\ 2} \sin^2\theta_1(t) \quad (4)$$

In case 2, as the Ar$^+$ laser beam enter the rubbed surface, a new local order of the nematic molecules appears, in planes parallel to the surfaces. Due to the presence of the dye molecules, the NLC molecules tend to orient themselves parallel to the Ar$^+$ laser beam polarization. Starting the irradiation, the nematic order imposed by the bulk director is not the same on both surfaces; the anchoring energy is larger on the rubbed one thus preventing a significant angular gliding of the nematic director in this plane. On the other surface the anchoring energy does not impose an easy axis (degeneracy) and the nematic director follows the bulk orientation. So, this is again a twisted nematic configuration as in case 1 but the probe beam impinges the surface at a certain angle θ_2 between the polarization direction (vertical) of He-Ne laser beam and the nematic director on the quasi-isotropic surface.

Let I_i the intensity of the He-Ne laser beam, linearly polarized at vertical direction, incident on the cell through the quasi-isotropic surface. We denote by $\theta_2(t)$ the rotating angle of the nematic director on this surface at irradiation time *t*. At the entrance in the sample, the He-Ne laser beam can be decomposed in two waves: the extraordinary wave (e. w.), polarized parallel with the nematic director on this entrant surface, having the intensity $I_i \cos^2 \theta_2(t)$ and the ordinary wave (o. w.), polarized orthogonal on the local nematic director, having the intensity $I_i \sin^2 \theta_2(t)$.

At the exit from the sample (at the rubbed polymer surface), the e. w. is vertically polarized while the o. w. is horizontally polarized. After the reflection on BS the e. w. is blocked off by the analyzer (its transmission direction is horizontal). The o. w. is reflected on BS, its intensity becomes $I_i r_\parallel{}^2 \sin^2 \theta_2(t)$ and it passes through the analyzer without attenuation. The transmitted intensity in this case is:

$$I_{tr2}(t) = I_i r_\parallel{}^2 \sin^2 \theta_2(t) \tag{5}$$

We remark that the relations (4) and (5) have the same form.

The optical fiber receives only a fraction $k < 1$ from the intensity transmitted by the analyzer. It also receives the dark signal I_{dark}, which is obtained by switching off the He-Ne laser beam.

The intensity $I_{tr}(t)$ received by the optical fiber at the irradiation time t, in both cases, has the following form:

$$I_{tr}(t) = kI_i r_\parallel{}^2 \sin^2 \theta(t) + I_{dark} \tag{6}$$

The presence of the factor kI_i in the relation (6) imposes a supplementary measurement to be made before irradiating the sample. Now the He-Ne laser beam propagates through the sample as an extraordinary wave. After the reflection on BS, its intensity becomes $I_i r_\perp{}^2$. The recorded intensity I_γ (when the analyzer is rotating by the angle γ with respect to the horizontal direction) is:

$$I_\gamma = kI_i r_\perp{}^2 \sin^2 \gamma + I_{dark} \tag{7}$$

By using the Eqs. (6) and (7), the time dependent rotating angle $\theta(t)$ can be calculated:

$$\sin^2 \theta(t) = \left(\frac{r_\perp}{r_\parallel} \right)^2 \frac{I_{tr}(t) - I_{dark}}{I_\gamma - I_{dark}} \sin^2 \gamma \tag{8}$$

The measurements were performed, for both cases, under the same conditions (power of the Ar$^+$ laser beam, γ, I_γ, I_{dark}), but the intensity of the transmitted probe beam at irradiation moment t in case 2 is smaller than that in case 1, $I_{tr2}(t) < I_{tr1}(t)$. The relation (8) allows the computation of $\theta_1(t)$ and $\theta_2(t)$ (see Fig.3).

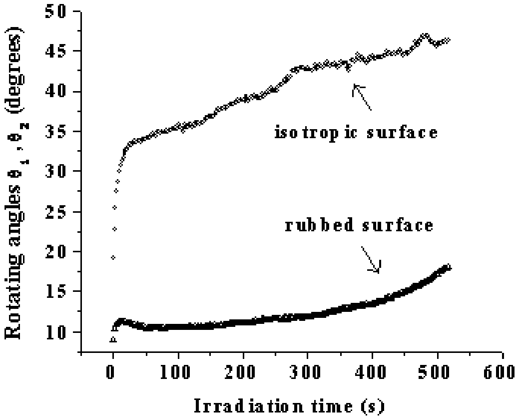

Fig. 3 - Rotating angles $\theta_1(t)$ and $\theta_2(t)$ versus irradiation time.

We conclude that the rotating angle of the nematic director within the quasi-isotropic surface in the case when the exciting laser beam enters this surface is greater than the corresponding value obtained by irradiating the rubbed surface, $\theta_1(t) > \theta_2(t)$.

The Ar$^+$ laser beam is attenuated during its propagation through the cell filled with 5CB+MR due to the MR molecules, which absorb this radiation. The anchoring energy on the rubbed surface is large preventing a rotation of the nematic director in this plane. On the quasi-isotropic surface, the anchoring energy does not impose an easy axis and the nematic director rotates under laser irradiation.

In case 1, the intensity of Ar$^+$ laser beam has a maximum on the quasi-isotropic surface (entrance surface) and a minimum on the rubbed surface (exit surface). In case 2, the maximum intensity of the pump beam is on the rubbed surface (which is the entrance surface in this case) and the minimum intensity is on the quasi-isotropic surface. The different values of the pump beam intensity on the quasi-isotropic surface in these two cases are responsible for the asymmetric response of sample.

CONCLUSIONS

The values of the transmitted probe light intensity passing through a DDNLC cell in case 2 (the exciting laser beam enters the oriented surface) are smaller than those from case 1 (the exciting laser beam enters the quasi-isotropic surface) and the time dependencies are different (asymmetric response). As a consequence the calculated values of the rotating angle of the nematic director within the quasi-isotropic surface show an asymmetry too. The explanation of this effect is based on: different nematic director configuration during irradiation, absorption of the Ar^+ laser beam in the DDNLC sample and different anchoring strength on the two surfaces.

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