

Photomanipulation of the anchoring strength of a photochromic nematic liquid crystal

G. Barbero,¹ L. R. Evangelista,^{1,2} and L. Komitov³

¹*Dipartimento di Fisica del Politecnico and INFM, Corso Duca degli Abruzzi, 24-10129 Torino, Italy*

²*Departamento de Física, Universidade Estadual de Maringá, Avenida Colombo 5790, 87020-900 Maringá, PR, Brazil*

³*Department of Microelectronics and Nanoscience, Chalmers University of Technology, S-41296 Gothenburg, Sweden*

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The behavior of the Fréedericksz threshold voltage of a planarly oriented sample filled with liquid crystalline azobenzene material possessing a nematic phase is investigated under UV illumination. It is found that the anchoring strength strongly decreases with the UV-exposure time due to the photoisomerization process that takes place in the sample. The changes in the anchoring strength are due to a selective adsorption of *cis* isomers at the surface. We show that the experimental data can be explained by a model that takes into account the changes in the molecular shape and in the net molecular dipole moment due to the *cis-trans* isomerization. We obtain a good agreement between the predictions of the model and the experimental data.

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I. INTRODUCTION

Liquid crystals (LCs) are organic materials with strong anisotropic physical properties. Due to their peculiar optical properties (transparency, homogeneity, birefringence, etc.) they are very attractive materials for a great variety of optical applications. The most known are the liquid crystal displays (LCDs) that over the last 20–25 years have developed from a simple niche products to strategic components.

Liquid crystal displays and devices consist, in general, of two solid substrates that (one or both) are transparent. These substrates form a cavity, usually of a few microns, filled with a liquid crystal material. The inner substrates' surfaces are precoated with transparent electrodes covered by thin alignment film for achieving a desired orientation of the liquid crystal molecules via solid surface/liquid crystal interactions. The uniform alignment of liquid crystals is crucial for LC displays and other LC devices and, therefore, it is a subject of many fundamental and experimental studies. A general requirement to liquid crystal alignment is to be stable with the time and temperature.

The performance of the LCDs and devices depends on the material parameters (bulk characteristics) of the liquid crystal as well as on the surface/liquid crystal interactions (surface characteristics). In particular, the characteristic features of the anchoring of the liquid crystal are very important for the performance of the liquid crystal devices since the strength of the anchoring affects strongly their threshold characteristics. The anchoring of liquid crystal to the solid substrate is a result of a very delicate balance between a number of interactions [1]. As we recently have shown, the photoisomerization process in photochromic liquid crystals that takes place under light illumination could change this balance and, thus, result in changes in the anchoring of the liquid crystal [2,3]. In some cases, these changes may trigger in-plane or/and out-of-plane anchoring transitions.

The bulk properties of thermotropic liquid crystals depend on the molecular structure and on the molecular interactions. The liquid crystal surface properties that are of vital importance for the alignment of LCs and thus for the optical appearance and the operation of these devices, depend, in ad-

dition, on the solid surface-liquid crystal interactions. Hence, both bulk and surface physical properties of the liquid crystal can effectively be controlled by external factors that result in changes of the molecular structure.

As known, reversible changes in the molecular structure may take place due to photoisomerization [4–6]. Some organic materials, such as azobenzenes, which also may exhibit liquid crystalline properties, undergo a *trans-* to *cis* isomerization upon light illumination. The photoisomerization process, however, may result in changes of the net molecular dipole moment and it may or may not result in changes of the molecular shape [3]. As a consequence, changes of bulk and/or of surface liquid crystal properties may take place. The photoisomerization process might be also reversible, if there is no chemical reaction or material degradation under light illumination. Therefore, the photoisomerization of liquid crystal attracts the interest of many researchers since the physical (bulk and surface) properties of the photochromic liquid crystals reversibly can be controlled by light without any changes in their chemical content. Photoinduced changes of the liquid crystal physical properties, however, could directly (e.g., photoinduced phase transitions and anchoring transitions) or indirectly (e.g., electric field assisted photoinduced effects) lead to significant changes of the optical appearance of the liquid crystal devices. These changes could be very fast (of the order of 100 μ s or even less) or very slow (a couple of minutes or hours) depending on the origin of the photoinduced effect, photochromic material, and the conditions of light illumination. The photoinduced effects in liquid crystals are very attractive for applications in photonic devices.

In this paper, we present the experimental results concerning the behavior of the Fréedericksz threshold voltage as a function of the UV-exposure time, for a cell of conventional sandwich-type, consisting of two parallel glass plates, filled with a photosensitive nematic liquid crystal [4-hexyloxy-(4'-hexyl)azobenzene]. By means of a relation connecting the anchoring strength to the threshold voltages, it is possible to determine the behavior of the anchoring strength as a function of the UV-exposure time. These new data can be explained in the framework of the theoretical models pro-

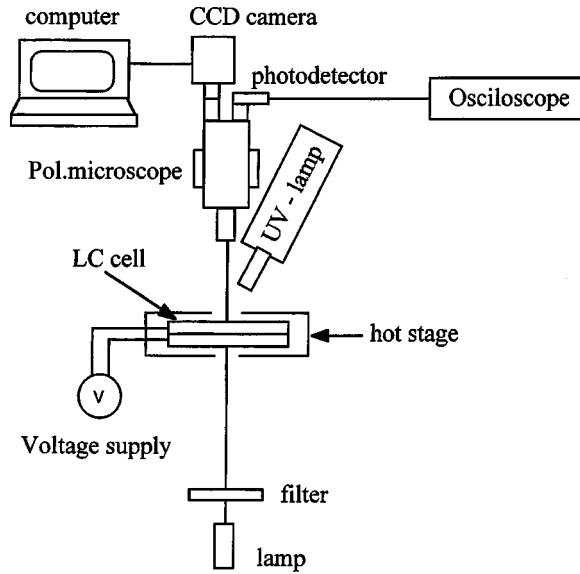


FIG. 1. Experimental setup.

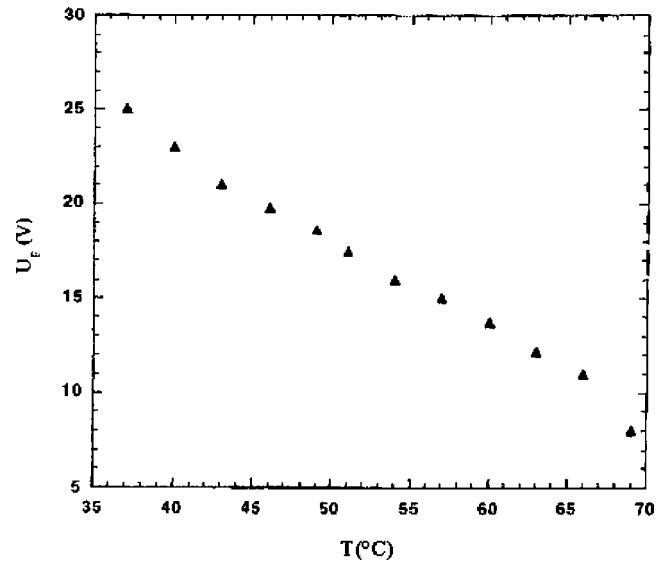
posed to describe the planar-homeotropic anchoring transition in azobenzene materials [7]. In the model presented in Ref. [7], it is considered that in the initial state all the molecules of the photosensitive material are in the *trans* state, being planarly oriented. With the increasing of the UV-exposure time the concentration of *cis* isomers increases, whereas the concentration of *trans* isomers decreases. The *cis* isomers are attracted to the solid substrate more than the *trans* isomers due to their larger electric dipole moment [3]. It follows that the surface density of the *cis* isomers increases with the illumination time. As discussed before, this process of selective adsorption of *cis* isomers onto the solid surface has drastic consequences on the threshold voltages and will be invoked here to explain the behavior of this quantity as a function of the illumination time.

Our paper is organized as follows. In Sec. II the experimental setup is described. In Sec. III we discuss the main experimental results. In Sec. IV we present the theoretical model and its connection with the experimental data. Some concluding remarks are drawn in Sec. V.

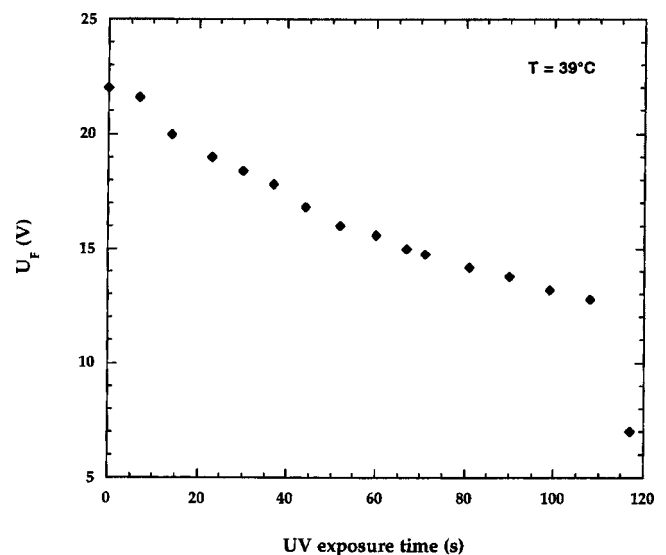
II. EXPERIMENT

The experimental cell used in this study is of conventional sandwich-type consisting of two glass substrates covered with transparent ITO (indium-tin oxide) electrodes precoated with a thin SiO_x layer. The SiO_x layer is deposited at normal incidence without any further treatment. The gap of the cell is fixed by Mylar spacers to be of about $10 \mu\text{m}$. The liquid crystal under study is a photosensitive nematic liquid crystal, 4-hexyloxy-(4'-hexyl)azobenzene.

The liquid crystal material has a small positive dielectric anisotropy ($\epsilon_a > 0$). The liquid crystal was introduced into the cell in the isotropic phase. After cooling the cell from the isotropic phase, the liquid crystal exhibited planar alignment with the preferred direction parallel to the material flow during the filling process. The cell was inserted in a Mettler F-52 hot stage in a setup shown schematically in Fig. 1. The

FIG. 2. Temperature dependence of the Fréedericksz threshold voltage U_F .

optical observations were performed by means of a polarizing microscope under crossed polarizers. The sample was protected from the light of the microscope lamp by yellow light filter. The sample was subjected to UV light for different periods of time. UV light was supplied by UV-curing units (Teklite) with an intensity of 70 mW cm^{-2} at the wavelength $\lambda = 365 \text{ nm}$. The illumination spot was about 1.5 cm in diameter, being with homogeneous distribution of the intensity in the cell area under investigation. Only one part of the sample was subjected to UV light. An ac voltage (at 1 kHz) was applied to the cell and the threshold voltage U_F for the Fréedericksz transition was measured as a function of the temperature and the UV-exposure time. The accuracy of the measurements of U_F was $\pm 0.5 \text{ V}$. This dependence is depicted in Figs. 2 and 3, respectively. As seen, U_F

FIG. 3. Dependence of the Fréedericksz voltage U_F on the exposure time of UV illumination.

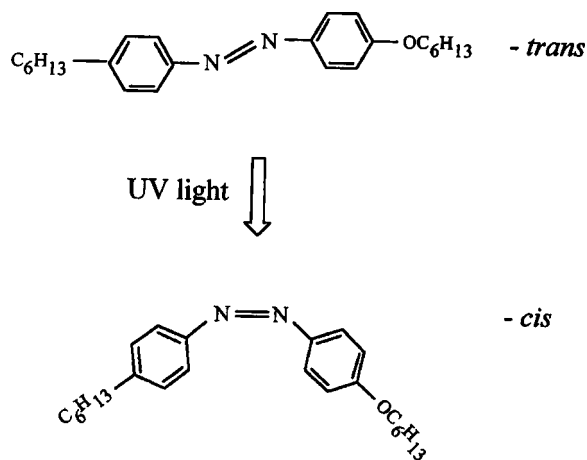


FIG. 4. Schematic presentation of photoisomerization of the 4,4'-disubstituted azobenzene nematic liquid crystal.

decreases with the temperature and the exposure time, thus implying that the anchoring strength undergoes changes.

III. DISCUSSION

Before UV illumination, U_F measured at 39°C was about 23 V. It was impossible to achieve any complete homeotropic alignment in the cell even at very high electric field near the electrical breakdown of one of the cells. There is always some residual birefringence left due to the presence of surface layers with liquid crystal molecules having tilted instead of homeotropic alignment. On approaching the clearing point, however, it was possible to obtain a complete transition from planar to homeotropic alignment by applying a high electric field. However, no temperature-induced anchoring transition from planar to homeotropic was found in this case. On the contrary, under UV illumination an anchoring transition from planar to homeotropic alignment was found to take place in the sample without applying an electric field. The mechanism of the light-induced alignment transition has been thoroughly studied [2,3] and could be shortly described as follows.

Upon UV illumination, the liquid crystal under study undergoes a *trans-cis* isomerization (Fig. 4). The concentration of the *cis*-isomer in the liquid crystal is increasing with the exposure time. Since the form of this isomer does not favor the liquid crystalline molecular order, due to its bent shape, the nematic state transforms into isotropic state above a critical concentration of the *cis* isomers in the liquid crystal volume [8]. Below this concentration, however, a transition from the initial planar to homeotropic alignment is observed to take place in a cell whose substrates are hydrophilic, as, for instance, the one covered by SiO_x . The *cis* isomer is generated under UV illumination in the volume as well as at the surface. However, the concentration of *cis* isomer at the surface increases much faster with the exposure time than the one in the volume since the *cis* isomer possesses a larger transverse dipole moment than the *trans* isomer that in turn favors the adsorption of *cis* isomer on the surface (selective adsorption) (see Fig. 5). Due to its form, however, the *cis* isomer anchored to the surface will affect the anchoring of

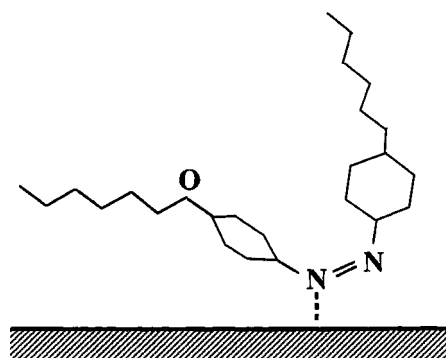


FIG. 5. Single *cis*-isomeric molecule adsorbed on the solid substrate.

the liquid crystal in a similar way as the surface-active agents do. Above a certain surface concentration, the *cis* isomer will promote a homeotropic alignment of the liquid crystal. Until the critical surface concentration of the *cis* isomer is reached, there is a continuous change of the anchoring strength with the UV-exposure time due to the selective adsorption process, as indicated by the dependence of U_F in Fig. 3. U_F was measured at 39°C , well below the clearing point (of about 30°C), as a function of the exposure time. As shown in Fig. 3, U_F successively diminishes with the exposure time and at about 2 min exposure, spontaneous transition from planar to homeotropic alignment takes place. In Fig. 6 is shown a sequence of photographs of the experimental cell after short UV illumination of the central part of the cell. As seen, no visible changes in the birefringence of the cell took place, thus indicating that there has been no detectable changes in the liquid crystal alignment due to the UV illumination [Fig. 6(a)]. When applying a voltage to the cell, the liquid crystal volume reorients first under the applied field (Frédéricisz transition) leaving the liquid crystal alignment in the subregion near the substrates to a large extent unaffected. The reorientation of the liquid crystal changes the birefringence of the cell [Fig. 6(b)]. However, increasing the voltage further, a transition to homeotropic alignment was found to take place in the illuminated region of the cell where the anchoring has been modified by the selective adsorption of *cis* isomer [Fig. 6(c)] whereas even much higher electric field is unable to result in such a transition in the unilluminated part of the cell at this temperature.

The field-assisted anchoring transition from planar to homeotropic seems to have potential for application in photonics. The recorded image can be visualized by applying an electric field. Depending on the exposure time, it would be possible to visualize images that will appear at different voltages.

IV. MODEL

Let us now interpret the above results concerning the dependence of U_F with the UV-exposure time in the framework of the model proposed in Ref. [7], which will be briefly reviewed. As stated above, we assume that in the initial state of the photosensible material all the molecules are in the *trans* state, and the planar orientation is achieved by means

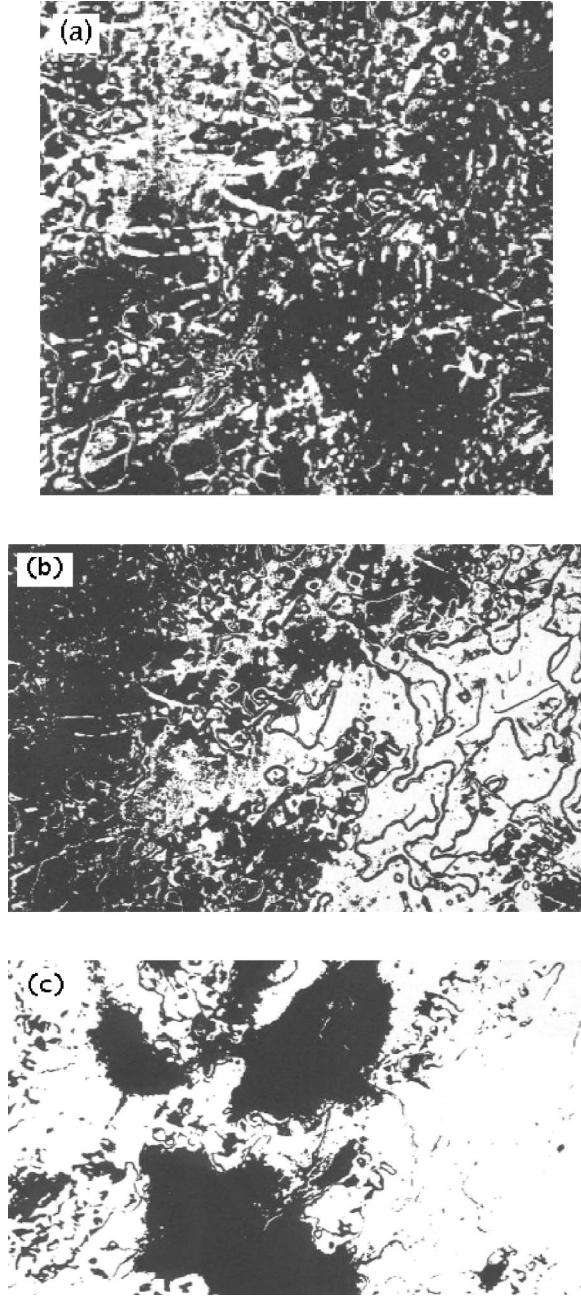


FIG. 6. Field-assisted transition from planar to homeotropic after UV illumination with UV light for short time. (a) No applied field ($U=0$), (b) $U_1 > U_F$, and (c) $U_2 > U_1$.

of the used surface treatment. With the UV irradiation the concentration of *cis* isomers increases and the concentration of *trans* isomers decreases [7]. The *cis* isomers, having larger dipole moment, are more strongly attracted by the cell surfaces. This effect can be easily understood by evaluating the interaction energy of electrostatic origin of a dipole \mathbf{p} with an isotropic substrate.

Let \mathbf{p} be the dipole moment of an isomer, at a distance r_0 from the substrate. We indicate by β the angle made by \mathbf{p} with the geometrical normal to the substrate, oriented towards the dipole. We use a Cartesian reference frame having the z axis coinciding with the normal to the substrate, and the

(x, z) plane containing the dipole \mathbf{p} . In this framework, $\mathbf{p} = p\mathbf{u}$, where $\mathbf{u} = (\sin \beta, 0, \cos \beta)$. According to the elementary electrostatics [9] the image of \mathbf{p} in the dielectric substrate, of dielectric constant ϵ , is

$$\mathbf{p}' = p \frac{\epsilon_0 - \epsilon}{\epsilon_0 + \epsilon} \mathbf{u}', \quad (1)$$

where $\mathbf{u}' = (\sin \beta, 0, -\cos \beta)$. The electrostatic interaction energy between the dipole \mathbf{p} and its image \mathbf{p}' in the substrate is

$$V = \frac{1}{4\pi\epsilon_0} \frac{(\mathbf{p} \cdot \mathbf{p}') - 3(\mathbf{p} \cdot \mathbf{z})(\mathbf{p}' \cdot \mathbf{z})}{(2r_0)^3}, \quad (2)$$

where \mathbf{z} is the unit vector along the z axis. By substituting Eq. (1) into Eq. (2), simple calculations give

$$V = -\frac{1}{4\pi\epsilon_0} \left(\frac{\epsilon - \epsilon_0}{\epsilon + \epsilon_0} \right) \frac{p^2}{(2r_0)^3} (1 + \cos^2 \beta). \quad (3)$$

Equation (3) shows that the interaction energy of the dipole with the substrate is proportional to p^2/r_0^3 , and depends on the angle formed by the dipole with the geometrical normal to the substrate, β . By assuming that r_0 is the same for the *trans* and *cis* isomers, we conclude that the larger is the dipole moment, the larger is the adsorption energy. Since the dipole moment of the *cis* isomer is larger than the one of the *trans* isomers, we expect that the surface concentration of *cis* isomer increases with the irradiation time.

A selective adsorption of *cis* isomers has an important effect on the orientation induced by the surface on the nematic liquid crystal. To analyze this effect, let us assume, for simplicity, that the bulk concentration of *cis* isomers remains small. This is equivalent to assume that the bulk nematic properties of the photosensitive liquid crystal do not change very much. This is not really the case, since the shift in the nematic-isotropic critical temperature is of several degrees, but this simplifying hypothesis allows one to understand in a simple way the effect of the *cis* isomers on the induced alignment. In this framework, the initial planar alignment is due to a molecular steric interaction “side to side,” where the electrical dipole of the *trans* isomer is parallel to the substrate. When a *cis* isomer is adsorbed at the surface, its molecular dipole, for steric reasons, is normal to the substrate. In this case the *cis* isomer gives rise to a structure having the shape of an asymmetric V, shown in Fig. 5. On an average, if the substrate is homogeneous, the symmetry axis of these objects of V shape is normal to the substrate, and the induced orientation on the liquid crystal is homeotropic, i.e., the nematic molecules are normal to the substrate. To connect the anchoring energy of the interface with the surface concentration of *cis* and *trans* isomers, we can proceed in the following manner. If the substrate is homogeneous, the surplus of energy due to the presence of the substrate is of the kind $f_s = f_s(\mathbf{n} \cdot \mathbf{k})$, where \mathbf{n} is the surface director and \mathbf{k} the geometrical normal to the substrate, supposed to be flat. Since \mathbf{n} is equivalent to $-\mathbf{n}$, $f_s = f_s[(\mathbf{n} \cdot \mathbf{k})^2]$. The surface orientation \mathbf{n}_e minimizing f_s is called “easy direction.” In the initial

state where *cis* isomers are absent, \mathbf{n}_e is parallel to the substrate ($\mathbf{n}_e \perp \mathbf{k}$). In the opposite limit, where only *cis* isomers are present at the surface, $\mathbf{n}_e \parallel \mathbf{k}$. In a first approximation, in which the easy directions are only the planar and homeotropic ones, f_S can be written in the Rapini-Papoular form [10]

$$f_S = \frac{1}{2} W \cos^2 \theta, \quad (4)$$

where θ is the angle between the normal and the surface nematic director, and W is the anchoring strength. The anchoring strength has two contributions: one coming from the *trans* isomers and another one coming from the *cis* isomers. It can then be defined as

$$W = c_{s,trans} W_t - c_{s,cis} W_c, \quad (5)$$

where $c_{s,trans}$ and $c_{s,cis}$ are the concentrations of *trans* and *cis* isomers at the surface, respectively. The structure of Eq. (5) is connected with the fact that the molecules in *trans* form favor a planar alignment, whereas the ones in the *cis* form induce homeotropic alignment, as discussed above. The quantities entering in Eq. (5) can be understood in the following manner. In the initial state (no UV illumination), all the molecules are in the *trans* state, i.e., $c_{s,cis} = 0$ and $c_{s,trans} = 1$. Therefore, in the initial state, the anchoring energy is connected with the presence of only *trans* isomers at the surface (in this case $W = W_t$); as the UV-exposure time increases, $c_{s,cis}$ also increases, tending to diminish W . W_c is then the anchoring strength of a nematic sample having only *cis* isomers adsorbed at the surface. By means of simple arguments it is possible to estimate the ratio W_c/W_t , if the interaction responsible for the surface orientation is mainly of steric origin. Let E be the interaction energy between a nematic molecule and an adsorbed surface molecule. In the case of planar orientation a nematic molecule interacts with one adsorbed molecule, and the involved energy is E . In the homeotropic orientation, induced by *cis* isomers, the number of first neighbors is, in average, four. Consequently, the energy for nematic molecule is $4E$. It follows that $W_c/W_t \sim 4$. Of course other interactions contribute to the surface energy, but they are usually negligible with respect to the hard core interaction considered in the steric interaction discussed above.

Let us consider now the influence of the UV irradiation time on the anchoring energy. According to the model proposed in Ref. [7], the density of *cis* molecules at the surface is given by

$$c_{s,cis} = \left[1 + \frac{x + (1-x)e^{-t/\tau}}{(1-x)(1 - e^{-t/\tau})} e^{-\Delta v} \right]^{-1}, \quad (6)$$

where τ is a characteristic time and x is a parameter that controls the fraction of *trans/cis* isomers after the illumination, because some kind of recombination has to be considered [11]. An estimation of x can be done by measuring the shift of the critical nematic-isotropic phase transition, as reported at the end of this section. Furthermore, $\Delta v = (V_{trans} - V_{cis})/k_B T$ is the difference between the adsorption ener-

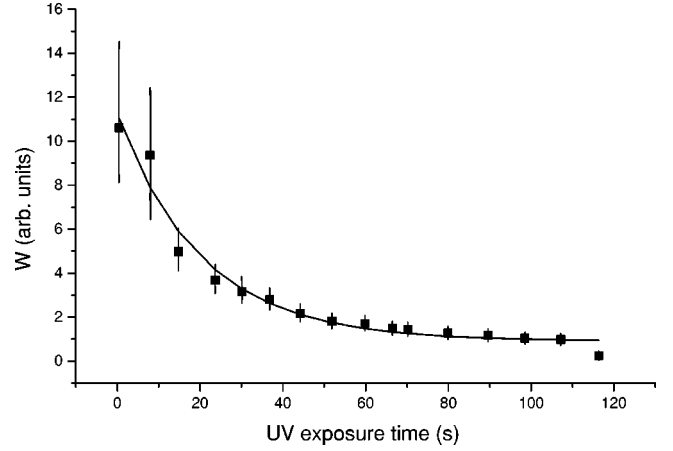


FIG. 7. Dependence of the anchoring strength W on the exposure time of UV illumination. The squares (with error bars) represent the data for W obtained from the data of Fig. 3. The solid line represents the theoretical curve.

gies of *trans* ($V_{trans} < 0$) and *cis* ($V_{cis} < 0$) molecules at the surface (in $k_B T$ units where k_B is the Boltzmann constant and T the absolute temperature). As a consequence of the assumption that at the initial time only *trans* molecules are adsorbed at the surface, we have $c_{s,trans} = 1 - c_{s,cis}$. Equations (5) and (6) are the theoretical tool to analyze the experimental data.

In Fig. 3 the Fréedericksz threshold voltages U_F are shown as a function of the UV-exposure time.

In order to obtain the anchoring strength as a function of the illumination time, we use the Rapini-Papoular expression for the anchoring strength as a function of the threshold voltages [1], which can be written as

$$W = \frac{K}{2d} \left(\frac{\pi U_F}{2 U_\infty} \right) \tan \left(\frac{\pi U_F}{2 U_\infty} \right), \quad (7)$$

where K is the elastic constant and d is the thickness of the sample. Furthermore, in Eq. (7) U_∞ is the threshold voltage for the case of strong anchoring at the surface. It is given by Ref. [1],

$$U_\infty = \pi \sqrt{\frac{K}{\epsilon_a}}, \quad (8)$$

where ϵ_a is the dielectric anisotropy of the sample. Note that W is actually measured in $K/2d$ and the critical voltage in U_∞ .

In Fig. 7 the values of W as a function of the UV-exposure time are shown. They were obtained by applying Eq. (7) to the data of Fig. 3 and considering that U_F was determined with an experimental error of about 0.5 V. The solid line represents the values of the best fit of these data, obtained by using the theoretical expressions given by Eqs. (5) and (6).

The parameters obtained from the best fit are: $\Delta v \sim 0.3$, $\tau \sim 21.0$ s, $W_c/W_t \sim 3.8$, and $x \sim 0.85$.

The positive difference between the adsorption energies of *trans* and *cis* molecules at the surface, $\Delta v = (V_{trans} - V_{cis})/k_B T \approx 0.3$, indicates that there exists a selective ad-

sorption of *cis* molecules at the surface responsible for the decreasing of the anchoring energy. This value for Δv can be justified using the expression proposed above for the adsorption energy connected with the electrostatic energy of dipolar origin. In fact, according to Eq. (3) we have

$$\Delta v = \frac{V_{trans} - V_{cis}}{k_B T} = \frac{1}{4\pi\epsilon_0} \left(\frac{\epsilon - \epsilon_0}{\epsilon + \epsilon_0} \right) \frac{2p_{cis}^2 - p_{trans}^2}{(2r_0)^3 k_B T}, \quad (9)$$

where the factor 2 in front to p_{cis}^2 comes from the angular dependence of the adsorption energy of dipolar origin. By assuming $p_{cis} \sim 5$ D, $p_{trans} \sim 1$ D [12], $r_0 \sim 6$ Å, and considering a glass with a relative static dielectric constant of the order of 4, we obtain $\Delta v \sim 0.4$ that is in reasonable agreement with the value of the best fit.

The value of $W_c/W_t \sim 3.8$ confirms our prediction according to which the steric interaction determines the orientation induced by the adsorbed isomers on the liquid crystal discussed above. This result indicates that for long UV-exposure time the system can undergo an anchoring transition from planar to homeotropic. Such a possibility is present in our model, but we have focused our attention to a case in which this anchoring surface transition has not been observed, since the technique used by us to measure the anchoring energy works only if the threshold voltage for the Fréedericksz transition is different from zero. For this reason, according to our best fit, the anchoring energy remains always positive, indicating that the easy direction remains always the planar one.

Finally, the parameter controlling the possibility of recombination is $x = 0.85$. This value can be evaluated directly by measuring the shift of the nematic-isotropic critical temperature in the absence, $T_{NI}(0)$, and in the presence of a given irradiation time, $T_{NI}(\Delta t)$. With our experimental setup for $\Delta t \sim 40$ s the shift of T_{NI} , $\Delta T_{NI} = T_{NI}(0) - T_{NI}(40 \text{ s}) \sim 6$ K. By assuming that the scalar nematic order parameter of the photosensitive liquid crystal can be evaluated using the Maier-Saupe theory, x can be estimated as follows. Without UV irradiation $T_{NI}(0) = 341.16$ K and with irradiation $T_{NI}(40 \text{ s}) = 338.16$ K. According to Maier-Saupe theory, $T_{NI}(0) = 0.22019v/k_B$, where v is a molecular parameter [9]. We define a reduced critical temperature by means of the relation $T_R = T_{NI}v/k_B$, and obtain $T_R(0) = T_{NI}(0)v/k_B$ and

$T_R(40) = T_{NI}(40)v/k_B = 0.22019[T_{NI}(40)/T_{NI}(0)] \sim 0.21$. According to the model proposed in Ref. [8] this reduced temperature corresponds to a density of isomers of the order of 0.1. Since $n_{cis}(\Delta t) = (1-x)[1 - \exp(-\Delta t/\tau)]$ [8], by assuming $\tau \sim 20$ s we obtain $x \sim 0.9$, which is comparable with the value of the best fit. From the discussion reported above it follows that only the characteristic time τ is a completely free fitting parameter, whereas the other parameters entering in the model can easily be connected with phenomena conditioning the observed effect of photomanipulation of the anchoring energy.

The photoisomerization process affects also the bulk properties of the liquid crystal. The *cis* isomer of the nematic under study is not compatible with the liquid crystalline order and thus it will affect the nematic scalar order parameter S [8,13]. Whereas the changes in the order parameter due to the temperature arise from the increased disorder caused by thermal fluctuations, the changes due to the photoisomerization arise from the increased concentration of *cis* isomer. Work is in progress along these lines and will be published elsewhere.

V. CONCLUSIONS

In this paper, we have presented an experimental technique to manipulate the anchoring energy of a photochromic nematic liquid crystal. We have shown how the UV-illumination process strongly affects the Fréedericksz threshold voltage and, consequently, the anchoring strength of a nematic liquid crystal. The experimental results are interpreted in the framework of theoretical models that take into account the changes in the molecular form and the molecular net dipole moment upon light illumination. The agreement between the models and the experimental data is rather good. Photomanipulation of the anchoring strength seems to have potential applications in photonics.

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