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Magnetic field-assisted liquid crystal alignment on a solid substrate: drift of easy orientation axis, anchoring energy and angular distribution of adsorbed liquid crystal molecules

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The anchoring properties of a film of anisotropically adsorbed liquid crystal (LC) molecules on a rigid substrate have been studied. The LC film was prepared by cooling it from the isotropic phase in the presence of a magnetic field parallel to the surface of the substrate. Relationship between the anchoring energy, easy axis direction and angular distribution of the adsorbed molecules, and changes in their angular distribution due to adsorption– desorption, were studied. The dependence of the anchoring energy on the duration and the temperature at which the LC film is annealed allowed an estimation of the activation energy of desorption of LC molecules on ITO surface, $\Delta E \approx 0.55$ eV. The results suggest that hydrogen bonds are responsible for the adsorption of LC molecules on the substrate.

1. Introduction

The traditional description of liquid crystal (LC) alignment on a solid surface suggests that an easy orientation axis results from a direct anisotropic interaction between a physically anisotropic surface and LC molecules, the translation mobility of which does not differ much from the mobility of the molecules in bulk LC. Since the first observation of a memory effect [1-3], it became clear that adsorption of LC molecules on the aligning surface can yield an anisotropic layer which itself aligns the LC. Subsequent studies confirmed that the layer formed of adsorbed molecules plays an important role in orienting the bulk LC that comes in contact with it. For instance, when a cell filled with LC is cooled from the isotropic phase to the nematic phase in the presence of a magnetic field, homogeneous LC orientation can be attained without surface treatment [2-4]. Light-induced adsorption of dye molecules on an isotropic polymer surface from the LC bulk also causes homogeneous alignment of the LC [5]. Investigations of the drift of the easy axis (or the "gliding effect" [4-8]) with time and temperature in electric, magnetic and optical fields showed that this effect is caused by the processes of adsorption and

Macroscopic characteristics of the alignment caused by the adsorbed layer depend on the angular distribution of the adsorbed molecules. Recently, Romanenko *et al.* developed a theory that describes director gliding in terms of changes in the angular distribution function of the adsorbed molecules and rotation of this function due to a bulk torque [9, 10]. This theory connects the macroscopic characteristics of the gliding with microscopic characteristics of the adsorbed LC molecules. For instance, the maximum of the angular distribution function of adsorbed elongated molecules specifies the direction of the easy axis, \mathbf{e} , and the width of the distribution is related to the anchoring energy, W.

The drift of the easy axis due to adsorptiondesorption of LC molecules on traditional polymer aligning surfaces is accompanied by a collective reorientation of flexible polymer fragments and LC molecules [11–13]. The latter process results in the drift of the easy axis along with AD processes. Both mechanisms of gliding are described by the same equations (see Romanenko *et al.* [9, 10]). The only



desorption of LC molecules on the aligning surface. The layer of adsorbed molecules should be considered as a mobile system wherein an active exchange between "bulk" and "surface" molecules occurs via the processes of adsorption and desorption (AD processes) of the LC and dopant molecules.

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difference between them is in the physical meaning of these equations. Therefore, it is difficult to differentiate between these two alternative mechanisms.

In this paper, we report experimental studies of the anchoring properties of a layer of anisotropically adsorbed LC molecules on a rigid substrate formed by cooling the LC from its isotropic phase in the presence of a magnetic field. The formation of reliable adsorbed layers of LC on rigid ITO coated surfaces aligned by a strong magnetic field allowed us to avoid complicating factors such as possible rearrangement of the aligning surface and inhomogeneities in the angular distribution of adsorbed molecules. The observed drift of the easy axis with time in nematic phase and upon annealing the LC cells in the isotropic phase, as well the changes in the anchoring energy after annealing, are explained in the framework of Reshetnyak's model [9, 10]. It allows us to establish the dependence of the angular distribution of adsorbed molecules, and the changes in it due to AD processes on the LC anchoring energy and the direction of easy axis. Experimental data reveal important information about the interaction between LC molecules and the substrate surface.

2. Experimental

A layer of adsorbed molecules of pentylcyanobiphenyl (5CB, clearing temperature, T_{NI} =35.4°C) was formed on ITO glass substrates. An empty symmetric cell, 5 µm thick, was heated to 70°C and placed between magnet poles so that the lines of magnetic field, $H \approx 0.8 \text{ T}$, were in the plane of the cell. Then the cell was filled with 5CB that was preheated to 70°C. The direction of filling was kept either parallel or perpendicular to H. After filling, the LC was cooled down to its nematic phase in 15 min, the field reduced to zero, and the cell removed from the magnet. A satisfactory uniform planar orientation of LC was observed in these cells. The direction of the alignment was along the direction of H and did not depend on the filling direction. This established that the magnetic field strength was sufficient to form a layer of adsorbed and aligned LC molecules.

To determine the anchoring energy of the adsorbed aligning layer, the symmetric cell was disassembled and the substrates used as a test surfaces in combined cells (figure 1) in which an ITO glass covered with a film of rubbed polyimide, PI2555 from Nissan, served as the reference substrate. This reference substrate provided negligibly small pretilt and strong planar alignment of 5CB. Therefore, the direction of the easy axis on the reference surface did not change at room temperature. The combined cells were made in a way such that the angle, θ_0 , between the rubbing direction of the reference substrate and the alignment direction on the test



Figure 1. Schematic representation of the hybrid test cell used to measure the twist angle.

substrate, was 45° . Since the thickness of the combined cell, $L=10\,\mu\text{m}$, was larger than that of the original symmetric cell and some of the LC was lost during its disassembly, additional 5CB was filled in the combined cells. Observations under a polarizing microscope showed a homogeneous twist structure confirming, for example, that the anchoring energy on the test surface was high enough to support a twist deformation in the cell.

The Mauguin regime for the propagation of light was valid in our case. Therefore, the probe beam followed the rotation of the director in the cell and its polarization was parallel to the director on the test surface. It allowed us to measure the twist angle in the cell, φ_{test} , by measuring the angle between polarizer and analyser [14].

Twist angle φ_{test} gives the orientation of the director on the test substrate. We obtained the value $\varphi_{\text{test}}=42^{\circ}$ at room temperature and observed a slow decrease of φ_{test} . This result is mainly caused by the drift of the easy axis toward the easy axis of the reference surface [8–10, 12]. However, a rapid change in φ_{test} was observed when the cell was annealed at a temperature $T > T_{NI}$ (T=65-120°C, figure 2). The annealing time, t_{ann} , was 2 h and it was much longer than the heating, t_{heat} , and cooling, t_{cool} , times of the cell (1–2 min). Therefore, possible changes of the twist angle during heating and cooling were negligible.

The decrease of the twist angle was faster with higher annealing temperature (figure 2). To check if this decrease is caused by a drift of the easy axis or the decrease in the anchoring energy on the test surface, we annealed the 45°-twist combined cell in the isotropic phase ($T=90^{\circ}$ C) for 22 h. After cooling to room temperature, this cell revealed significant decrease of the twist angle ($\varphi_{test}=22^{\circ}$). We disassembled the cell and rotated the test substrate by 45° toward the rubbing axis



Figure 2. Experimental (dots) and theoretical (solid) dependence of the twist angle on thermal annealing time for annealing temperatures of 65° C (top curve), 80° C, 85° C, 90° C, 95° C, 100° C and 120° C (lowest curve).

on the reference substrate. In this geometry, the cell demonstrated a perfect planar alignment with zero twist that denotes no change of the easy axis on the test surface. Thus, the decrease of the angle ϕ_{test} after annealing is caused by a decrease of the anchoring energy on the test surface.

3. Theoretical model and discussion

Below, we describe a model which yields the direction of the easy axis and the value of the anchoring energy from the angular distribution function of the molecules adsorbed on the substrate. The changes in the angular distribution of the adsorbed molecules arise from the AD processes described above.

First, consider the formation of the adsorbed layer in a symmetric cell. During cooling, just after the transition from the isotropic phase to the mesophase, molecules in the bulk of LC are preferably oriented along the direction determined by **H**. In this case, the adsorbed molecules, which mimic the angular distribution of the molecules in the bulk, cause the easy axis, \mathbf{e}_{test} , to be parallel to **H**. We presume that the angular distribution of the long axes of the adsorbed molecules, $F_{S}(\varphi_{s})$, is determined as a projection of the bulk distribution, $F_{V}(\Omega_{V})$, onto the surface:

$$F_S(\varphi_S) = \int_0^{\pi} F_V(\Omega_V) \sin \theta_V \, d\theta_V, \qquad (1)$$

where θ_V is the polar angle between the long axis of molecule and the normal to the substrate, $\varphi_S = \varphi_V$ is the azimuth angle between the long axis of molecules and the easy axis, \mathbf{e}_{test} , Ω_V is the solid angle (θ_V , φ_V) in the polar reference frame. The angular distribution function of the LC molecules in the bulk in the Maier–Saupe approach is given by [15]:

$$F_V(\Omega_V) = \frac{1}{N} \exp\left\{\beta S \frac{3\left[\sin^2 \theta_V \cos^2 \varphi_V - 1\right]}{2}\right\}, \quad (2)$$

where *S* is the nematic order parameter in the bulk, *N* is a normalization constant, $\beta = \frac{\alpha}{k_B T}$, and α is the amplitude of the intermolecular interaction potential.

The interaction of the "bulk" molecules with the adsorbed molecules is proportional to the concentration of the adsorbed molecules, N_S , molecular anchoring coefficient, w_0 and it depends on angular distributions

of the "bulk" and the "surface" molecules:

$$W = \frac{1}{2} w_0 N_S \int_{-\pi/2}^{\pi/2} d\phi_V \int_{-\pi/2}^{\pi/2} d\phi_S F_S(\phi_S) \sin^2(\phi_V - \phi_S) \int_{0}^{\pi} F_V(\Omega_V) \sin^3 \theta_V \, d\theta_V.$$
(3)

The distribution $F_S(\varphi_s)$ is a *symmetric* function. In this case, equation (3) is reduced to the traditional form of Rapini–Popular surface potential [16],

$$W_{RP} = \frac{1}{2} W_0 \sin^2(\mathbf{e}_{\text{test}} \mathbf{n}), \qquad (4)$$

where the anchoring energy, W_0 , is expressed through the concentration of the adsorbed molecules, N_S , molecular anchoring coefficient, w_0 and the parameter λ as: $W_0 = w_0 N_S \lambda$ with

$$\lambda = \int_{-\pi/2}^{\pi/2} \cos(2\varphi_V) d\varphi_V \int_{0}^{\pi} F_V(\Omega_V) \sin^3 \theta_V d\theta_V \int_{-\pi/2}^{\pi/2} F_S(\varphi_S) \cos(2\varphi_S) d\varphi_S.$$
(5)

Here, the parameter λ characterizes the anisotropy of the angular distribution function of adsorbed molecules. Since the bulk distribution function, $F_V(\Omega_V)$, is determined by the order parameter, *S*, [see equation (2)], there is a one to one correspondence between the parameters λ and *S*. The dependence $\lambda(S)$ is presented in figure 3. One can see that $\lambda=1$ when S=1 and, for typical value of S=0.6 at room temperature, $\lambda\approx0.4$. The surface concentration N_s is described by the kinetic equation that takes into account the probability of adsorption and desorption of molecules and their angular distribution on the surface and in the bulk:

$$\frac{d(N_S F_S(\varphi_S))}{dt} = A_+ N_{VS} \int_0^{\pi} F_V(\Omega_V) \sin \theta_V \, d\theta_V - A_- N_S F_S(\varphi_S), \, (6)$$

where A_+ and A_- are the probabilities of the adsorption (A_+) and the desorption (A_-) of LC molecules per unit time, $N_{VS}=aN$, and *a* is the intermolecular distance in the bulk of LC perpendicular to the surfaces. In a stationary state achieved after cooling, $A_+N_{VS}=A_-N_S$.

During the second stage of the experiment, the substrate with adsorbed LC layer was tested using a hybrid test cell. Due to a finite value of anchoring energy, the director on the test substrate, \mathbf{n}_{test} , does not coincide with the easy axis, \mathbf{e}_{test} , and a free energy of LC in a cell per unit surface area is written as follows:

$$F = \frac{1}{2} \int_{0}^{L} dz K_{22} \left[\frac{\partial \varphi(z, t)}{\partial z} \right]^{2} + W + W_{ref}, \qquad (7)$$

where L is thickness of the cell, K_{22} is the Frank's twist elastic constant, φ is the direction of the director, W is the anchoring energy on the test substrate (equation (3))



Figure 3. Dependence of parameter λ on the nematic order parameter S.

and W_{ref} is the anchoring energy on the reference substrate.

The anchoring energy on the reference surface is suggested to be infinitively strong. Therefore, the boundary conditions for the director on the reference and test surfaces are written as:

$$\left. \frac{\partial W(\varphi_{\text{test}})}{\partial \varphi_{\text{test}}} + K_{22} \frac{\partial \varphi(z)}{\partial z} \right|_{z=L} = 0,$$
(8)

where $\varphi_{\text{test}} \equiv \varphi_{\text{director}}(L)$.

Equation (8) provides a balance of the bulk and surface torques on the tested surface. Minimization of equation (7) with the boundary conditions of equation (8) results in a linear solution for the twist of the director in the direction perpendicular to the substrates of the cell:

$$\varphi(z) = \frac{\varphi_{\text{test}}}{L} z. \tag{9}$$

Taking into account equation (9), equation (8) can be used to write:

$$\varphi_{\text{test}} = -\frac{\partial W(\varphi_{\text{test}})}{\partial \varphi_{\text{test}}} \frac{L}{K_{22}}.$$
 (10)

In the case of the Rapini–Popular surface potential, equation (4), equation (10) reduces to the well known expression that describes the relationship between the anchoring energy and the twist of the director in the combined twist cell [14, 16]:

$$\xi \sin 2(\varphi_0 - \varphi_{\text{test}}) = 2\varphi_{\text{test}}, \tag{11}$$

where $\xi = W_0 L/K_{22}$ is the anchoring parameter, $\varphi_0 = 45^\circ$ is the angle between \mathbf{e}_{ref} (parallel to the rubbing direction) and the easy axis of test surface which is along \mathbf{e}_{test} (or parallel to **H**). Substituting the experimental values $\varphi_{test} = 42^\circ$, $L = 10 \,\mu\text{m}$ and $K_{22} = 3 \times 10^{-12} \,\text{N}$ in equation (11), we obtain the value of the anchoring energy $W_0|_{t=0} = 4 \times 10^{-6} \,\text{Jm}^{-2}$ at room temperature. It should be noted that this value is unexpectedly high and is of the same order of magnitude as for photoaligning substrates [13].

Mismatching $\varphi_0 - \varphi_{test}$ between \mathbf{e}_{test} and \mathbf{n}_{test} causes the observed drift of the easy axis toward \mathbf{e}_{ref} in a nematic phase [7, 9–10]. Since the maximum of the angular distribution function of the LC molecules in the bulk is parallel to \mathbf{n}_{test} , a permanent exchange of molecules between the bulk and the surface due to AD processes leads to an enrichment of the surface with the adsorbed molecules preferably oriented parallel to \mathbf{n}_{test} . It results in a rotation of the easy axis, \mathbf{e}_{test} , and modification of the distribution of the adsorbed LC molecules

under the influence of a reorientation torque from the reference surface. Since the distribution function $F_S(\varphi_s)$ becomes non-symmetric during the drift of the easy axis [9, 10], the surface potential W is described by equation (2) and the time dependence of twist angle φ_{test} is determined by equation (10). The angle between \mathbf{n}_{test} and \mathbf{e}_{test} was small ($\approx 3^\circ$) due to relatively strong anchoring and consequently, the reorientation torque was weak. It resulted in very slow drift of the easy axis.

Much larger changes in φ_{test} were observed after heating the cell above T_{NI} , because, unlike a mesophase, the distribution of molecules above T_{NI} is isotropic and the adsorption of these molecules is also isotropic. At the same time, the angular distribution of desorbing molecules is anisotropic because these molecules belong to the anisotropic adsorbed layer formed in the mesophase. In this case, the exchange of molecules between the test surface and the bulk decreases the anisotropy of the adsorbed molecules that in turn, reduces the anchoring energy. Higher temperature speeds up the desorption process. It causes a decrease in the concentration of anisotropically distributed adsorbed molecules. Therefore, an increase of the temperature results in a faster decrease of anchoring energy.

Since only the anisotropically adsorbed molecules contribute to the anchoring energy, the estimation of W_0 reduces to the calculation of the concentration of the anisotropically adsorbed molecules $N_{S,a}$ after a fraction of these molecules is desorbed in the isotropic phase. In this case, the distribution function $F_S(\varphi_S)$ is constant (symmetric) and the twist angle φ_{test} is determined by equations (11) and (4). The decrease of the concentration $N_{S,a}$ due to desorption of molecules is described by equation (6) where $A_+N_{VS}=0$ and $F_S(\varphi_S)=\text{const.}$, thus:

$$N_{S,a} = N_{S,a,0} e^{-A_{-}t}.$$
 (12)

Substituting this dependence in equations (12) and (4) we obtain an expression for the anchoring energy,

$$W_0(t) = w_0 \lambda N_{S, a, 0} e^{-A_- t} = W_0|_{t=0} e^{-A_- t}.$$
 (13)

The value $W_0(t)$ is related to the measured twist angle $\varphi_{\text{test}}(t)$ by the equation (11). We fitted the experimental curves $\varphi_{\text{test}}(t,T>T_{NI})$ by the numerical solutions of equation (11) taking into account equation (13). The only variable parameter was the probability of desorption, A_- . The results of fitting are presented in figure 2 for the measured value $W_0|_{t=0} \approx 4 \times 10^{-6} \,\text{J m}^{-2}$ and different temperatures of annealing.

The temperature dependence of the probability of desorption, obtained from the fits, is presented in figure 4 and is well approximated by the Arrhenius law



Figure 4. Dependence of the probability of desorption on the temperature of thermal annealing.

$$A_{-} = A_{-0} \exp(-\Delta E/kT),$$
 (14)

where the activation barrier of desorption $\Delta E \approx 0.55 \text{ eV}$, and the probability of desorption at the room temperature $A_{-}\approx 0.7 \times 10^{-6} \text{ s}^{-1}$ (figure 5). The estimated value of A_{-} correlates with the magnitude $A_{-}=1.1 \times 10^{-6} \text{ s}^{-1}$, which was estimated from the experimental dependence $\varphi_{\text{test}}(t)$ of the relaxation of the easy axis in nematic phase.



Figure 5. Dependence of $\ln A_{-}$ on 1/T which yields an activation energy $\Delta E=0.55$ eV.

The estimated value ΔE lies within the order of magnitude to the activation energy as the hydrogen bonds $E_H=0.13-0.30 \text{ eV}$ [17]. Therefore, we are led to believe that hydrogen bonds are responsible for the adsorption of LC molecules on the tested surface.

It should be noted that a deterioration of magnetically-induced alignment observed in previous studies [3, 4] was also described by the Arrhenius law. The authors explained it by the in-plane rotational diffusion of the LC molecules adsorbed on the surface. The measured value of the activation energy of rotation of 8CB molecules on polyimide surface was 2.16 eV [3] and 0.45 eV for 5CB molecules on polyvinyl alcohol surface. In contrast to our model, the in-plain rotation model does not assume desorption of LC molecules. Since the value $\Delta E \approx 0.55 \,\text{eV}$ is close to the energy of H-bond breaking, our models looks more realistic and well founded. At the same time, additional theoretical and experimental studies are needed to ultimately establish the mechanisms responsible for the changes on the angular distribution of the adsorbed molecules on the ITO surface.

4. Conclusions

The results of our studies show that the layer of adsorbed LC molecules formed by cooling the LC cell from the isotropic phase in the presence of a magnetic field provides rather strong anchoring. The characteristics of the anchoring (anchoring energy value, easy axis direction and its drift) are governed by the angular distribution of the adsorbed molecules on the aligning surface. In particular, the decrease in the anchoring energy upon annealing the LC cell at a temperature above the transition to the isotropic phase is caused by an effective exchange between isotropically distributed "bulk" LC molecules and anisotropically distributed "surface" LC molecules via AD processes. The dependence of the anchoring energy on the duration and temperature at which the LC film was annealed allowed an estimation of the activation energy of desorption of LC molecules on ITO surface, $\Delta E \approx 0.55 \text{ eV}$. The results

suggest that hydrogen bonds are responsible for the adsorption of LC molecules on the substrate.

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