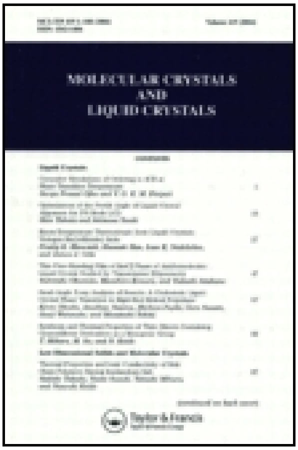

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Surface-Modified Hysteresis of Short Pitch Cholesteric-Nematic Mixtures under Externally Applied Fields

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Dynamic hysteresis behaviors are subjected to testing in short pitch cholesteric-nematic liquid crystal layers at the various boundary conditions. Modification of hysteresis properties near the field induced cholesteric-nematic transition was observed for the differently treated boundary layers and inspected by direct optical methods. It was found the interaction energy between liquid crystal (LC) and boundary layers plays rather essential role in nucleation process than director configuration at the interface and if this interaction energy is essential for tangential constrains, the nucleation occurs mainly at the surface interface and then permeate into the bulk gradually. The experimental results show an influence of surface factor even for a big ratio d/P_0.

Keywords: cholesteric liquid crystals; hysteresis; boundary; nucleation

INTRODUCTION

Since the Greubel [1] first reported that field induced nematic state can be saved even under the field strength below the cholesteric-nematic (CN) transition threshold provided that surface thickness inhomogeneties were reduced, many papers have been devoted to this problem [2-6]. Kashnov *et. al.* [2] were the first to suppose reverse nematic to cholesteric transition occurs both in the bulk and at the surface. Later, Kawachi and Kogure [3] have investigated kinetics of relaxation from electric field induced nematic state in long pitch cholesteric liquid crystals. They were also found the reverse nematic to cholesteric transition to be a nucleation process, which occurs around bulk and surface defects. It was additionally found that the surface treatment plays no role upon characteristics of nucleation process i.e. the nucleation is an exceptionally bulk phenomenon. Meanwhile, such results seems to be very strange, at least if originate from the nucleation theory of *Van Sprang* [7], which is based on the symmetry reasonings. Because of the unique properties of Bragg selective reflection and bistability properties in short pitch cholesterics it is feasible to account their molecular statistic characteristics by direct optical methods using both reflection and transmission techniques.

In this paper we focus on investigation of hysteresis properties near the field induced CN transition for the various boundary conditions.

THEORY

The experimental results indicate that for real samples the boundary conditions play an important role especially for low d/P_o ratio. To account the influence of boundary conditions on the critical fields of texture and phase conversions we used modified approach of of *Kawachi* and *Kogure* [8] elaborated by *Van Sprang* and *Van de Venne* [9]. According to their description, the threshold fields are the crossovers of total free energy curves in an applied electric field and differ from those of *Kawachi* and *Kogure* by incorporation of surface free energy terms. In this way the derived expressions for critical fields are following [9]:

$$E_{cn} = 2\sqrt{2}\left[\left(\frac{\pi}{P_o}\right)^2\left(\frac{K_{22}}{\varepsilon_o\Delta\varepsilon}\right)+\left(\frac{F_{sn}-F_{sc}}{\varepsilon_o\Delta\varepsilon d}\right)\right]^{1/2} \tag{1}$$

$$E_{nc} = \frac{\pi}{P_o}\left[\frac{4K_{22}^2-[K_{33}(P_o/d)]^2}{\varepsilon_o\Delta\varepsilon K_{33}}+\frac{4F_{sn}}{d\varepsilon_o\Delta\varepsilon}\right]^{1/2} \tag{2}$$

$$E_{c'c} = 2\sqrt{2}\left[\frac{F_{sn}-F_{sc'}}{\varepsilon_o\Delta\varepsilon d}\right]^{1/2} \tag{3}$$

where E_{cn}, E_{nc}, $E_{c'c}$ - are the critical fields for CN transition, reverse NC transition and focal conic deformation respectively $F_{sc'}$; F_{sc}, F_{sn} - are the densities of surface free energy in the planar, focal conic and induced nematic states respectively, d - being the cell thickness, K_{22}, K_{33} -are the Frank elastic constants and $\Delta\varepsilon$ - is dielectric anisotropy of liquid crystals. Expressions (1-3) were derived in the assumption that for the planar anchoring conditions $F_{sc}=0$ and for homeotropic $F_{sn}=0$.

RESULTS AND DISCUSSION

The research has performed with the variety of aligning layers that provide the different director configuration at the interface. Preparation of cells has been performed in the following way. Standard glass plates with conducting ITO electrodes were covered by electron beam evaporated SiO_2-Al_2O_3-SiO_2 dielectric protective layers to avoid the shorts. The total thickness of layer was laser controlled and to be about 2200A°. Then the aligning layer was deposited onto protecting layer. Four differently prepared and treated aligning layers were used. Polyimide (PI) surfactant AD-9103 (Ukraine) was spin coated, dried and imidized. When no additional treatment is applied it provides the planar orientation of molecules at the surface, where local director is coplanar with surface plane. Additional treatment of polyimide layers by rubbing provides homogeneous alignment of molecules at the interface, where director is collinear with rubbing direction. Octadecyltrichlorosilane (OTS) films were prepared by dipping and provide homeotropic alignment at the surface, where director is normal to the surface plane. Oblique deposited by cathode reactive sputtering in glow discharge plasma layers silicon oxide SiO_x were also used as aligning layers. Prepared in this manner layers provide weakly tilted alignment of molecules at the interface. After the preparation of alignment layers, the cells were assembled and then filled in a vacuum chamber by liquid crystals.

Cholesteric liquid crystals used in our experiment were a mixture of cholesterol derivatives CH12-M (Ukraine) which was added into nematic host E7 (BDH Ltd.) in ratio 3:2 by weight. The resulting mixture has a temperature independent helical pitch $P_o \approx 0,33\mu m$ and wavelength of maximum selective reflection from Grandjean texture observed in the region of green light $\lambda_m=540nm$ under the normal incidence. The dielectric anisotropy of liquid crystal material is positive $\Delta\varepsilon = +2,6$.

The dependence $(U_{cn})^2 d=f(d)$ according to eq.(1) yields a straight line, which crossovers the ordinate at a point $8(F_{sn}-F_{sc})/\varepsilon_0\Delta\varepsilon$. The respective curves are demonstrated in Fig.1 for the various boundary conditions. In case of homeotropic boundary conditions $F_{sn}=0$, so at once one can obtain F_{sc}. For the planar constrains one can define F_{sn} from cell thickness dependences of $(U_{c'c})d^2$ value. The calculated values of surface free energy terms obtained from experimental curves has brought into Tab.I and found to be of the reasonable magnitude. The remarkable feature is that the E_{cn} magnitudes differ for the cells containing the same surfactant PI with and without mechanical treatment. The threshold fields for unwinding of helix and F_{sn} associated therewith has somewhat declined after treatment of polyimide by rubbing. It has additionally found that the quasi-static hysteresis curves have modified too and became wider. The last one is not clear, because if originate from the nucleation theory elaborated by *Van Sprang* [7] and taking into account experimental results of [8], the birth of cholesteric texture from field-induced nematic state takes place at the sites where the symmetry breaks occur e.g. surface and bulk inhomogeneties, dust particles etc. Our reasonings are that the mechanical treatment of (PI) should increase the surface density of nucleation centers and as a consequence the time for reverse CN transition to be fastened. In order to check this assumption we have provided a series of dynamic hysteresis investigations.

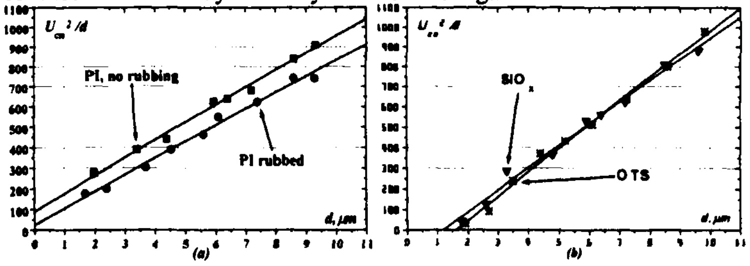


FIGURE 1. The values of $(U_{cn})^2d=f(d)$ as a function of cell thickness.

TABLE I Surface free energy terms for various aligning layers

Surfactant	$F_{SC'}$, (J/m^2)	F_{SC}, (J/m^2)	F_{SN}, (J/m^2)
PI, no rubbing	0	$2.9\cdot10^{-4}$	$5.5\cdot10^{-4}$
PI rubbed	0	$3.2\cdot10^{-4}$	$3.9\cdot10^{-4}$
OTS	$1.8\cdot10^{-4}$	$5.4\cdot10^{-4}$	0
SiO$_x$	$2.1\cdot10^{-4}$	$4.3\cdot10^{-4}$	$7\cdot10^{-5}$

Schematic representation of experimental setup is shown in Fig.2. It consists of the light source (LS), collimator (C), polarizing microscope (M) with the embeded photomultiplier (PM), analog to digital and digital to analog converter (ADC/DAC) and personal computer (PC). The resulting signal of rectangular form simulated by PC, passing through the DAC is applied to the cell. Any changes in transmission or selective reflection of light are detected by electron photomultiplier and converted to the computer by ADC.The dynamic hysteresis measurements as a whole were carried out using conventional technique with the exception of some peculiarities. First the electric field higher than critical field for CN transition E_{cn} was applied and system is switched into homeotropic state. Then the applied field was reduced from field higher than E_{cn} to a bias field $E_b<E_{cn}$. The time of bias field action T_b to be one of the parameters of dynamic response was fixed for each hysteresis curve. Peculiarity the of experiment lies in the very fact, that electric field was reduced from E_{cn} to E_b not gradually as it usually occurs in case of quasi-static hysteresis measurements but stairly as shown in Fig.2. In this case the homeotropic to planar and homeotropic to focal conic modes start to compete with each other at once after the bias field is settled. So the final state of a system depends on how quickly bias field is turned off [8].

Dynamic electro-optical response curves of four aligning layers are shown in Fig.3 for the different bias time T_b. The curves show the transmittance of a system when the bias voltage U_b from field induced nematic state was applied for a bias time and then turned off quickly. Maximum intensity of light corresponds to a focal conic state while the minimum respects to a planar state.

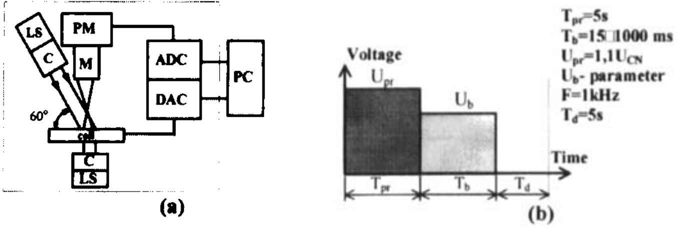


FIGURE 2 Schematic representation of experimental setup for dynamic hysteresis measurements (a) and sequence of driving voltages (b).

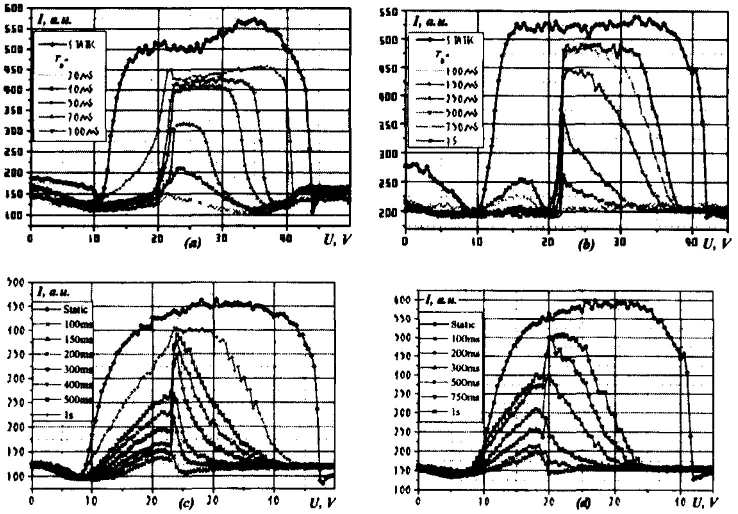
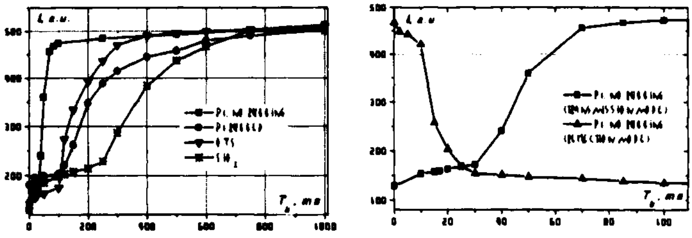


FIGURE 3. Dynamic electrooptical response of the cholesteric systems for samples with different aligning layers (PI no rubbing (a) (d=4,5μm), PI rubbed (b) (d=4,4μm), OTS (c) (d=5,2μm), and SiO_x (d) (d=4,8μm)). Label "static" corresponds to a conventional static response curve of the system.

As it could be seen from Fig.3 all hysteresis loops have the different shape at the same T_b. On the sites with maximum propagation velocity of focal conic domains it was implemented comparative Fig.4 taken from Fig.3. From the curves displayed in Fig.3*b*÷3*d* nearly the same growth and propagation velocity of cholesteric texture is observed. This is the same result as obtained in [8]. However, two aligning layers of polyimide treated by rubbing (Fig.3b) and polyimide with no rubbing (Fig.3a) indicate strongly different hysteresis loops. The nucleation time became about an order of magnitude longer after the treatment of polyimide by rubbing and to be about the same magnitude as for ODTS and SiO_x aligning layers. This kind of result seems to be inconsistent with our assumption of increasing surface defects concentration in rubbed polyimide if no other approach is applied. Increasing of surface defects concentration has to be resulted in the decreasing of nucleation time, but the experimental results indicates vice versa. According to this new approach we believe that not only molecular alignment at the interface must be taken into account, but the interaction energy of molecules with the aligning surface too. Following that assumption one can easily explain such strange behavior of hysteresis curves. In case of strong surface anchoring and planar boundary conditions the nucleation occurs mainly at the surface and then gradually permeates into the bulk. For the weak surface anchoring dominates nucleation around the bulk and surface defects. Because the distance between neighboring nucleation centers is much wider than LC layer thickness, the reverse NC transition in former case occurs much faster. In favor of our assumption may also testify the curve in Fig.4b obtained by measurements in the reflective modes, which shows the reflectance from the interfacial LC layer at first.



(a) (b)

FIGURE 4. Kinetics of nucleation process in cholesteric systems obtained with the various measuring modes.

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