Production and investigation of statically deformed structures in nematic liquid crystals

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The possibility of existence of four symmetrically and optically different structure is experimentally demonstrated in a planar liquid-crystal layer with initial static splay when excited by an alternating magnetic field in various regimes. The case is discussed of a layer with initial static bending. The energetics and dynamics of the change of molecular orientation are analyzed. A new phenomenon, surface-deformation modulation (SDM) of the phase of light passing through a liquid-crystal layer is observed. This modulation increases strongly the contrast of the defects of a surface in contact with a statically deformed liquid-crystal layer.

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Statically deformed structures in nematic liquid crystals (NLC) are attracting increasing interest in connection with practical requirements. Any deformation in a liquid crystal (LC) can be resolved into three elementary deformations —splay (*S*), twist (*T*) and longitudinal bending (*B*). Therefore the study of the properties of NLC with deformed structures of these three types is of decisive significance for the understanding of more complicated structures and for subsequent investigations. At the present time the most investigated and widely used structure is the one with static *T* deformation (so called "twist-effect" devices). The study of structures with static *S* and *B* deformations (*S* and *B* structures) were started only most recently,$^{1)}$ mainly because they were found to possess bistability properties important for the development of bistable and certain matrix devices.$^{1-5}$ These structures were investigated in greater detail in Ref. 5. There, however, as in other studies,$^{1-3}$ a simplified picture was observed of the structural transformations following excitation of the liquid-crystal layer, the reason being that the initial surface tilt of the director region was close to $\pi/4$. In the general case this picture becomes more complicated at other tilt angles.^{4}

We report here an experimental investigation of the picture of structural transformations following the excitation of a liquid-crystal layer with static *S* deformation in its most complete (at present) form, and discuss the case of *B* deformation. A new electro-optical effect in a statically deformed liquid-crystal layer is described.

EXPERIMENT

To obtain statically deformed liquid-crystal layers, cells of the "sandwich" type were used. In this structure the LC is contained between flat glass plates with transparent electodes of indium oxide and orienting coatings of polyimide film. The fan-like orientation in the volume of the LC crystal, typical of splay, was ensured by their inclined orientation at the wall with mutually opposite tilt at the different walls. The tilt at the wall was obtained by the polyimide form in one direction. The sign of the angle is uniquely connected with the polishing direction. Therefore, to obtain splay we used cells with parallel polishing direction of the opposite walls, while in the control cells the polishing was in antiparallel directions, where an undeformed pretilt *P* structure was obtained. Measurement of the average tilt angle by a method similar to that described in Ref. 6 yielded a value 6–9° relative to the LC-layer plane for an antiparallel cell.

In all the cells we used the liquid-crystal mixture esternitryl ZhK-614 with positive dielectric anisotropy. The basic measurements were made at room temperature on cells with LC layer thickness 20 μm.

The electro-optical parameters of the cells were measured with the cells excited with alternating sinusoidal voltage of 50 Hz frequency. In the optical measurement system, a cell with polarizers was placed between the condensor and objective of the projection unit. The cell image was projected with magnification 2–3$^\times$ on a screen with a hole (2 mm diameter) near the optical axis, through which light from a small local region of the cell was incident on the measuring instrument. The cell was placed in a plane perpendicular to the optical axis and could be moved in this plane. The measurements of the voltage-color characteristics were made at normal and oblique incidence of the beam of light on a cell placed between parallel polarizers, whose polarization direction made an angle of 45° with the cell polishing direction. As a result of modulation of the state of polarization of the light beam$^{2)}$ by the cell, some of the wavelengths are extinguished in such a system by the second (exit) polaroid, and the outgoing light beam becomes colored, providing color indication of the phase shift locally at each point of the cell.^{8} Its color coordinates (in the CIE system) were measured with a photoelectric colorimeter. For the temporal measurements we used an FÉU-69 photomultiplier with an S1-18 oscilloscope.

RESULTS AND DISCUSSION

1. Of decisive importance for the production of statically deformed structures are the boundary condition, in this case the static tilt angle α:

$$\alpha_0>0, \quad \alpha_d<0, \tag{1}$$

where α_0 is the tilt angle of the director at one wall, α_d at the other, and d is the thickness of the LC layer. The most interesting practical case is that of mirror symmetry relative to the central plane of the LC layer: $|\alpha_0|\sim|\alpha_d|=\alpha_i$. For small

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values of α_i in the (unexcited) cell with such boundary condition there exists a structure with static splay (S structure).

According to Frank's theory, with increasing α_i and when condition (1) is satisfied the energy of the splay of the structure increases. At a certain $(\alpha_i)_{\rm cr}$, which depends mainly on the ratio of the Frank moduli K_{11} and K_{33}, B deformation becomes energywise favored; therefore for values $\alpha_i > (\alpha_i)_{\rm cr}$ the structure of the LC layer in the cell will be deformed, with static B deformation (B structure). An estimate of the free energies in accordance with Frank (for details, see, e.g., Ref. 5), shows that for the usual ratio $K_{11}/K_{33} \sim 1$ the critical angle is $(\alpha_i)_{\rm cr} \sim 45^\circ$ for many LC. This leads to two consequences:

a) To obtain cells with deformed B structure the boundary conditions must ensure a large angle α_i; our estimate yields the value $70 \lesssim \alpha_i < 90^\circ$ for the customarily employed NLC (the NLC parameters were taken from Ref. 9).

b) When α_i is close to $(\alpha_i)_{\rm cr}$, a small change of α is sufficient for the S structure to change over to the B structure, and conversely, i.e., the energy potential barrier of the transition is a minimum here.

In Refs. 1–3 and 5, $(\alpha_i)_{\rm cr}$ is close to 45°; the orienting coatings were obliquely sputtered oxides that ensure values $\alpha_i \approx 40^\circ$ (Ref. 5) and 35–39° (Refs. 1–3) relative to the LC-layer plane (the measurements were made, just in our case, for a tilted P structure). The obvious physical difference between our case and those mentioned above is in the height of the energy potential barrier of the $S \rightarrow B$ transition, which in our case was close to the maximum in view of the smallness of α_i (6–9°). This leads to qualitatively new phenomena when the cell is excited with an electric field.

2. When the voltage applied to the cell with initial S structure (S cell) normal to the LC layer is smoothly increased and reaches a value $U_1 \approx 2.2$–2.4 V, one can distinctly see a structural transition: the new (second) structure appears in the form of smeared-out spots simultaneously over the entire area of the cell and crowds out the initial one rapidly. The contrast of the spots increases noticeably when the observed region is moved farther away from the optical axis of the system along the polishing direction (i.e., with increasing light incidence angle $|\varphi|$ in the polishing plane). The color of the beam observed on the screen at the exit from the cell near the optical axis hardly differs from the color of the initial S structure before the transition, thus indicating that the corresponding optical phase shifts are close.[8]

With further increase of the voltage above $U_2 \approx 4.8$–5 V, a second structural transition is even more strongly pronounced: the next (third) structure appears as a result of sharply outlined spots and crowds out the preceding one at a rate proportional here to the difference U–U_2. The optical phase shifts of the structures differ radically, and disclinations can be noted on the boundaries. This last structure exists already all the way to saturation at 15 V and is metastable, since it is destroyed only when the voltage is lowered below $U_3 \approx 2.1$–2.2 V; the time of its decay when the voltage is turned off is relatively long and reaches 8–12 sec (see Fig. 1).

When the voltage is applied to a control cell with initial P structure (P cell), no transitions are observed at any control regime, and all its properties correspond to the known properties of the tilted P structure.

A careful comparison of all the experimental facts has made it possible to construct a hypothetical scheme of the onset of the structures and of the transitions between them (see Fig. 2). The initial deformed S structure goes over at a voltage U_1 into a structure similar to the tilted P structure, which in turn goes over at the voltage U_2 into a deformed B structure. We present below a basis for this scheme.

3. When the electric field is turned on it tends to align the LC molecules along the force lines; their orientation changes slightly, with the exception of the molecules of the center layer, which at low voltages are close to an unstable equilibrium (but do not reach it), see Figs. 2b and 2f. At the voltage U_1 the splay becomes energetically unprofitable, the molecules of the central layer are in an unstable equilibrium, and in the case of weak pinning of the molecules to the walls the molecular orientation is "pushed out" with zero tilt in one direction or another (see Figs. 2c and 2g), accompanied by detachment of the molecules at one of the walls from the direction of the preferred orientation (which is determined

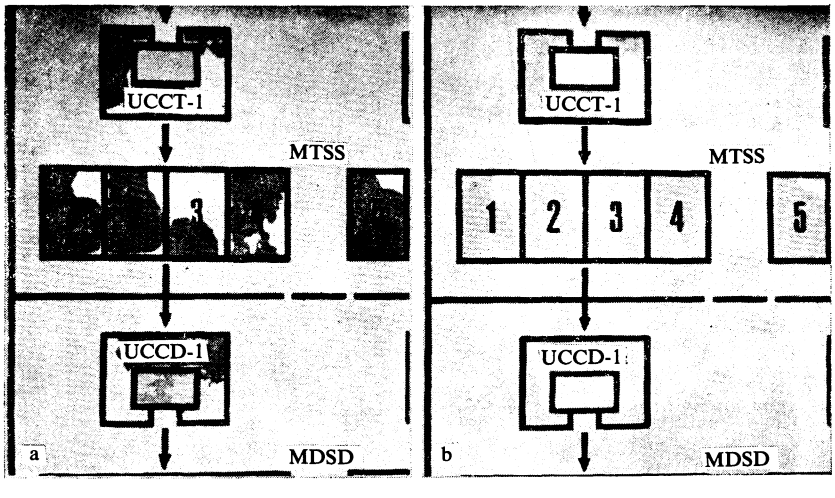

FIG. 1. Display of the structural transition $S \leftrightarrow B$ on a screen; the exciting voltages 8 V and is applied only on part of a memory system marked by the numbers: a — 3 sec after application of the voltage on the unexcited cell, b — 3 sec after removal of the voltage (performed immediately after the first photograph): dark regions — relaxing B structure, $B \rightarrow S$ transition, light region — the rapidly relaxed S structure no longer differs from the unexcited background. The shown section of the LC memory circuit is 24.5×28 mm.

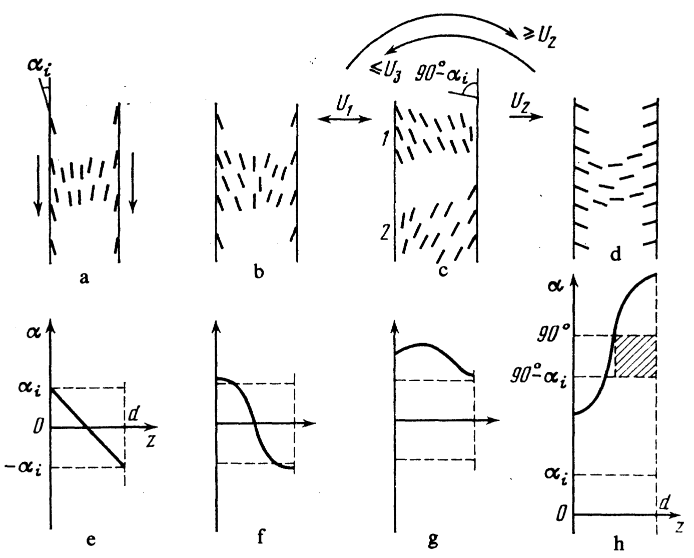

FIG. 2. Diagram of the structures and the transitions produced in an LC cell with static S deformation upon excitation: a — $U = 0$; b — $0 < U < U_1$, S structure; c — $U_1 < U < U_2$, both types of analogs of an inclined structure, P_1 and P_2, are shown; d — $U_2 < U$, metastable B structure, goes over only into an S structure at $U \leqslant U_3$. The change of the tilt angle α of the molecule across the LC layer for cases a–d is shown respectively in Figs. e–h: the coordinate z is perpendicular to the walls, d is the thickness of the layer. The curve in Fig. g is only for P_1, and the shaded region in Fig. h is the forbidden band (a similar region is located at $-90° < \alpha < -90° + \alpha_i$ for the molecules near the first wall $0 < z < d/2$).

by the interaction at the wall and is described by α_i) and by formation of an analog of a tilted P structure.

The resultant structure was identified in accordance with the following experimental attributes:

a) A considerable and monotonic change of the optical phase shift is observed when the sign and magnitude of the angle φ of incidence of the light on the cell is changed.

b) The sign of the rotation angle of the molecules of the central layer depends on random factors, therefore the formation of two different energetically degenerate analogs of a P structure with opposite signs of α_0 and α_d is equally probable. Indeed, simultaneous formation of different regions with structures P_1 and P_2 was observed in all the experimental S cells at $U \geqslant U_1$ (Fig. 2c).

c) The P_1 and P_2 structures differ only in the case of oblique incidence of the light ($\varphi \neq 0$). When the angle of incidence φ reverses sign the neighboring regions P_1 and P_2 "exchange" colors, i.e., the optical phase shift

$$\Delta_1(\varphi) = \Delta_2(-\varphi), \quad \Delta_2(\varphi) = \Delta_1(-\varphi),$$

and their color-voltage characteristics are strictly identical, see Figs. 3 and 4.

d) All the measured electro-optical parameters of the P_1 and P_2 structures of an S cell, including the response times, coincide or are close to the parameters of a P cell with tilted P structure (Figs. 3 and 4).

The formation of the structures $P_{1,2}$ is the most significant qualitative difference between the general pictures of the structural transformations in the S cell from that described in Refs. 1–3 and 5. In place of the two stable states observed in those references, separated by a small potential barrier ("bistability"), the decrease of α_i led in our case not simply to an increase of the height of the barrier but also to the appearance, as follows from the general picture of the experiment in Fig. 2 and from a comparative estimate, of a local energy minimum that characterizes a third state degen-

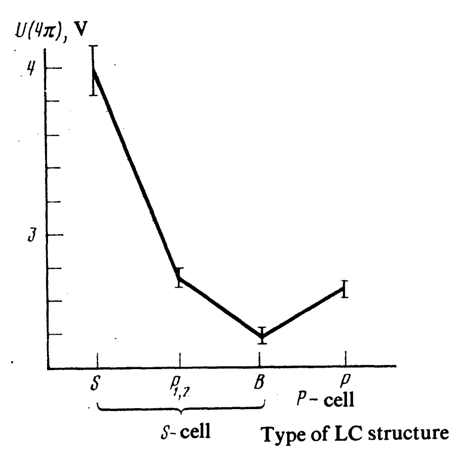

FIG. 3. Dependence of the voltage corresponding to an optical phase shift $\Delta = 4\pi$ at a light wavelength 550 nm on the type of the LC structure (for the S structure the measurements are approximate and were made using the duration of the $S \rightarrow B$ transition).

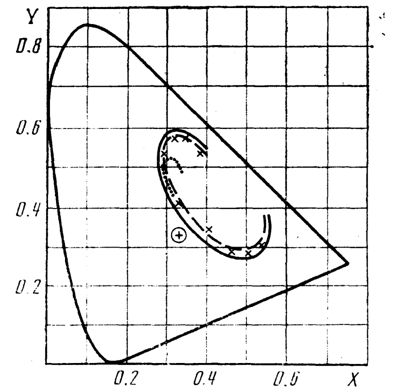

FIG. 4. Segments of color curves for different structures in paraxial light beams: dotted curve — S structure, dashed — $P_{1,2}$ structures, solid — B structure, crosses — P structure of control P cell. On each curve the point closest to the upper edge of the "color triangle" corresponds to a phase shift of 4π (550 nm).

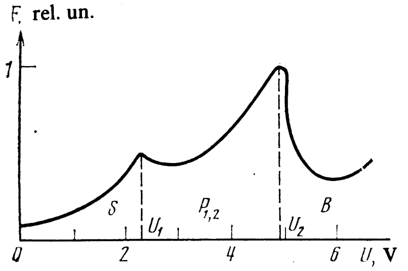

FIG. 5. Dependence of the averaged free energy of the LC layer in an S cell with small α_i on the voltage applied normal to the layer. Dashed lines — boundaries of the regions of existence of different structures.

erate in the two orientations of the boundary regions of the LC, Fig. 5. The depth of this minimum is obviously connected with the strength of pinning of the molecules of the LC to the bounding surfaces—when the pinning is increased it should tend to zero. In view of the energy degeneracy of P_1 and P_2, we consider hereafter only P_1.

4. When the voltages increase from U_1 to U_2, the molecules at the second wall ($z = d$ in Fig. 2) reach a tilt $\alpha = 90° - \alpha_i$, i.e., normal to the preferred orientation. Here they enter into an unstable equilibrium relative to the torque of the wall forces, but there is no equilibrium relative to the field torque, therefore the field rotates the molecules to a tilt $\alpha = 90°$; the increase of α and the final equilibrium tilt are furthermore again determined by the competition between the field and wall torques. In other words, at $U = U_2$ a jump-like increase takes place in the angle of tilt of the molecules at this wall, by more than $2\alpha_i$ (Figs. 2d, h).

Thus, for a static tilt angle there exists here a sort of forbidden band described by the relation

$$90°-\alpha_i \leqslant \alpha \leqslant 90° \qquad (2)$$

(this band is shaded in Fig. 2h; a similar band at $-90° \leqslant \alpha \leqslant -90° + \alpha_i$ for the molecules at the other wall occurs in the case of P_2), i.e., at no value of the voltage can the molecules in the region $z > d/2$ in the structural sequence $S \rightarrow P_1$ have a static angle tilt in the range (2).

This explains the metastability of the structure with the highest voltage. In analogy with the situation of Sec. 1, B deformation becomes here energetically more favorable, and therefore at voltages $U \geqslant U_2$ a static deformed B structure is produced in the cell (Fig. 2, 5). Our comparative estimates, with the aid of the results of Ref. 5, of the free energy of an LC layer shows that the depth of the potential minimum of the B structure exceeds the depth of the local minimum $P_{1,2}$, a fact understandable from the geometry of the B structure: the molecules in the central layer are situated along force lines, and the tilt of the molecules at the wall is close to the preferred value. The condition for the decay of the molecule is an increase in the energy of the B deformation, which indeed takes place at low voltages $\leqslant U_3$. Here, however, the transition to an energetically favored S structure is made difficult and prolonged (8–12 sec), since this calls for rotation of the molecules of the central layer in the viscous LC medium immediately through 90° (see Sec. 2 and Fig. 1b). A similar prolonged transition is observed when a voltage $U > U_2$ is applied to the unexcited cell (i.e., transition from the S to the B structure), and in this case its duration $\tau_{\text{tr}} \sim (U - U_2)^{-1}$. In the latter case the picture is similar to that described in Refs. 1–3 and 5: centers of nucleation of the B structure are formed on the defects of the LC layer, and this structure increases on them uniformly in all directions at a finite rate (Fig. 1a).

It can be seen from Fig. 2 that for different structures, by virtue of their geometry, there should be observed unequal values of the optical phase shifts at identical voltage, or, conversely, different voltages to obtain one and the same phase shift. Experiment confirms the latter (see Fig. 3), and the relative voltages agree with the scheme of the structural transitions.

Near the stability limit U_3, the behavior of the B structure becomes more complicated. Whereas the temperature dependence of its response times (to the turning on and off) for a given change of the optical phase transition far from the stability boundary has the usual monotonic character (see Fig. 6a), near the boundary the dependence becomes nonmonotonic (Fig. 6b), becoming steeper when this boundary is crossed (Fig. 6c), something not observed earlier in NLC.

It is most important for applications that the B structure has a symmetry plane passing through the central layer

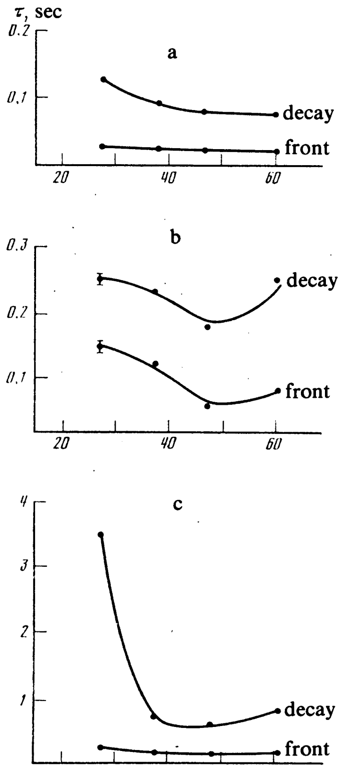

FIG. 6. Temperature dependence of the times of the response of the B structure for a given change of the phase shifts: a — switching of the control voltage from a level corresponding to $\Delta = 2\pi$ to the level $\sim 1.2\pi$ (for 590 nm, yellow ↔ brownish color, voltages 3.6 ↔ 9.1 V at 27° C). b — The same for $\Delta = 4\pi \leftrightarrow 2.2\pi$ (550 nm, green ↔ yellow, 2.5 ↔ 3.6 V). c — The same for turning off the voltage from the 3.6-V level (transparent ↔ yellow, 0 ↔ 3.6 V; the duration of the pause between the packets of the sinusoidal pulses is 4 sec, i.e., short enough to prevent a $B \rightarrow S$ transition).

$z = d/2$. This decreases noticeably the dependence of the phase shift on the light incidence angle φ, and this leads to a considerably better homogeneity of the color of the output beam over the area of the cell compared with the asymmetric P structure. In addition, for rays incident at equal angles but opposite signs, the phase shifts are equal, therefore the saturation of the colors of the B structure is noticeably improved compared with the remaining ones even in paraxial rays (Fig. 4). With increasing φ, the difference in the saturations also increases. Our measurements have shown that the symmetry of the B structure makes it possible to raise the aperture of the projecting light beam in the optical system to 25–27°, compared with the 4° for the usually employed inclined, homogeneous, and homeotropic structures. This is very important for controllable optical filters,[7] for memory systems and phase plates,[4,8] and for bistable devices.[1–3]

5. When the frequency of the control voltage was lowered to 1–3 Hz in a B structure, we observed for the first time an increase, by many times, of the contrast of small defects of the boundary surfaces, defects usually unobservable under a microscope at any magnification. These defects, in the form of bands and ripple, are noticeable on Fig. 7 (cf. Fig. 1) in the excited regions of the LC layers. At these frequencies, the LC molecules react by changing orientation even during each half-cycle of the sinusoidal voltage, i.e., they "swing into oscillations." At the zero points of the sinusoid, the structure is beyond the stability region, but owing to its duration the deformation transition from the B structure cannot extend fully over one-quarter of the period—on the average the structure becomes balanced at the stability region. In the regions of the surface defects, the local values of α_i differ from the mean value, shifting also the local voltage stability region to one direction or another. At dynamic equilibrium there appear therefore deformation-transition, or "pretransition" regions, which are inhomogeneous over the depth of penetration into the LC layer because of the difference between the local inhomogeneities of the voltage-stability boundary.

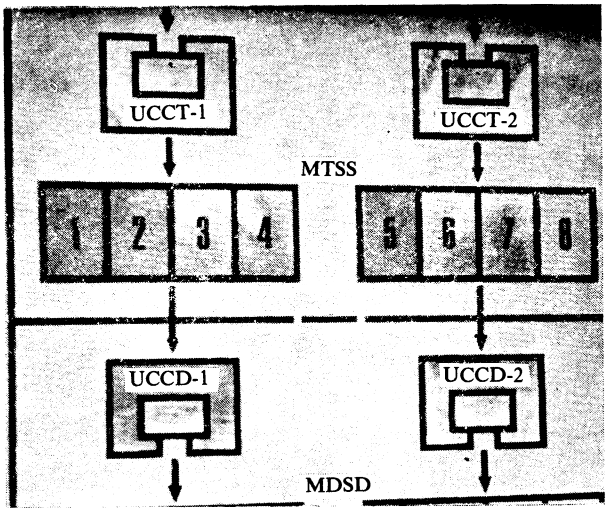

FIG. 7. SDM of the phase of the transmitted light. Voltage 6.5 V, frequency 3 Hz. The contrast of the surface defects (of the bands and of the ripple on most exciting sections of the memory) on the black-and-white photograph is small because of partial integration of the varying color picture during the time of the frame exposure (0.1 sec). No analogous defects are visible on the surrounding unexcited background.

Judging from all the foregoing, a "standing wave" of the deformation transition $B \rightarrow S$ or $B \rightarrow P$, modulated by the surface defects, is produced and it is the one which causes the spatial modulation of the phase shift of the transmitted light beam, a modulation that illustrates the relief of the defects. The phase shift at each point of the cell changes regularly at double the frequency of the control voltage, but the difference between the phase shifts of the defected and defect-free regions can be observed at any instant of time (Fig. 7). The LC plays here the role of an "amplifier" (or "developer") that perturbs the action of the small surface defects on the passing light waves. This phenomenon makes it possible to observe defects with the unaided eye, we have called it surface-deformation modulation (SDM) of the phase of the transmitted light.

CONCLUSION

Thus, in a flat nematic LC layer with initial static splay, at various excitation regimes and amplitudes of the exciting alternating electric field, there can be obtained reversibly four symmetrically and optically different structures:

1) An S structure with mirror symmetry about the central plane.

2) Two energetically degenerate structures $P_{1,2}$ — close analogs of an axisymmetric tilted P structure.

3) B structure with mirror symmetry about the central plane.

The structure with the highest voltage is in this case metastable. In other words, using the terminology of Refs. 1–3 and 5, in place of the bistability described there we have observed experimentally in the present paper tristability of a statically deformed NLC layer.

Starting from the foregoing results, and also from the physical equivalence of the S and D effects, which are modifications of the Freedericksz effects for different (homogeneous and homeotropic) initial orientations of the LC layer, one should expect for a thin cell (less than 30 μm) with initial static B deformations and with large α_i (see item 1 of the preceding section), filled with an NLC with $\Delta\varepsilon < 0$, the picture of the structural transitions that appears when the film is excited will be the inverse of the case analyzed above, that of a cell with S deformation and $\Delta\varepsilon > 0$. Stated in greater detail, when the voltage is smoothly increased one should observe first a transition from the B into the $P_{1,2}$ structures, and then from the $P_{1,2}$ into the S structure, and here, naturally, the metastable structure should already be the S structure. To be sure, in thicker layers (30 μm and more), there can arise here also other deformation phenomena,[10] capable of changing the overall picture.

Depending on the regime of the excitation of the LC layer, we observed in the experiment structural transitions both with conservation ($S \leftrightarrow B$) and with change $\overleftarrow{S}\ \overline{\leftrightarrow P_{1,2} \rightarrow B^{\downarrow}}$ of the type of symmetry. Simultaneously, in different regions over the area of the layer, one could observe up to three different structures, e.g., $P_{1,2}$ and B.

We observed for the first time in a liquid crystal, and analyzed qualitatively, the electro-optical SDM effect which manifests itself in a considerable enhancement of the contrast of small surface defects that are in contact with the liquid crystal with static *P* formation, at low frequencies of the control voltage.

On the basis of the described phenomena it is possible to develop a new method of determining certain molecular constants of LC by measuring the characteristics of the described transitions and of the SDM effect under conditions that the adhesion energy of the LC molecules to the surface is determined beforehand, and taking into account in an exact Frank calculation of the free energies.

Statically deformed *S* and *B* structures are quite promising for applications in bistable devices controllable, in optical filters, and also in memory systems and controllable phase plates. Their application, by virtue of their symmetry, makes it possible to improve the parameters of such devices and lower considerably the requirements on the optical systems with which they operate.

Great interest attaches also to the possibility of using *S* and *B* cells to obtain thorough electric or flexoelectric samples of the LC type, described in Refs. 9 and 11. This possibility can be realized when the *S* cell is filled with a liquid crystal with molecules of wedge-shaped form, while the *B* cells are filled with molecules of arc-shaped form, having nonzero electric dipole moment.

In conclusion I am grateful to N. V. Murtazina for help in producing part of the experimental samples and Z. G. Petrenko for interest in the work, as well as Professor A. S. Sonin for very helpful discussion of the results.

[1] In Refs. 1–3 the *S* and *B* structures were named "horizontal" and "vertical." Obscuring the true geometry of these structures (which correspond more readily to homogeneous and homeotropic structures), these designations do not agree with the customary designation of the third basic type of statically deformed structures, namely twist structure, let alone with relation of the concepts of verticality and horizontality for LC cells. In the present paper we retain the terminology of Ref. 4.

[2] But not the rotation of the plane of polarization of linearly polarized light, as stated in error in Ref. 7.

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