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Strain-texture transition in type-A smectics

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To explain the physical nature of the transition of the homotropic smectic texture into a confocal one, we investigated the mechano-optical characteristics in smectic liquid crystals. A mechano-optical hysteresis is observed in smectics of type *A*. An experimental setup capable of investigating the mechano-optical effects in liquid crystals is described. The transition of the homotropic structure into a confocal one, due to stretching of the smectic layer, is investigated in detail. It is shown that this transition is from one texture into another, and is not a phase transition. This texture transition is responsible for the observed mechano-optical hysteresis. It is established that both textures, the homotropic and the confocal, are stable for the smectic type-*A* phase.

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A smectic liquid crystal of type A has a layered structure in which the long axes of the molecules are perpendicular to the layers, whose thicknesses are of the order of the molecule length, 20–30 Å. The optical properties of smectic-A are analogous to the optical properties of a solid uniaxial crystal. The mechanical properties have a large anisotropy. Thus, the elasticity measured perpendicular to the layers is closer to the elasticity of the solid crystal, while the elasticity measured along the layers is similar to the elasticity of the nematic liquid crystal.

Delaye *et al.*[1] and Clark and Meyer[2] have reported observation of mechanical instability in smectic A, wherein flexure of the smectic layer with a definite period takes place. The instability appeared at small strains, lower than a certain critical value. The flexure relaxation time was of the order of several milliseconds. The reason for the instability is that the tensile force that tends to separate the smectic layers is balanced by the wavy distortion that is produced in the layer. It was noted in the cited papers that tensile strains exceeding the critical value lead to a transition from the homotropic texture to a texture that consists of bright regions—domains. The relaxation time of such a domain texture is long, on the order of a minute. The causes of the appearance of the domain structure at large strains were, however, not considered in[1,2]. We shall henceforth call the transition into a domain texture a strain-texture transition (STT).

STT were later observed in a new class of smectics of type A,[3,4] which are obtained when two nematics are mixed. These systems offer the advantage that they are in the smectic liquid-crystal phase at room temperature.

To investigate the STT we developed the experimental setup shown in Fig. 1. A liquid-crystal layer 1 of approximate area 1 cm^2 was placed between two glasses 2. The glasses are coated on the inside by a transparent

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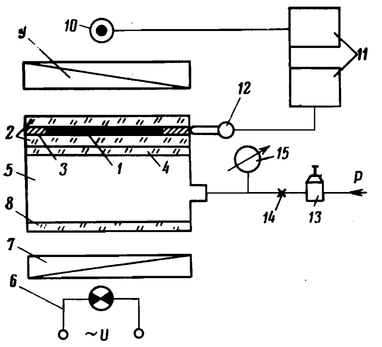
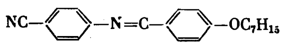


FIG. 1. Experimental setup for the investigation of the mechano-optical properties of liquid crystals.

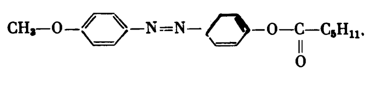
conducting SnO_2 layer, so that an electric field can be applied on the liquid-crystal layer. The layer thickness is determined by the thickness of the elastic gasket 3. The liquid-crystal cell produced in this manner is mounted on the transparent surface 4 of a hermetically sealed chamber 5. Light from source 6 passes through polarizer 7, the glass bottom of the chamber 8, the liquid-crystal cell, and the analyzer 9 and is incident on the photomultiplier 10. The output of the photomultiplier is connected to the recording instrument 11. Thermocouple 12 records the temperature. The pressure source is compressed nitrogen that flows from the glask through a pressure reducer 13, a valve 14, and a manometer 15 into chamber 5 and applies uniaxial pressure uniformly over the entire liquid-crystal layer area. The setup makes it possible to measure the dependence of the change of the transparency of the liquid-crystal layer on the pressure at various temperatures, to investigate the scattering of the light by the liquid-crystal layer, to carry out microscopic observations of the sample, and to take photographs by placing the chamber in the field of view of a polarization microscope.

The STT was investigated in substances forming the smectic phase A obtained by mixing nematics, and in the pure substance 4-nitrophenyl-4-octyloxybenzoate, which is in the smectic phase A at temperatures from 49 to 61 °C. The following mixtures were investigated:

1) 50% *p*-*n*-heptyloxybenzylidene-*n*′-aminobenzonitrile

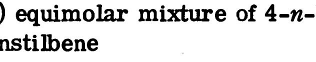


+50% ether 4-metoxybenzene-4-oxycaproic acid

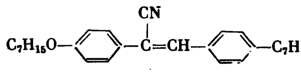


2) 45% 4-cyanphenyl ester of 4-*n*-hexylbenzoic acid

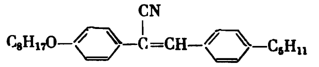
+55% 4-*n*-hexyloxyphenyl ester of 4′-*n*′-butylbenzoic acid



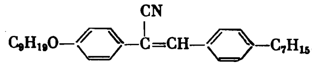
3) equimolar mixture of 4-*n*-hyptyloxy-4′-*n*-heptyl-α-cyanstilbene



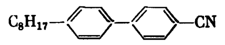
4-*n*-octyloxy-4′-*n*-amyl-α-cyanstilbene



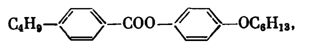
4-*n*-nonyloxy-4-*n*-heptyl-α-cyanstilbene



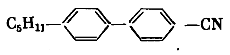
4) equimolare mixture of 4-*n*-octyl-4′-cyandiphenyl



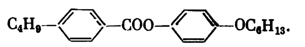
4-*n*-hexyloxyphenyl ester of 4′-*n*-butylbenzoic acid



5) Equimolar mixture of 4-*n*-amyl-4′-cyandiphenyl



4-*n*-hyxyloxyphenyl ester of 4′-*n*-butylbenzoic acid



The phase transition points of the mixtures were:

1) 28–75 °C, 2) 12–41 °C, 3) 8–43 °C, 4) 2–50 °C, 5) 6–39 °C.

As a result of the investigations, we obtained a family of hysteresis curves for the dependence of the transparency of the liquid-crystal samples on the pressure in the range from zero to 1 kgf/cm². Figure 2 shows these plots for five different smectic liquid crystals. The abscissas show the pressures, and the ordinates the ratios of the light intensity passing through the sample in the absence of pressure to the light intensity corresponding to the given pressure.

In the absence of pressure, the sample had the texture shown in Fig. 3a and a minimal transparency. The light, after passing through an inhomogeneous sample, was

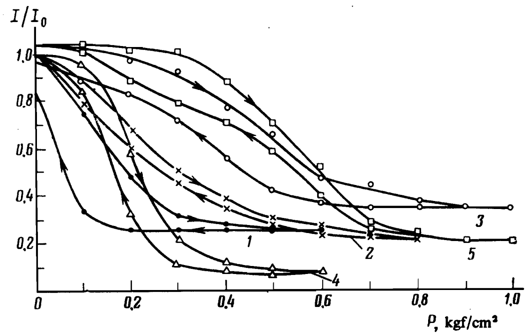


FIG. 2. Mechano-optical hysteresis (dependence of the relative light transmission on the pressure). The numbers on the curves correspond to the numbers of the mixtures.

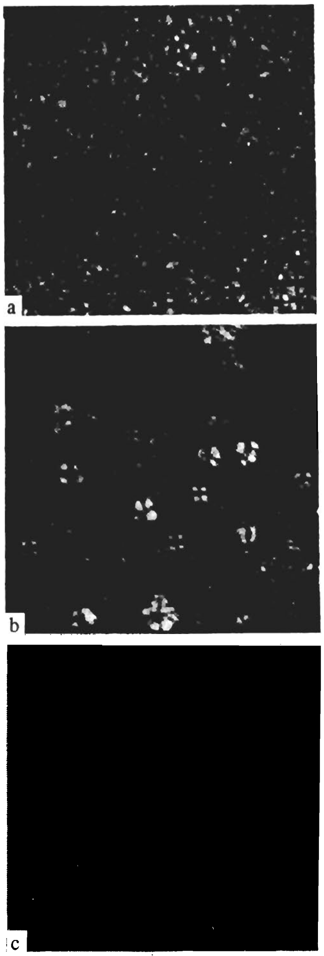


FIG. 3. Microphotographs of sample in crossed polaroids: a—domain texture, b—homotropic texture, c—appearance of domains.

scattered by the texture. The pressure compressed the smectic layers and caused the texture to vanish and the transparency of the liquid crystal to increase. At a certain critical pressure the transparency reached a maximum, and this correspond to the transition of the liquid-crystal layer to the homotopic smectic phase—Fig. 3b. Gradual removal of the pressure led to a dilatation of the homotropic smectic in a direction perpendicular to the smectic layers. Individual domains—regions in which the homotropic orientation was destroyed—appeared in the layers (Fig. 3c). With decreasing pressure, the number of such regions has increased and led to a decrease of the sample transparency.

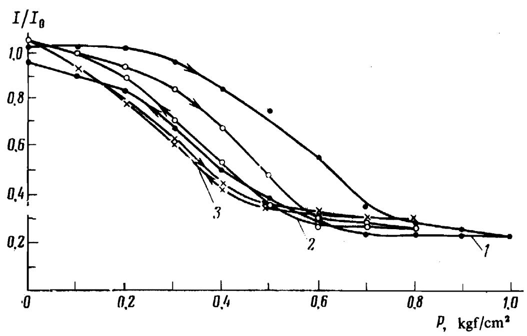


FIG. 4. Mechano-optical hysteresis of substance No. 2 at the temperatures: 1) 15 °C, 2) 25 °C, 3) 35 °C.

Optical hysteresis is observed for all the investigated substances. With decreasing pressure, the liquid-crystal layer retained for some time a homotropic orientation, until the tensile strain reached a definite value P_{tr} for each substance. At the pressure P_{tr}, the transparency began to decrease. Since the measurements were performed in unpolarized light, the threshold is not sharply pronounced on the plots of the intensity against pressure. It is seen from Fig. 2 that each substance has its own transparency characteristics and its own value of hysteresis. In this case the half-width of the hysteresis loops is apparently determined by the viscosity of the smectic phase. The half-width of the hysteresis tend tends to zero when the sample is heated (Fig. 4). The mechano-optical characteristics obtained in polarized light show that hysteresis is present, but takes a different form—Fig. 5. This form of hysteresis can be attributed to the vanishing of the domains with increasing compression stress. The domains vanish gradually, in agreement with the smooth character of the plot I against P. In the case of tension, the strain accumulates, as it were, and at a certain critical pressure P_{tr} the homotropic texture (Fig. 3b) goes over into the domain structure (Fig. 3a), corresponding to the reverse dependence of I on P. Formation of only several domains is sufficient to allow the light to pass through the crossed polaroids to the photomultiplier. Naturally, the form of the hysteresis in crossed Nicols is reversed, i.e., in the absence of pressure the light passing through the sample is a maximum, and at the highest pressure and at homotropic orientation it is a minimum.

To determine the dimensions of the domains produced upon elongation of the smectic layers, we investigated the scattering of light. A liquid-crystal layer 50 μm thick was placed between two transparent glasses coated on the inside with a conducting transparent layer. When a static pressure was applied to the layer its thickness decreased to 49.7 μm, after which the layer, was transferred to a state with homogeneous homotropic orientation by applying an orienting electric field. The homotropic texture was preserved after removal of the electric field. The pressure was gradually decreased until the light-scattering ring appeared. The ring appeared at a strain corresponding to linear elongation of the layer by 0.3 μm in a direction perpendicular to the surface. The pressure was then gradually decreased and this led to a definite increase of the layer thickness Δl and

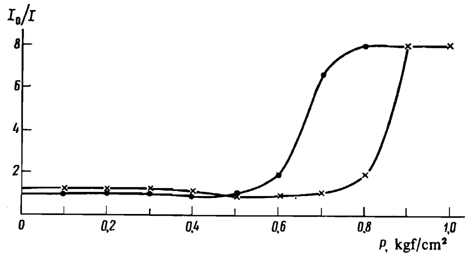


FIG. 5. Mechano-optical hysteresis of substance No. 4 in crossed polaroids.

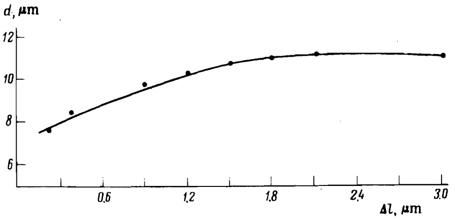


FIG. 6. Dimension d of the domains produced in the homotropic texture vs the elongation Δl of the liquid-crystal layer. Sample thickness $h=50$ μm.

correspondingly to a tension strain. For each Δl we determined the scattering angle and respectively the dimension d of the scattering region (domain). It turned out that with increasing tension the domain dimension increased up to $\Delta l=2$ μm, but remained constant with further increase in tension (Fig. 6). At $\Delta l=1.5$ μm there appears a texture that produces a good diffraction picture consisting of four or six diffraction maxima. Such a diffraction picture corresponds to domains of quadratic or triangular shape. Increasing Δl by more than 1.5 μm erases the sharp geometrical boundaries of the domains and produces a diffuse light-scattering ring. The domain dimension (Fig. 6), starting with 0.9 and down to 0.3 μm, decreases rapidly with decreasing strain. The described experimental investigations were made at strains larger than 0.3 μm, which exceeded by two orders of magnitude the strains considered in[1,2], and we therefore did not observe the mechanical instability that was produced in those investigations.

In contrast to the results of[1,2] the strains used by us led to the appearance of a stable texture (Fig. 3a), which did not vary with time. However, if the stability of the homotropic texture is increased by applying an orienting field, then mechanical instability can be observed also at strains greatly exceeding the critical value. Increasing the voltage applied to the sample decreases rapidly the time of vanishing of the domain texture produced by stretching the smectic layers. In the domain texture the director is inclined to the normal and it can be assumed that in the domain the director orientation tends to be planar. Then the transition from the confocal texture to the homotropic one under the influence of the field E will be the usual field-induced reorientation of the director from planar to homotropic.

The reorientation time τ of the field effect of the nematic phase is expressed in the form[5]

$$\tau=\gamma_1\left(\frac{\Delta\varepsilon E^2}{4\pi}-\frac{k_{ii}\pi^2}{d^2}\right)^{-1}, \tag{1}$$

where γ_1 is the viscosity coefficient in a direction perpendicular to the layers, $\Delta\varepsilon$ is the anisotropy of the dielectric constant, k_{ii} is the elasticity coefficient, d is the thickness of the deformed liquid-crystal sample, and E is the electric field intensity. Since the quantities γ_1, $\Delta\varepsilon$, k_{ii}, and d are constant, it follows that relation (1) for the smectic phase can be represented in the form

$$\tau=A/(E^2-B), \tag{2}$$

where A and B are constants. Consequently, the time τ of vanishing of the domain texture should have a quadratic dependence on the orientic field E. This is precisely the dependence observed in experiment for τ (see Fig. 7). It is seen that within the limits of the measurement errors the point fit a straight line well. Consequently, relation (1) describes correctly the reorientation effect in the smectic A and can be used to calculate the viscosity coefficient or the elasticity coefficient of a smectic liquid crystal.

Analyzing the experimental results, we can conclude that when the layers in the smectic phase A are stretched the homotropic texture changes into a domain texture. Owing to the tension-induced inclination of the smectic layers, the homotropic homogeneous orientation breaks up into domains. The domain dimensions depend on the deforming force. Tension that increases the layer thickness by up to 3% increases the domain diameter. An ordered domain picture capable of producing diffraction is then formed. When the layer thickness increases by more than 3%, the texture is mechanically damaged, but the domain dimension is unchanged (Fig. 6). The domain dimension depends also on the sample thickness. With decreasing sample layer thickness the average domain dimension decreases:

h,μm:	5	15	20	30	50	70	100
d,μm:	4.2	4.9	5.2	5.25	8.05	8.7	9.25

The reason is that when the sample thickness h is decreased the number of smectic layers taking part in the flexture also decreases.

The presence of mechano-optical hysteresis and its temperature dependence demonstrate that an important role in the transition from one texture to another is played by the value of the internal friction of the smectic layers.

The transition of a confocal texture into a homotropic one under the influence of an electric field corresponds to reorientation of the director in the electric field, from the planar position to a normal one, because the investigated substances have positive values of $\Delta\varepsilon$.

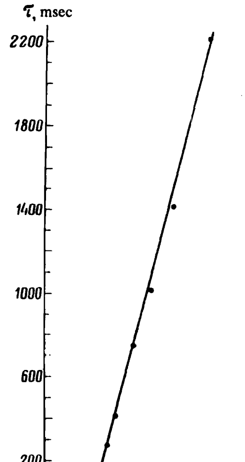


FIG. 7. Dependence of the time τ of the transition of the domain texture into homotropic on the electric field intensity E.

All these facts indicate once more that the smectic phase A can be in two stable textures: homotropic, which practically corresponds to the liquid-single-crystal state of the sample, and domain texture, corresponding to liquid-polycrystalline state. In this case tension destroys the liquid single crystal state and converts the layer into a liquid-polycrystalline state, while the electric field orients the molecules along the field and makes the sample a liquid single crystal.

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Contribution to the theory of high-frequency properties of ferromagnets

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We develop a unique method, based upon a special representation for the spin operators, to calculate the high-frequency, kinetic, and thermodynamic properties of magnetic substances. We use this method to calculate the high-frequency susceptibility, the spectrum and the damping the spin waves, the thermodynamic potential, and the magnetization of a Heisenberg ferromagnet. We show that the components $\chi_{xx}(\mathbf{k},\omega)$ and $\chi_{yy}\mathbf{k},\omega)$ in this representation reduce simply to single-particle Green functions of the quasi-particles.

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1. INTRODUCTION

A large number of papers has been devoted to calculating the high-frequency properties of ferromagnets. The theory of these effects is mainly based upon the following approaches: a phenomenological one, going back to Landau and Lifshitz's well known paper[1]; the kinetic equation method, developed by Akhiezer[2] on the basis of the Holstein-Primakoff (HP) representation[3] for spin operators; the Green function method, using the HP representation of the Dyson-Maleev (DM) representation[4]; finally, the Green function method using the Bogolyubov-Tyablikov,[5] the Vaks-Larkin-Pikin,[6] or the Izyumov-Kasan-Ogly[7] spin operators.

By now we have reached an understanding of the basic regularities connected with the high-frequency properties of magnetic substances such as resonance effects, parametric excitation of spin waves, and also thermodynamic properties at low temperatures. There are, however, a number of problems which are important not only for constructing a theory of magnetically ordered crystals, but also for a correct understanding of experiments. Among these there is, first of all, the problem of a consistent evaluation of the components of the high-frequency (hf) magnetic susceptibility tensor and the problem, closely connected with it, of determining the spectra and interaction of quasi-particles. Traditionally this problem is solved by changing from spin operators to Bose operators using the HP representation. The dynamical interaction of the spins is then replaced by the interaction of the Bose particles. The HP Hamiltonian can serve as a basis for a consistent calculation of the thermodynamic properties of a magnetic substance. As far as the interaction of the spin system with an external field is concerned, the situation here is appreciably more complicated.

An external field acts directly on the spin variables which are in an essentially non-linear way connected with the HP spin wave operators. We shall show that the calculation of the hf susceptibility tensor $\chi(\mathbf{k},\omega)$ cannot be reduced to a calculation of the single-particle Green functions of the HP spin waves.

Although the Green function method for spin operator[6,7] is consistent, its use for describing the properties of a magnetic substance at low temperatures is hardly justified due to its cumbersomeness. In that temperature region it is more convenient to use Bose operators. One must then give up the preference for the representation in which one of the spin operator components is linearly connected with the Bose operators. This is just the situation in the case of the DM representation and the representation suggested in Ref. 8. In Ref. 9 the component $\chi_{+-}(\mathbf{k},\omega)$ of the hf suscep-