Concerning a new type of instability in liquid crystals

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A new type of azimuthal orientational instability, produced by the electric field, has been observed in the nematic phase of liquid crystals at temperatures close to the temperatures of the transition to the smectic phase. A possible explanation of the observed effect is proposed.

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It is known that the usual Williams "domains" (WD) exist in nematic liquid crystals with positive anisotropy of the electric conductivity $\sigma_a > 0$ and with negative anisotropy of the dielectric constant $\epsilon_a < 0$. The geometry of the observed picture is uniquely determined by the initial planar orientation: the WD are parallel to the y axis if the molecules in the unperturbed states are parallel to the x axis (the electric field E is directed along the z axis perpendicular to the xy plane of a layer of thickness l). With decreasing σ_a, the threshold E_W for the onset of orientation modulations $\exp(ikx)$, where $k \sim l^{-1}$, increases abruptly,[1] and no WD are produced at $\sigma_a \leq 0$. Therefore the formation of an inhomo-

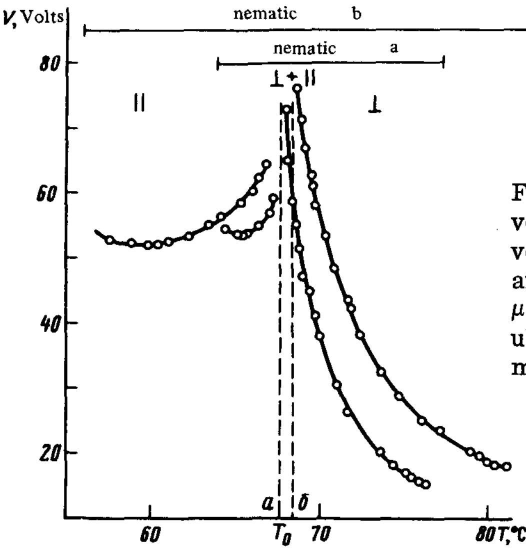

FIG. 1. Dependence of the threshold voltage on the temperature in the investigated samples of compounds (a) and (b): $\omega = 50$ Hz, layer thickness 35 μ, ⊥—Williams domains, ∥—new modulated instability, ⊥+∥—region of mixed domains.

geneous nematic structure in a substance with $\sigma_a \leq 0$ and $\sigma_a < 0$ would offer evidence of the existence of a new physical phenomenon in such an anisotropic medium.

Distortion of the WD in the substance 4-*n*-butoxybenzylidene-4′-*n*-octylaniline (a) as $\sigma_a \to 0$ (with decreasing temperature) was reported in[2]; this points to the possible existence of a new instability mode.

We report here an investigation of the nematic phases of substance (a) and the substance 4-*n*-butoxybenzylidene-4′-*n*-heptylaniline (b).

We have observed that in the region $T < T_0$ a new type of instability appears in these substances, wherein all the perturbations satisfy the law $\exp(iky)$.

Samples in the form of thin layers ($l = 15$—$35\ \mu$) between transparent conducting electrodes were placed in an oven whose temperature could be maintained constant within $\pm$ 0.2 °C, or else varied at a constant rate on the order of 0.5 deg/min. The optical system made it possible to carry out the observation and photography in parallel with the recording of the transparency of the

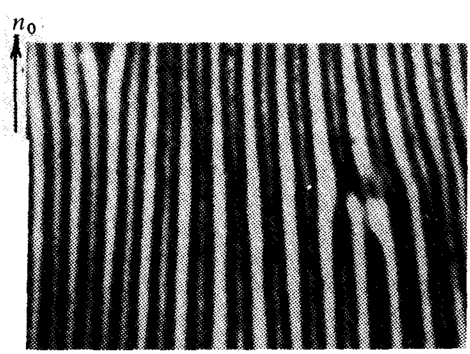

FIG. 2. Modulated structure at $T < T_0$ in sample (b): $n_0\uparrow$ —direction of initial orientation, layer thickness 35 μ, external electric field perpendicular to the plane of the layer.

samples in polarized light. The phase-transition points were registered by an optical method. The threshold voltage at which the instability set in was determined from the dependence of the transparency on the applied voltage at constant temperature. The conductivity of the correspondingly oriented samples was measured in a constant field of 0.1 V.

The temperatures at which nematic mesophases existed in the investigated samples were 63.2—77.6 and 55.7—82.5 °C for (a) and (b) respectively.

It was observed that σ_a reverses sign at temperatures T_0 equal to 67.2 °C for (a) and 68.4 °C for (b). Figure 1 shows the temperature dependence of the threshold of WD formation at $T > T_0$ and of the new type of instability at $T < T_0$ of the investigated compounds. Figure 2 shows a photograph of the orientational modulations at $T < T_0$. In the vicinity of the point T_0 we observed the coexistence of both types of instability (the observed picture is similar to that described in[2]), which became spatially separated in the presence of a small temperature gradient. The temperatures T_0 determined from the reversal of the sign of σ_a and from the dependence of the threshold E_c on T agree within the limits of experimental error. We note that no instability is observed when the investigated substances go over into the smectic phase.

Optical observations have shown that the obtained picture of the fringes is characterized by periodic deviations of the molecules from the x axis in the xy and xz plane, respectively, through angles $\psi \sim \exp(iky)$ and $\theta \sim \exp(iky)$. Such an azimuthal orientational structure can be due, in principle, to different physical factors.

First, the onset of ψ-deviations is possible as a result of the electrohydrodynamic effect if all the molecules in the unperturbed state are oriented at an angle $\theta_0 \neq 0$ in the xz plane.[3,4]

Another cause of the considered phenomenon may be the threshold piezoelectric instability, which is uniquely characterized by the described structure of the orientational perturbations. The threshold of this effect, which is connected with the existence, in the liquid crystals, of a linear connection between the strains ($\partial\psi/\partial y$, $\partial\theta/\partial y$, $\partial\psi/\partial z$, $\partial\theta/\partial z$) and the external electric field $E(t)$, is determined by the expressions

$$E_c \approx \begin{cases} \dfrac{2\pi k}{l\,|e_1 - e_2|\,(1+\mu)}, & \omega < \dfrac{K}{\gamma}\left(\dfrac{\pi}{l}\right)^2 \\[2ex] 4\sqrt{\dfrac{\gamma\omega}{|\epsilon_a|\,(1-|\mu|)}}, & \omega > \dfrac{K}{\gamma}\left(\dfrac{\pi}{l}\right)^2 \dfrac{1}{1-|\mu|} \end{cases}$$

where K is the elastic modulus, e_1 and e_2 are the piezoelectric moduli, γ is the viscosity of the substance, ω is the frequency of the electric field, and

$$\mu = \frac{\epsilon_a k}{4\pi(e_1 - e_2)^2}.$$

We emphasize that this threshold effect, in contrast to[4], exists only at $|\mu| < 1$.

The values of the piezoelectric moduli are determined by the constant dipole moment of the molecules or of the molecular groups, and can change noticeably when the short-range order is restructured in the liquid crystal. In substances of the type investigated in the present paper, the possibility of formation of specific molecular groups in the nematic phase has been noted.[2] This phenomenon arises when the temperature is lowered and the system approaches the point T_{NS} of the transition to the smectic phase, the formation of which depends essentially on the dipole-dipole interaction of the molecules.[1] The observed temperature dependence of $E_c(T)$ can be attributed to the known[1] increase of the elastic moduli and of the viscosity in the immediate vicinity of the point T_{NS}, and also to the suggested decrease of the quantity $|e_1 - e_2|$ far from T_{NS}. The measured frequency dependence $E_c(\omega)$ agrees qualitatively with the presented expression. In the vicinity of the point T_{NS}, according to our estimates, $(e_1 - e_2)^2 \sim 10^{-7}$ cgs esu.

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