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$$a/b \approx \sigma_0 = H_0 MV / kT_0. \quad (2.12)$$

In order for expressions (2.11), from which this equation was obtained, to be valid it is necessary to satisfy the condition  $\sigma_0 \gg 1$ . The minimum lies therefore in the region of low temperatures (or large values of  $V$ ) only if  $b \ll a$ . At other values of the parameters, a minimum also exists and can be obtained graphically. Thus, Fig. 4 shows a plot of the normalized line width  $(\Delta\Omega + \Delta\omega)/(b + a)$  at  $b = 2a$ . It is seen that the minimum is located in this case at  $\sigma_0 = 2$ . The temperature  $T_0$  corresponding to the minimum line width separates the region where the magnetodipole relaxation mechanism predominates ( $T < T_0$ ) from the region of predominance of the internal relaxation mechanisms ( $T > T_0$ ).

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## Electroconvective flow in nematics. Velocities and deformation

A. A. Razumov and E. A. Kirsanov

*State University, Ivanovo*

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The optical parameters, orientational deformation angles, and velocities of electroconvective flow in nematic liquid crystals were determined. Finite values of the velocities, independent of the electric field frequency, were obtained at the instability threshold.

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### INTRODUCTION

Electroconvective flow in nematic liquid crystals is due to the motion of an electric space charge under the action of an external static or alternating field. This flow is associated with an electrohydrodynamic instability described in several papers.<sup>1,2</sup> The experimental situation is usually as follows.

A nematic crystal is in a flat cell between glass plates coated with transparent and electrically conducting films. The long axes of the nematic molecules are oriented parallel to the glass plates along the rubbing axis  $x$ . When a threshold voltage  $V_{th}$  is reached in the conduction regime,<sup>3</sup> a system of parallel vortex tubes, oriented along the  $y$  axis, i.e., at right-angles to the rubbing direction, appears in the nematic. The existence of a velocity gradient results in a hydrodynamic displacement and corresponding deformation of the orientations of the long axes of the molecules. The maximum angle of inclination  $H_0$  of the molecules is observed at the center of a vortex tube; the molecules remain parallel to the cell walls in the regions of ascending and descending fluxes (along the line of sight). Reorientation of the long axes of the molecules results in a periodic variation of the refractive index along the  $x$  axis. This produces a refractive-index gradient which

can focus light whose electric vector is parallel to the  $x$  axis.<sup>4</sup> The focal lines observed in a polarizing microscope are known as the Williams domains.<sup>5</sup>

### EXPERIMENTAL RESULTS

We determined experimentally the parameters of vortex tubes (domains). We observed experimentally<sup>6</sup> a difference between the focal lengths above the regions of ascending and descending fluxes (Fig. 1a). The asymmetry of the optical pattern was due to inequivalence of the convex and concave parts of the pattern of orientation lines, as viewed relative to the light beam vector. A beam passing at an angle to the local optic axis (coinciding, in our case, with the orientation line) became bent, which was not allowed for in the purely gradient "quasiisotropic" model.<sup>4</sup> Our measurements revealed a considerable difference between the upper, average, and lower focal lengths  $f_u$ ,  $f_{av}$ , and  $f_l$ , as well as alternation of the lengths between the lower focal lines  $l$  and  $\lambda-l$  for samples of  $n$ -azoxyanisole (PAA) and  $n$ -methoxybenzylidene- $n'$ -butylaniline (MBBA) in a wide range of voltages and frequencies. Thus, the optical pattern made it possible to determine unambiguously the ascending and descending (relative to the vertical light beam) parts of the flow of the liquid, as well as

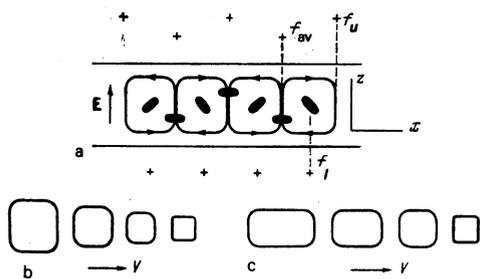


FIG. 1. Schematic representation of electroconvective vortex flow (the crosses represent the optical focusing lines): a) cross section through a cell; b) changes in the vortex tube in the  $a < a_c$  case; c)  $a > a_c$  case.

the centers of the vortex tubes where the velocity gradient was highest.

A complete description of the electrohydrodynamic instability required determination of the velocity of flow of the liquid in the vortex tube. This velocity was related to the density of the space charge and varied from a certain maximum value on the boundary of the vortex tube to zero at its center and near the surfaces of the glass plates. The velocity of flow was estimated usually by finding some average velocity  $v$  equal to the velocity of motion of dust particles dragged by the liquid. Velocity measurements were carried out in this way by Durand *et al.*<sup>7</sup> and by Bertolotti *et al.*<sup>8</sup>

We investigated the velocity of electroconvective flow in the nematic phase of PAA (sample thickness  $d = 100 \mu$ , temperature  $t = 122^\circ \text{C}$ ) and MBBA ( $d = 100 \mu$ ,  $t = 33^\circ \text{C}$ ) in a cell with glass plates coated with  $\text{SnO}_2$  films. The cell was placed in a thermostatted chamber controlled by a RT-049 regulator. Temperature was measured with a V7-16 ohmmeter and a KTM-5 thermistor placed on a step in the cell in direct contact with the investigated nematic. The time of revolution of an impurity particle along a closed trajectory between the two upper focal lines was measured with an F5080 frequency meter and chronometer. At voltages slightly above the threshold value ( $V - V_{th} \sim 1.5 - 2$  volts) we found that linear domains (parallel vortex tubes) became unstable and secondary flow took place. The optical pattern of the domains exhibited periodic perturbations, which made it difficult to measure the velocity. Stable results were obtained in a narrow range of voltages near  $V_{th}$ ; at a fixed frequency we obtained four to five values of the velocity as a function of the voltage.

The velocities corresponding to different frequencies of the electric field were calculated using the vortex flow models. Figure 1a shows a model of electroconvective flow in which the vortex tubes are bounded by the maximum-velocity lines. It is natural to assume that impurity particles move along trajectories coinciding with the vortex "boundaries." In fact, visual observations indicate that the motion of particles is vertical (along the  $z$  axis) in the region of the upper focal lines. In this model the calculated velocity depends on the assumed shape of the vortex tube. If we postulate that the dimensions of the tube are the same along the  $x$  and  $z$  axes, we find that the trajectory of a particle is

$\sim 4a$ , where  $a = \lambda/2$  (Fig. 1a). Then,

$$v = 4a/t, \quad (1)$$

where  $t$  is the revolution time of a particle traveling on a closed trajectory. The relative error in our determinations of the velocity did not exceed 3%.

When the threshold value  $V = V_{th}$  was reached, the appearance of focal lines of the domains was accompanied by simultaneous circular motion of the impurity particles in the  $xz$  plane. The particle revolution times in neighboring vortex tubes were the same within the limits of the experimental error.

An analysis of the graphs (Figs. 2a and 2b) indicated that in the case of MBBA the following dependence was well satisfied:

$$v - v_0 = B(\omega)(V^2 - V_0^2), \quad (2)$$

where  $B(\omega)$  decreases uniformly on increase in the electric field frequency;  $v_0$  and  $V_0$  are constants independent of the frequency. The velocity  $v_c$  of the electroconvective flow at the electrohydrodynamic instability threshold was a finite quantity practically constant for all the investigated particles. A similar situation was also observed in the case of PAA, with the exception of low voltages near  $V_{th}$  (Fig. 2b). The domain period  $\lambda$  measured with an eyepiece micrometer decreased on increase of the voltage and this was due to reduction of the vortex tube dimensions along the  $x$  and  $z$  axes (Fig. 1b).

The characteristic deviations from the straight lines for PAA could be explained by introducing a maximum critical vortex size along the  $z$  axis. For  $a > a_c$ , the vortex tubes were no longer cylindrical (Fig. 1c). An increase in the voltage did not affect the size of the vortices along the vertical  $a_c$ , but  $a$  decreased to  $a_c$ . When this critical dimension was reached, the vortex continued to decrease uniformly along both directions. For the vortex with the critical thickness  $a_c$  the average flow velocity was

$$v = (2a + 2a_c)/t. \quad (3)$$

In the case of PAA we selected the critical width corresponding to the velocity  $v$  just in front of the inflection of the function  $v(V^2)$ . The velocities for  $a > a_c$  calculated from Eq. (3) were found to lie on the same line in accordance with the expression  $v - v_0 = B(\omega)(V^2 - V_0^2)$ .

On this basis we can represent the vortex flow in PAA

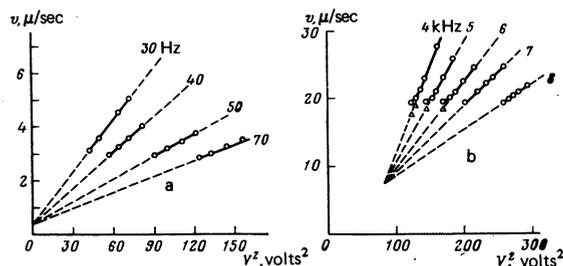


FIG. 2. Dependences of the velocity on the voltage across the cell: a) MBBA; b) PAA. The circles represent the velocities calculated from Eq. (1) and the triangles those calculated from Eq. (3).

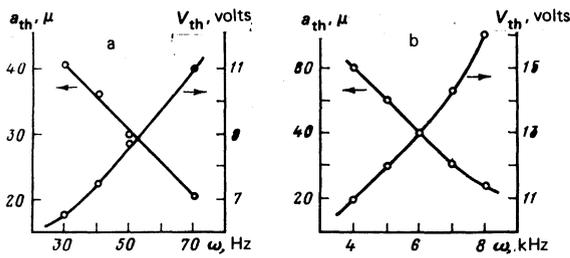


FIG. 3. Dependences of the threshold voltage  $V_{th}$  and diameter of a vortex tube  $a_{th}$  on the electric field frequency: a) MBBA; b) PAA.

as follows: first a cylindrical vortex appears of size  $\sim a$  and the velocity is  $v_c = 4a/t$ . The subsequent formation of a domain system (10–100 msec) alters the shape of the vortex, the flow near the walls rapidly slows down, and the average velocity decreases to  $v = (2a + 2a_c)/t$ . The change from  $a < a_c$  to  $a > a_c$  thus alters the calculation formula for the average flow velocity. The results of calculations are presented for PAA in Fig. 2b. The finite velocity near the electrohydrodynamic instability threshold should give rise to finite values of the orientational deformation of the nematic.

The angle  $\Theta_0$  of the maximum deflection of a molecule at the center of a vortex tube was determined by the Penz method<sup>4</sup> for the focal lengths. We measured the parameters of linear vortex tubes (domains) in the conduction regime in a sample of PAA  $d = 20 \mu$  thick (Fig. 3). Three focal lengths and the domain period  $\lambda$  were found as a function of the threshold voltage  $V_{th}$  when the external field frequency was reduced. The focal lengths  $f_u$ ,  $f_{av}$ , and  $f_l$  decreased on increase in the threshold voltage  $V_{th}$  (Fig. 4). An increase in the applied voltage  $V$  at a fixed frequency resulted in a more rapid reduction in the focal lengths, as represented by the dashed lines in Fig. 4.

The birefringence of a sample, causing splitting of the upper focal lengths, was ignored in Refs. 4 and 9. Therefore, in calculations based on the "quasiisotropic" Penz formula<sup>4</sup> we took the average value of the upper focal length  $\bar{f} = \frac{1}{2}(f_u + f_{av})$ . Then, the angle of inclination of the molecules in the center of the vortex tube was given by

$$\Theta_0 = [(n_{\parallel} - n_{\perp}) k d \bar{f}]^{-1/2}. \quad (4)$$

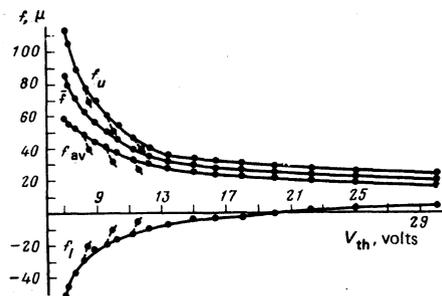


FIG. 4. Focal lengths for three positions of optical focusing by domains  $f_u$ ,  $f_{av}$ , and  $f_l$  plotted as a function of the threshold voltage  $V_{th}$  at  $t = 121^\circ\text{C}$ .

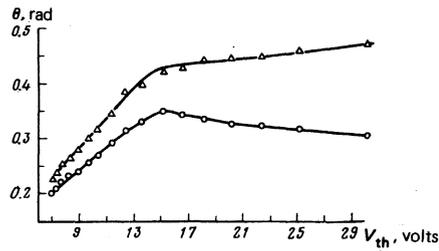


FIG. 5. Dependence of the angle of inclination of the molecules  $\Theta_0$  on the threshold voltage  $V_{th}$ . The circles represent the results calculated from Eq. (4) and the triangles those calculated from Eq. (5).

Here,  $n_{\parallel} - n_{\perp}$  is the birefringence of an oriented sample;  $k$  is the wave vector of the perturbation along the  $x$  axis;  $\bar{f}$  is the average focal length measured from the center of the nematic layer.

The dependences  $\Theta_0(V_{th})$  were found to consist of two regions (Fig. 5). On increase of  $V_{th}$  from 7 to 15 V the angle  $\Theta_0$  increased from 10 to 20°, and between 15 and 30 V the value of  $\Theta_0$  decreased from 20 to 17.5°. It was difficult to account for the maximum at 15 V.

More reliable results were obtained using the model of a vortex tube in which the dimensions of the vortex along the  $z$  axis are much less than the thickness of the sample. When the orientational deformation occurs only inside a vortex tube bounded by the maximum-velocity lines, the optical focusing conditions are modified somewhat. The effective thickness of the nematic layer becomes  $\lambda/2$  and the angle is given by

$$\Theta_0 = [1/2 \lambda (n_{\parallel} - n_{\perp}) k^2 \bar{f}]^{-1/2}. \quad (5)$$

The values of  $\Theta_0$  calculated from Eq. (5) increase practically linearly from 13 to 24.5° on increase of  $V_{th}$  from 7 to 15 V. Between 15 V and 30 V the angle  $\Theta_0$  increases slowly (from 24.5 to 26.5°). Thus, the angles of inclination of the molecules  $\Theta_0$  relative to the initial orientation are finite and fairly large already at the actual measured electrohydrodynamic instability threshold.

## DISCUSSION OF RESULTS

It should be pointed out that these were the first-ever determinations of the velocity of flow and angles of orientational deformation at various threshold voltages and frequencies. The experimental dependences of the velocity of flow in the case of electroconvective cells of hexagonal shape were reported by Durant *et al.*<sup>7</sup> for a static field applied to MBBA. They obtained a dependence of the type

$$v = A(V^2 - V_{th}^2),$$

where  $A$  is a constant. According to this formula, the velocity of electroconvective flow should gradually decrease to zero on approach of  $V$  to  $V_{th}$ . A similar expression for a static field was obtained by Bertolotti *et al.*<sup>8</sup> in the case when the field vector  $E$  was parallel to the cell plates. The average velocity of flow obeyed the dependence  $v \propto (E^2 - E_{th}^2)$ .

Carroll<sup>9</sup> used the Helfrich-Orsay theory<sup>3</sup> to calculate the dependence of the velocity  $v$  and angle  $\Theta_0$  for a small excess over the threshold in a static field. Expanding the moments of the forces as a series in  $\Theta^2$ , he obtained the following relationships in a simple nonlinear approximation:

$$\Theta_0^2 \propto (1 - V_{th}^2/V^2), \quad v \propto \Theta_0 E^2.$$

Clearly, the approach to the threshold should give near-zero values of the velocity and angle in the immediate vicinity of  $V_{th}$ . The experimental dependence  $\Theta_0(V)$  was in good agreement with the calculations. In all these investigations<sup>7-9</sup> the measured or calculated values of  $v$  and  $\Theta_0$  tended to zero on approach of  $V$  to  $V_{th}$ .

Our experiments showed that the quadratic dependence of the velocity on the voltage was retained in the case of an alternating electric field but was more complex. The velocity  $v_c$  at  $V_{th}$  was a constant independent of the field frequency. In the case of PAA this velocity was  $v_c \approx 20 \mu/\text{sec}$ ; in the case of MBAA, we obtained  $v_c \approx 3 \mu/\text{sec}$ . The finite velocity  $v_c$  correspond to finite and fairly considerable deformation angles  $\Theta_0$  (from 13 to 26°) for values of  $V_{th}$  in the investigated frequency range. Thus, the results of experimental determinations of the velocities and deformation angles were in mutual agreement and indicated considerable nonlinear

effects even at the threshold of the electrohydrodynamic instability in the conduction regime.

These effects, ignored in most investigations, may be responsible for the growth of the initially small perturbation to the relatively large values of the velocity and deformation found by us. Our results differ considerably from those published earlier and it is not yet clear whether they can be explained within the framework of the existing theories of the electrohydrodynamic instability.

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## Phase transitions in a cubic crystal with dipolar forces and an anisotropic correlation function

A. I. Sokolov and A. K. Tagantsev

V. I. Ul'yanov (Lenin) Leningrad Electrotechnical Institute  
*Zh. Eksp. Teor. Fiz.* **76**, 181-192 (January 1979)

The static critical behavior of a cubic ferroelectric (ferromagnet) is studied taking into account both the dipolar and the crystal anisotropy of the correlation function of the order-parameter fluctuations. It is shown that the interaction of the critical fluctuations can lead in this case to a curious effect—a phase transition to an ordered phase that is not energetically the most favorable from the standpoint of the Landau theory. The form of the diagram of states in the coordinates of inverse correlation length, anisotropy of the correlator, and anisotropy of the anharmonicity is established in the critical region. The features that arise in the diagrams of states of the system as a result of the interaction of the fluctuations with the anisotropic spectrum are discussed.

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### 1. INTRODUCTION

The problem of phase transitions in cubic crystals can be regarded at the present time as classical for the theory of static critical phenomena. Indeed, several dozen papers have been devoted to the critical thermodynamics of systems with cubic and hypercubic symmetry,<sup>1</sup> and the most important of the results obtained have already appeared in reviews and books.<sup>1-4</sup> While studying the simplest model of this type, Wilson and

Fisher<sup>5</sup> encountered for the first time two very interesting phenomena, having, as was made clear later, an extremely general character—the change of the order of the phase transition in an anisotropic system under the influence of the critical fluctuations, and what it is now customary to call asymptotic symmetry or isotropization. Models with generalized cubic symmetry have been investigated in detail by various methods based on the ideas of the renormalization group, and the corresponding problem has succeeded in becoming something