Threshold characteristics of electroconvective flow in nematics

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The characteristics of the electrohydrodynamic instability in nematic *p*-azoxyanisole are measured at various simple thicknesses. A nonlinear dependence of the threshold domain voltage on the thickness and an exponential dependence of this voltage on the external-field frequency are observed. Empirical relations for the threshold characteristics are determined for various frequency intervals between 20 Hz and 44 kHz. A comparison is made with the threshold characteristics of the instability in methoxybenzylidene-butylaniline.

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INTRODUCTION

Electrohydrodynamic instability in liquid crystals has been investigated in quite sufficient detail.[1] Particular attention was paid to this instability in nematics with planar orientation and negative dielectric anisotropy.[2–4] The theory of the conduction regime of the electrohydrodynamic instability was developed on the basis of the Carr-Helfrich model,[5,6] i.e., on the basis of an anisotropic mechanism of this instability. A recently developed theory has explained well the experimental dependence of the threshold voltage V_{thr} on a number of parameters of the material.[4]

The frequency dependences of $V_{\text{thr}}(\nu)$ and of the period $\lambda(\nu)$ of the electroconvective domains were determined in a number of studies.[3–6,9] The dependence of the parameters of the electrohydrodynamic instability on the thickness d of the sample was investigated in Refs. 5 and 8. In these papers, the threshold dependences were considered in the limiting cases of the low-frequency conduction regime and of the high-frequency dielectric regime. For the conduction regime, the known Orsay formula[6,8] was derived in the one-dimensional approximation:

$$V_{\text{thr}}^2(\omega)=\frac{V_0^2(1+\omega^2\tau^2)}{\zeta^2-(1+\omega^2\tau^2)}, \qquad (1)$$

where τ is the dielectric-relaxation time, ζ is the Helfrich constant, and V_0 is a constant that depends on the parameters of the material under the assumption $\lambda \approx 2d$ (d is the sample thickness). It follows from (1) that the conduction regime is limited by a critical frequency

$$\omega_c=2\pi\nu_c=(\zeta^2-1)^{1/2}/\tau. \qquad (2)$$

Equation (1) was used to plot[8] at $\zeta^2 = 4.5$ a curve that agreed well with the experimental data for methoxybenzylidene-butylaniline (MBBA). The comparison was made against seven experimental points in the interval from 20 to 80 Hz. It is noted already in that reference, however, that the optimal value of ζ for the theoretical curve closest to the experimental points is smaller for thin samples ($d < 30$ μm) and larger for thick ones ($d > 100$ μm).

The conditions for the calculation of the threshold of the conduction regime were the relations $T \gg \tau$ and $\omega\tau < 1$ (T is the curvature relaxation time). Under these conditions the space charge q is modulated at a frequency ω, and the curvature of the orientation is practically constant. The electroconvective flow has an almost constant velocity direction independent of that of the external field.

Gruler and Meier[10] presented for *p*-azoxyanisole (PAA) and MBBA plots of $V_{\text{thr}}(\nu)$ without any theoretical interpretation whatsoever. Kashnow and Cole[9] described the $V_{\text{thr}}(\nu)$ dependence for well purified MBBA (~5×10^{11} Ω-cm). They, however, defined the critical frequency as the frequency at which the conduction and dielectric regimes "cross" (~9 Hz). Carroll[11] measured $V_{\text{thr}}(\tau)$ for MBBA but did not indicate in his paper the method used to determine ν_c (~600 Hz). Meyerhofer and Sussman[12] have observed that the Orsay theory[6] does not describe well enough the conduction regime in MBBA at low frequencies ($\nu < 10$ Hz), but at a certain choice of V_0 and ζ^2 the $V_{\text{thr}}(\nu)$ curve agrees well with the experimental points at frequencies from 10 to 300 Hz. Grebenkin *et al.*[7] obtained the $V_{\text{thr}}(\nu)$ dependence for *p*-butyl-*p*'metoxyazoxybensene of varying thickness and electric conductivity. Experimental results and tabulated data were used in the calculation. The calculated values of ν_c were smaller by a factor 4–5 than the critical frequencies suggested by the experimental curves. Matsumoto and Kawamoto[14] investigated a mixture of nematics made from Schiff bases, in which the electric conductivity was varied by varying the ion impurity. The critical frequency $\nu_c (\propto \rho^{-1})$ was determined from the crossing of the conduction and dielectric regimes. The functional $V_{\text{thr}}(\nu)$ dependence was not analyzed.

An attempt was made[14] to determine ν_c directly from the experimental curve. Equation (1) was transformed into

$$(Y^2-1)/\omega^2=\tau^2+Y^2/\omega_c^2, \qquad (3)$$

where $Y = V_{\text{thr}}/V_0$. In modified coordinates $[(Y^2-1)/\omega^2$ and $Y^2]$, the plot of Eq. (1) is a straight line. A sufficiently linear dependence was obtained for MBBA, and ν_c was determined from the slope (570 Hz).

Goscianski[15] eliminated the restriction due to the constancy of the wave vector k of the perturbation, and transformed Eq. (1) into

$$V_{\text{thr}}^2(\nu)=V_{\text{thr}}^2(0)\,\frac{k^2(\nu)}{k^2(0)}\,\frac{1+(\zeta^2-1)\nu^2/\nu_c^2}{1-\nu^2/\nu_c^2}. \qquad (4)$$

Goscianski, however, did not check the agreement between this equation and the experimental frequency dependence. He determined ν_c from the crossing of the two regimes.

It has been stated[16,17] that the critical frequency ν_c is equal to the reciprocal dielectric relaxation time ($4\pi\sigma/\varepsilon$). In this case the Helfrich parameter ζ^2 is always equal to 2, in contradiction to its definition in terms of the material parameters of the medium.

The absence of a dependence of V_{thr} on the thickness d, at least at low frequencies, we noted by Penz[18,19] for PAA and by a number of others[6,8] for MBBA and similar compounds. At the same time, Penz observed no frequency dependence of V_{thr} whatever in the 50–500 Hz interval.

It follows from the foregoing brief survey that the $V_{\text{thr}}(\nu)$ frequency dependences obtained in experiments were compared with sufficient rigor with the theoretical equations only in the case of MBBA. There is also a different approach to the determination of the critical frequency ν_c. At the same time there is a complete absence of empirical relations between V_{thr} and ν, obtained directly from the experimental data without invoking theoretical models. To reveal such relations we have investigated the classical nematic crystal PAA in the frequency interval from 20 Hz to 40 kHz, i.e., in a frequency range of three orders of magnitude.

EXPERIMENTAL RESULTS

We have separated three qualitatively different sections on the $V_{\text{thr}}(\nu)$ threshold curve: a plateau, a drop, and a rise (Fig. 1). The arrangement of these sections was the following: a high-frequency rise of V_{thr} (11–44 kHz), a linear plateau (1–7 kHz), and a low-frequency drop (20–200 Hz). In the intervals between these sections, the experimental points occupied certain intermediate positions.

In the plateau region, V_{thr} increased slightly with increasing thickness d and was independent on the frequency ν. For thicknesses from 20 to 140 μm in the temperature interval from 118 to 132° C, the threshold voltage V_{thr} ranged from 7 to 10 V.

On the rise section, the threshold voltage increased substantially with the sample thickness (Fig. 2). The threshold curves do not differ outwardly (Figs. 1 and 3) from the usual conduction-regime frequency dependences described in Refs. 7–13.

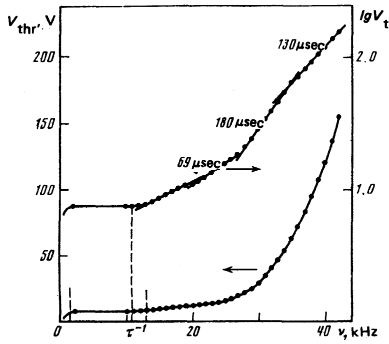

FIG. 1. Threshold voltage vs. electric-field frequency in PAA (d = μm, 132 °C, $\rho = 2\times10^8$ Ω-cm, $\tau \sim 90$ μsec). The coefficients α are indicated on the upper curve.

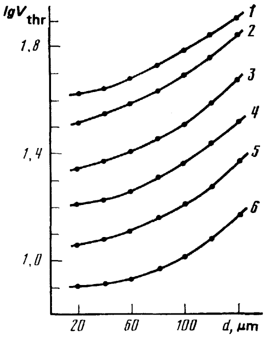

FIG. 2. Dependence of the threshold voltage on the sample thickness at various frequencies: 30 kHz (1), 28 kHz (2), 24 kHz (3), 20 kHz (4), 16 kHz (5), and 12 kHz (6) (PAA, 122 °C).

Lowering the frequency below 1 kHz led gradually to a low-frequency drop of the threshold voltage (Fig. 1). For the PAA sample (d = 30 μm, 120° C), V_{thr} decrease from 9 V (on the plateau) to 2 V (at 20 Hz). The period λ of the electroconvective domains increased at the same time from 50 μm (on the plateau) to 178 μm (at 20 Hz).

To compare the characteristics, we obtained the threshold $V_{\text{thr}}(\nu)$ dependence for MBBA (Fig. 4). The threshold curves on Figs. 1, 3, and 4 are quite similar in shape.

DISCUSSION OF RESULTS

Our main purpose was to determine the empirical dependences of the threshold parameters of the electroconvective flow directly from the experimental data. The choice of the characteristic frequency intervals within the electrohydrodynamic instability regime was determined by the following considerations.

The most strongly pronounced region on the threshold curve is the linear plateau. The high-frequency rise section is separated from the plateau by an intermediate frequency interval 4 kHz. We can therefore exclude a possible influence of the plateau regime on the high-frequency instability region. On the other hand, the

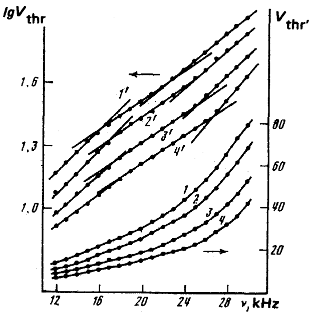

FIG. 3. Frequency dependence of threshold voltage at various sample thicknesses: 140 μm (1), 120 μm (2), 80 μm (3), and 20 μm (4) (PAA, 122 °C); 1′ to 4′ are curves 1 and 4 replotted in a logarithmic scale.

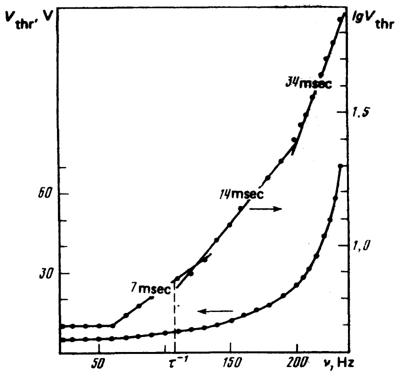

FIG. 4. Dependence of the threshold voltage on the frequency in MBAA ($d = 25\,\mu$m, 23 °C, $\rho \sim 2\times10^{10}\,\Omega$-cm, $\tau \sim 9$ msec). The coefficients α are marked on the upper curve.

experimental data are far enough from the "dielectric" regime, which is usually not reached in experiments with PAA.[18,19] Therefore the influence of the dielectric regime on the character of the instability in the rise region can likewise not be particularly strong.

The low-frequency drop regime (20–200 Hz) is separated from the plateau by an intermediate interval of 800 Hz.

We note immediately that the dependence of the electrohydrodynamic-instability parameters on the frequency and on the sample thickness have in these intermediate sections a complicated "intermediate" character. Empirical relations in sufficiently simple form have been established in the characteristic drop, plateau, and rise intervals.

The absence of double modulation of the light and the circular motion of the impurity particles in the entire frequency interval from 20 Hz to 40 kHz point to an anisotropic mechanism of the Carr-Helfrich instability,[5,6] connected with the modulation of the space charge. $V_{thr}(\nu)$ curves similar to those shown in Figs. 1 and 3 are usually interpreted within the framework of the Orsay theory for the conduction regime. However, neither Eq. (1) nor the modified Eq. (3) is in satisfactory agreement with experiment. The choice of the empirical equations has shown that the best agreement with the experimental point is given by an exponential dependence of the $\exp\alpha\nu$ type. Accordingly, all the curves investigated by us are divided in the rise region into three matched sections (Figs. 1 and 3). It is clear that the experimental shape of the curve and a functional dependence of the type (1) cannot be observed simultaneously.

Examination of the voltage-rise threshold curves raises the natural question of the value of the critical frequency ν_c that limits the conduction regime within the framework of the Orsay theory.[6,8] It was shown in the Introduction that ν_c is a quantity that can be calculated but not measured directly from the threshold dependences. In the case of an exponential dependence of V_{thr} on ν, the concept of a critical frequency that limits the conduction regime loses meaning. The presence of one electrohydrodynamic instability regime of one type or another is determined then only by the threshold. This last argument is confirmed by the well known fact that the two regimes (dielectric and conduction) cross and are superimposed in a certain frequency interval. Our results show that Eqs. (1) and (3) do not hold for nematic PAA and that the concept of the critical frequency of the conduction regime becomes meaningless. Further examination of the control curve for MBBA has shown a similar exponential dependence on the three successive frequency intervals (Fig. 4).

In a nematic PAA, in contrast to MBBA at high frequencies, the threshold voltage increases abruptly with the sample thickness (Fig. 2), likewise contradicting the traditional form of the Orsay theory.[6,8] An essential role in the interpretation of threshold dependences is played by the dielectric relaxation time τ. For PAA, an estimate of the resistivity yields $\rho \sim 2\times10^{8}$ Ω-cm and an average dielectric constant $\varepsilon \sim 5$. Then $\tau \sim 90\times10^{-6}$ sec and $\tau^{-1} \sim 11$ kHz. It is clear that $\omega\tau$ is certainly larger than unity on the rising section. The condition $\omega\tau \lesssim 1$ is satisfied only at frequencies lower than 3 kHz, i.e., on the lower part of the plateau. For MBBA ($\rho \sim 2\times10^{10}$ Ω-cm, $\varepsilon \sim 5$) we obtain $\tau \sim 9\times10^{-3}$ sec and $\tau^{-1} \sim 110$ Hz. Actually the entire section where V_{thr} increases with frequency is located above τ^{-1}. The condition $\omega\tau \leqslant 1$ is not valid starting already with 20 Hz.

The coefficients α in the exponential form of the curve were determined from slope of the plot in coordinates $\log V_{thr}$ and ν. For PAA ($d = 20$ μm, 132° C, $\rho = 2\times10^{8}$ Ω-cm) the values obtained for the three linear segments were

$$\alpha_1 \sim 70\cdot10^{-6}\,\text{sec},\ \alpha_2 \sim 180\cdot10^{-6}\,\text{sec}.\ \alpha_3 \sim 140\cdot10^{-6}\,\text{sec}.$$

For MBBA ($d = 25$ μm, 23° C, $\rho = 2\times10^{10}$ Ω-cm) they were

$$\alpha_1 \sim 7\cdot10^{-3}\,\text{sec},\ \alpha_2 \sim 14\cdot10^{-3}\,\text{sec},\ \alpha_3 \sim 34\cdot10^{-3}\,\text{sec}.$$

It should be noted that α has in both cases a value close to the dielectric-relaxation time τ. Penz[19] mentions a possible connection between the shapes of the threshold curves and the time of dielectric relaxation of a "liquid-crystal" capacitor. Inasmuch as in the conduction regime the space charge q is modulated, it is probably more correct to speak of relaxation of the space charge. It can be assumed that $V_{thr} \sim \exp\tau_q\nu$, where τ_q is the space-charge relaxation time under conditions of electroconvective flow. It is quite probable that $\tau_q \sim \tau$. In this case, when ρ is increased the $V_{thr}(\nu)$ curves will shift towards the low-frequency region. This can be easily seen from a comparison of the PAA and MBBA samples, upon addition of ion impurities to MBBA,[13] when the sample is cooled,[20] and when samples with different degrees of purification are investigated.[10]

In the low-frequency regime, where the condition $\omega\tau \leqslant 1$ is actually satisfied, empirical relations of the type $V_{thr}^2 \propto \nu$ and $V_{thr} \propto \exp\beta\nu$ were observed. This modification of the electrohydrodynamic instability must be considered separately. We note here that the strong decrease of $V_{thr}(\nu)$ at frequencies lower than 10 Hz was observed in MBBA.[12] In this sense there is a correlation between the PAA and MBBA curves.

We can thus draw the following conclusions. In nematic PAA the threshold $V_{thr}(\nu)$ curve has a complicated structure and can be divided into three characteristic sections: plateau, drop, and rise. A number of simple empirical curves describe these sections of the threshold curve. At high frequencies the threshold voltage reveals an exponential frequency dependence (exp $\alpha\nu$) that makes meaningless the concept of a critical frequency that limits the conduction regime. In the rising sections of V_{thr}, the condition $\omega\tau > 1$ is satisfied. The coefficients α are comparable in magnitude with the dielectric relaxation time. (Similar results were obtained in the rise region form an MBBA sample.) In contrast to MBBA, in nematic PAA the threshold voltage increases substantially with increasing sample thickness.

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