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## Liquid Crystals

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# Electrohydrodynamic instabilities in nematic liquid crystals with large positive dielectric anisotropy

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### Electrohydrodynamic instabilities in nematic liquid crystals with large positive dielectric anisotropy

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The conditions for initiation of EHD instability at frequencies of 50–150 Hz and for DC for eight nematic materials having a positive dielectric anisotropy ( $\Delta \varepsilon > 3$ ) have been experimentally studied. The dependences of the threshold voltage for the EHD instability on electrical conductivity, as well as on the translational and rotational viscosities of the LC materials have been found. For the measurements substances with strongly differing (from  $10^{-9}$  to  $5 \times 10^{-7} \Omega^{-1} m^{-1}$ ) electrical conductivities and profound temperature dependence of the viscosities were chosen. Analysis of the results based on the isotropic model of EHD instability shows good agreement between theory and experiment.

#### 1. Introduction

The electrohydrodynamic (EHD) instability effect in nematic liquid crystals is rather well known. Hydrodynamic flows arising in a nematic due to moving electric charges and the threshold conditions for the occurrence of flows are determined by a number of physical characteristics of the material, especially by the dielectric anisotropy  $\Delta \varepsilon$ . Until now, liquid crystals (LCs) exhibiting low dielectric anisotropy, both positive and negative in sign, have been most often investigated. As for LCs having a high (of the order of 5–15) positive  $\Delta \varepsilon$ , observations of EHD instabilities and the conditions for their initiation in such LCs are rather few in number [1–3], though these LCs are known to be widely used in practice.

Two mechanisms for the initiations of EHD instabilities in the above mentioned LCs are known. In principle, for an electrooptic cell with a planar orientation of molecules, an instability of the Carr-Helfrich type [4] may arise at any value of  $\Delta \varepsilon$ . But for LCs having a high, positive  $\Delta \varepsilon$ , prior to the advent of the EHD instability, a Fréedericksz transition takes place at a voltage of about 1 V. As a result, the sample becomes quasi-homeotropically oriented and in such an orientation the initiation of the Carr-Helfrich instability is impossible. Thus, it is evident that for LCs with  $\Delta \varepsilon > 0$ , the Carr-Helfrich mechanism is not realized whatever the geometries of the experiment. The second mechanism of instability [1], the so-called isotropic mechanism (evidence for the mechanism was first found for isotropic liquids [5]) is realized for an initially homeotropic orientation of the LC molecules and for  $\Delta \varepsilon > 0$ .

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In the case of a direct current (DC), as well as for a low frequency alternating current (AC), the threshold voltage  $U_{LF}$  needed for the initiation of EHD instabilities is given by [1]

$$U_{\rm LF}^2 \sim A\eta \sigma \varepsilon^{-2},\tag{1}$$

where  $\eta$  is the viscosity, A is a numerical coefficient, and  $\sigma$  is the electrical conductivity. The threshold voltage for frequencies above the critical value  $v_c$  is determined by the viscosity  $\eta$  and the field frequency v, but is independent of the conductivity  $\sigma$ 

$$U_{\rm HF}^2 \sim A' \eta v \varepsilon^{-1}.$$
 (2)

Relation (1) was corroborated qualitatively making use of MBBA for which the conductivity ( $\sigma$ ) and  $\Delta \varepsilon$  were varied by means of doping ( $\sigma$  between  $10^{-9}$  and  $5 \times 10^{-7} \Omega^{-1} m^{-1}$  and  $\Delta \varepsilon$  between 0 and +3).

The present work was aimed at an experimental study of the validity of the isotropic model, i.e. whether relations (1) and (2), resulting from the model, account for the analysis of EHD instability initiation in nematic substances having different chemical structures. It is evident that using various LC substances with strongly differing conductivities and temperature variations of viscosity, we may attempt to obtain a general idea of the process of initiation of EHD instabilities in the substances explored.

#### 2. Experimental

The LC materials investigated in this work are characterized in the table; together with the pure, single component nematics enumerated 1–5, we also studied 5CB with the addition of a dopant (6) which decreased the conductivity, and also two multicomponent LC mixtures (7 and 8). The large values of dielectric anisotropy are due to the presence in the molecules of a polar CN group for which the dipole moment lies along the longitudinal axis of the molecule. The conductivities of the substances explored vary greatly, from  $1.1 \times 10^{-8}$  to  $850 \times 10^{-8} \Omega^{-1} m^{-1}$ .

For the observation of EHD instability, planar glass cells with a thickness of the LC layer of  $50 \,\mu\text{m}$  were used. The initial orientation of the nematic was planar. With increasing voltage, a Fréedericksz transition with a threshold voltage of about 1 V was found for all the investigated samples. Further increase in voltage up to U = 1-5 V (DC) and up to a few tens of volts (AC, v > 50 Hz) in quasi-homeotropically oriented (due to a Fréedericksz transition) samples results in the appearance of light dots against the dark background (crossed polarizer). The dots were thinly distributed and became more and more mobile with increasing voltage.

#### 3. Results

Threshold values  $U_{LF}$  corresponding to the advent of the instabilities in the samples explored are given in figure 1 (for DC) and figure 2 (for 50 and 100 Hz). The characteristic behaviour of the threshold voltage in different substances with respect to the voltage mode (direct or alternating) differs. For AC (see figure 2),  $U_{HF}$  is seen to increase with decreasing temperature within the temperature range of existence of a given nematic. Moreover, a systematic increase of  $U_{HF}$  from sample 2 ( $U_{HF} \le 70$  V) to sample 3 ( $U_{HF} \ge 70$  V) with decreasing electrical conductivity is observed. The threshold values for DC (see figure 1) for different LCs lie within 2–4 V, though the electrical conductivity varies from sample 2 to sample 3 by about 3 orders of magnitude. As follows from the above reasoning, we should make use of the isotropic mechanism model to analyse the observed EHD instability.

		Transition temperatures/°C		Conductivity/ × $10^8 \Omega^{-1} m^{-1}$		
	Molecular structure	$T_{\rm CN}$	T <sub>NI</sub>	σ	Δε	3
1	C <sub>0</sub> H <sub>13</sub> O-{COO-{CN	71	83	420-850	11.3	14·8
2	C <sub>0</sub> H <sub>17</sub> O-CN	89	128	100-430	9.8	12·9
3	CaH11	15	35	5–11	9.0	11·8
4		33	43	17–26	8.6	16.3
5		30	55	13–16	3.0	8·5
6	Sample 3+dopant	15	35	1–3	9·0	11.8
7	LC mixture	0	67	_	7.5	9.0
8	LC mixture	- 30	71	_	9.3	8∙4

Electrophysical characteristics of the LC materials investigated.

The value of  $\Delta \varepsilon$  is given for a temperature 3°C below  $T_{\rm NI}$ .

 $\varepsilon$  is constant (to an accuracy of  $\pm 2$  per cent), within the temperature range corresponding to the existence of a nematic phase.



Figure 1. Threshold voltage  $U_{LF}$  for initiation of EHD instability (DC) versus temperature. Numbering of the curves corresponds to the sample numbers in the table.

The numerator in expression (1) for  $U_{\rm LF}$  contains the factor  $\sigma\eta$ . According to the well-known law for liquids with an ionic mechanism of conductivity (nematic LCs belong to this class of liquids) the product of viscosity and conductivity is temperature independent:  $\sigma\eta \sim \text{constant}$ . The dielectric permittivity in the denominator of (1), which we assume to be the mean dielectric permittivity of an anisotropic nematic, is equal to  $\varepsilon = (\varepsilon_{\parallel} + 2\varepsilon_{\perp})/3$  (see the table) and varies with temperature by no more than a few per cent. Therefore, the right-hand part of equation (1) and hence  $U_{\rm LF}$  should be slightly temperature dependent. This is evidenced by the experimental data of figure 1.

In the case of AC ( $v \ge 50$  Hz), the temperature dependence of  $U_{\rm HF}$ , according to equation (2), should be governed by the temperature variations of the viscosity:  $U_{\rm HF} \sim \eta^{-0.5}$ . Since EHD flows yield involved translational as well as rotary motions of



Figure 2. Threshold voltage  $U_{\rm HF}$  for initiation of EHD instability (AC) for frequencies 50 Hz (1–6) and 100 Hz (1'-6') versus temperature.



Figure 3. Ratios  $U_{\rm HF} \gamma_1^{-0.5}$  (dark points) and  $U_{\rm HF} \eta^{-0.5}$  (light points) as functions of temperature for the LC samples studied.

the LC molecules, we may suggest that the threshold voltage should be associated with the translational viscosity  $\eta$ , as well as with the rotational viscosity  $\gamma_1$ . The translational viscosity was measured using a capillary method for the three LC materials, **3**, **7** and **8**. The rotational viscosity was obtained for samples **3** and **8** and for **1**, **2** and **5**. Measurements of  $\gamma_1$  were performed making use of the study of the free relaxation time of the director in a planar layer of LC following the procedure outlined in [6]. We found that the rotational viscosity of the samples lie in the interval from 0-15 to 1-1 P and increase with decreasing temperature as  $\gamma_1 \sim \exp(W/kT)$ . Such a temperature dependence of  $\gamma_1$  is in accordance with the dependence of  $U_{\rm HF}$  (see figure 2). It follows from relation (2), valid for fairly high frequencies, v, for which the threshold voltage is no longer related to the conductivity, that  $U_{\rm HF}$  should vary as the square root of the viscosity. Figure 3 shows temperature dependences of the ratios  $U_{\rm HF}\gamma_1^{-0.5}$  and  $U_{\rm HF}\eta^{-0.5}$ . It is seen that, for all of the substances explored, these relations are



Figure 4. Plot of electrical conductivities of the LC samples against the values of  $\varepsilon U_{LF}^2 U_{HF}^{-2}$ found from equation (3) at a temperature 3°C below  $T_{NI}$ .

practically constant. Hence, temperature variations of the threshold voltage  $U_{\rm HF}$  of the EHD instability at  $v > v_{\rm c}$  are dictated by the temperature dependence of the viscosity of a nematic. Making use of the experimental data, we can separate a contribution of the conductivity into a threshold voltage. From equations (1) and (2) it follows that

$$\sigma \sim A'' \varepsilon U_{\rm LF}^2 U_{\rm HF}^{-2}.$$
 (3)

Here, as above, A'' is a numerical constant. For all of the investigated nematics, the values of the right-hand part of equation (3) were calculated for 3°C below the isotropic-nematic phase transition temperature, making use of the values of  $\varepsilon$ ,  $U_{\rm LF}$  and  $U_{\rm HF}$ .

The values found for  $\varepsilon U_{LF}^2 U_{HF}^{-2}$  are presented in figure 4. They are plotted against the conductivities of the same samples, obtained by immediate measurements [7] of  $\sigma$ . It is seen that the experimental values of the squares of the threshold voltages are proportional to  $\sigma$  for the samples measured.

Thus, the experimental data demonstrate the validity of equations (1), (2), resulting from the isotropic model of the EHD instability for nematics with a high positive dielectric anisotropy.

#### 4. Concluding comments

The conditions for initiation of EHD instability have been studied using a number of liquid crystal materials having high, positive dielectric anisotropy. The dependence of the threshold voltage for the EHD instability on viscosity and electrical conductivity of the substances investigated has been determined, and the dependences agree with an isotropic mechanism for the EHD instability.

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