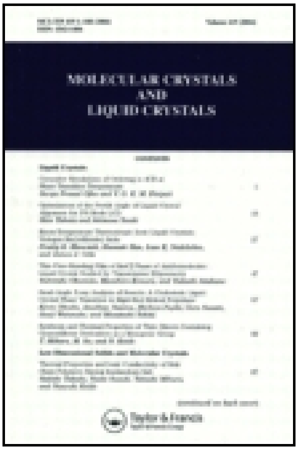

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The Electric Current Caused by a Pulse Flow of Liquid Crystals†

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The oscillating flow of a liquid crystal induces an electric charge across electrodes of a cell which is due to both the electro-kinetic and direct flexo-electric effects. These two contributions can be separated. The flexo-electric coefficient and the anchoring energy of an azoxy-compound mixture are calculated. The technique allows the determination of phase transition points, especially for ferro-electric liquid crystals.

1. INTRODUCTION

According to the original paper of R. Meyer[1] flexo-electric polarization should appear in a mechanically distorted layer of a liquid crystal. As far as we know all the experimental investigations of the flexo-electric distortion[2–5] dealt, in fact, with an inverse flexo-electric effect when an external electric field caused the distortion. The principal difficulties in observing the direct flexo-electric effect arise from both the screening of the relatively small flexo-electric polarization ($P_f = 10^{-13} - 10^{-11}\mathrm{C.cm}^2$) and a number of parasitic effects such as a tribo-electric and an electro-kinetic ones discussed below.

The aim of the present work is to investigate the direct flexo-electric effect and to calculate the corresponding flexo-coefficient and anchoring energy for homeotropically oriented liquid crystal layers.

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2. TECHNIQUE

In our experiment, the pulsed air flow causes the flow of a drop of a liquid crystal accompanied by the distortion of its structure. The principal peculiarity of this technique is an absence of any moving solid parts (as compared, say, with[6]) which drastically diminishes various noises resulting from tribo-electric and capacity effects. Additional alternating air pressure was delivered to an open end of a planar capillary consisting of two glass plates with SnO_2 electrodes on one of them (see insert to Fig. 1). A gap between the plates was fixed with teflon spacers. The surface of the plates was treated with chromium chloride stearate to have homeotropically oriented liquid crystalline layers, or was rubbed with polyvinylketal to obtain the homogeneous orientation.

When air pressure alternating with frequency, say $\omega = 2\pi f$ ($f = 50$ Hz) is applied to a cell, the liquid crystal oscillates around some equilibrium position and induces an alternating electric charge across the electrodes. This charge was amplified and integrated by a charge-coupled amplifier with an input resistance of 10^9Ohm, and then was recorded by a storage oscilloscope. The condition $\omega\tau_c \ll 1$ where τ_c is the space charge (Maxwell) relaxation time should be fulfilled to give the highest possible amplitude of the signal.

The experiment was carried out on the azoxy compound mixture with a nematic range from -5 to $+75$°C (a mixture A[7]), p-octyl-p′-

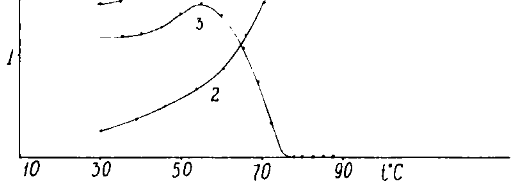

FIGURE 1 Temperature dependences of 1) the flow-induced charge; 2) the extrapolated value for the electro-kinetic contribution; 3) the difference between them (the flexo-electric term) for a homeotropically oriented layer (thickness = 110 μm) of mixture *A*. Insert: experimental set-up; 1) cell, 2) charge-sensitive amplifier, 3) storage oscilloscope; 4) polarizing microscope.

cyanobiphenyl (8CB) and mixtures of both of them. There were also some experiments with ferroelectric liquid crystals.

The curve 1 in Fig. 1 illustrates the typical temperature behaviour of the electric signal generated by a homeotropically oriented layer of mixture A when alternating pressure with amplitude $p = 1.5\text{Pa}$ is applied to a sample.

3. DISCUSSION OF THE FLEXO-ELECTRIC AND THE ELECTRO-KINETIC EFFECTS DUE TO THE FLOW OF A LIQUID CRYSTAL

In order to find a relationship between the flexo-electric charge and parameters of a liquid crystal, we solved equations for unidimensional motion of a homeotropically oriented layer caused by steady-state additional pressure. Let us write down the equation of motion for a mass of liquid:

$$-\frac{\partial p}{\partial x} + \frac{\alpha_2}{d^2} \cdot \frac{\partial^2 v}{\partial \tilde{z}^2} = 0$$

and the motion equation for a director

$$Er^{-1} \cdot \frac{\partial^2 \theta}{\partial \tilde{z}^2} = 2\tilde{z} - 1; \qquad Er = \frac{\alpha_2 \cdot v_0 \cdot d}{K_{33}}$$

with the corresponding boundary conditions

$$W\theta = K_{33}\frac{\partial \theta}{\partial \tilde{z}}, \qquad \tilde{z} = 0$$

$$W\theta = -K_{33}\frac{\partial \theta}{\partial \tilde{z}}, \qquad \tilde{z} = 1$$

Here, Er is the Ericksen number, the Z-axis is directed perpendicularly to the glass surface, $\tilde{z} = z/d$, d is the layer thickness, θ is a director deviation angle with respect to the Z-axis, P is the additional pressure, v_0 is the velocity of a liquid crystal in the central plane of a layer, α_2 is a Leslie coefficient, W is an anchoring energy and K_{33} is an elastic modulus for the bend distortion.

Taking the coupling between the distortion θ and flexo-electric charge q_{fe} into account,

$$q_{\text{fe}} = l\int_0^1 e_{33}\frac{\partial \theta}{\partial \tilde{z}}\, d\tilde{z}$$

we can arrive at the following expression for the flexo-electric charge on meniscus of a liquid crystalline drop

$$q_{\mathrm{fe}} = \frac{2Ere_{33}l}{12 + 6W \cdot d/K_{33}} \tag{1}$$

where l is a length of the meniscus, e_{33} is the corresponding flexo-coefficient.

4. THE RESULTS FOR MIXTURES A

It is easily seen in Fig. 1 (curve 1) that electric charge is generated not only in the nematic state but even in the isotropic phase where there is no flexo-electric polarization. In the latter case, the electro-kinetic effect, that is, the appearance of a so-called flow potential is responsible for the induced charge. The flow potential appears due to the shift of a diffusion part of a double layer, formed at an interface between a liquid and a solid support, by the flow of the liquid. This double layer always exists in any liquid crystal due to its finite ionic conductivity.

The magnitude of the electro-kinetic charge caused by a laminar flow (this is our case as the Reynolds number $\mathrm{Re} = 0.1 \ll 2000$) depends on the parameters of a liquid:[8-9]

$$q_{\mathrm{ek}} = \frac{A \cdot \epsilon\zeta}{\eta} \tag{2}$$

Here, ζ is a so-called ζ-potential, ϵ and η are the dielectric permittivity and viscosity of the liquid (taken for mixture A from [7]), A is a constant independent of temperature.

Thus, in the isotropic phase we measure only the electro-kinetic charge and both contributions (the electro-kinetic and flexo-electric) are measured in the nematic and other liquid crystalline phases.

Under conditions of weak temperature dependence of the ζ-potential, equation (2) allows the analytic extrapolation of the isotropic part of curve 1 in Fig. 1 into the nematic phase. Then the difference between the experimental curve (curve 1) and the extrapolated one (curve 2) corresponds to the flexo-electric charge, which is sought for (curve 3). Using equation (1) for two different cell thicknesses one can calculate parameters e_{33} and W. For mixture A we have $e_{33} = 10^{-5}\mathrm{dyn}^{1/2}$ and $W = 5 \cdot 10^{-5}\mathrm{dyn.cm}^{-1}$. These data agree with those determined from the investigations of the inverse flexo-electric effect. Our accuracy does not allow the temperature behav-

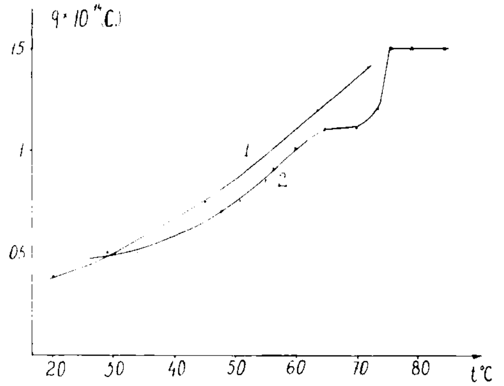

FIGURE 2 Temperature behaviour of the electro-kinetic charge (calculated curve 1) and flow induced charge (experimental curve 2) for a homogeneously oriented layer of mixture *A* (thickness = 100 μm).

iour of these parameters to be studied though, in principle, a possibility is not excluded.

In Fig. 2, the temperature behaviour of the induced charge is shown for a homogeneously oriented layer of mixture *A*, the director being perpendicular to the flow direction. For such geometry the flexo-electric polarization should not arise as there is no corresponding distortion. Indeed, the experimental curve for charge generated in the nematic phase coincides with the curve for electro-kinetic effect.

5. ELECTRIC CHARGE GENERATION IN 8CB AND ITS MIXTURES WITH AZOXY-COMPOUNDS

In the isotropic phase the electric response for a step of pressure has different signs for azoxy-compound mixture A and 8CB. It means

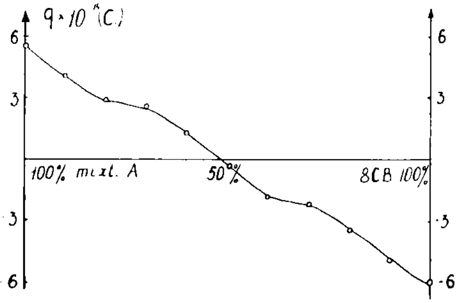

FIGURE 3 Dependence of the flow-induced charge on composition of mixtures 8CB with azoxy-compounds (mixture *A*).

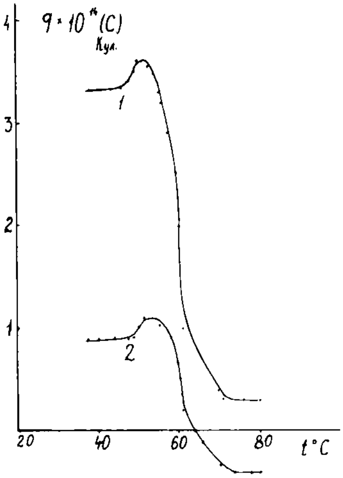

FIGURE 4 Flow induced charge as a function of temperature for a compensated mixture (1 : 1) of *A* with 8CB. Cell thicknesses $d = 110$ μm (1) and 60 μm(2).

that the diffusion parts of their double layers are charged in opposite ways, positively for 8CB and negatively for mixture *A*. Since these substances may be mixed in any ratio there should exist a composition with zero charge of the diffusion part. Such an "isoelectric" point corresponds to a mixture 1 : 1 of the azocompounds and 8CB, Fig. 3. For this mixture the temperature behaviour of the flow-induced charge was investigated at two different cell thicknesses, $d = 60$ and 110 μm (Fig. 4). The experimental curves are similar to curve 3 in Fig. 1 which was obtained by a mathematical subtraction. So, by a choice of the proper mixture composition it is possible to reduce essentially the electro-kinetic contribution to the induced charge and to study net flexo-electric phenomena.

The same "isoelectric" state for an individual liquid crystalline compound in a cell with SnO_2 electrodes may be obtained using small quantities of surface-active substances as impurities. For instance, the isoelectric point is achieved at a concentration as small as 10^{-9} (by weight) of biologically active valinomycine in 8CB.

The charge generation effect may effectively be used for the determination of phase transition temperatures in liquid crystals. In Fig. 5, the phase transitions "isotropic liquid-nematic" and "nematic-smectic *A*" are shown by arrows. As compared with the DTA method this technique is especially useful for the detection of thermodynamically weak but "electrically strong" transitions, for example between smectic *A* and chiral (ferroelectric) smectic *C* phases, Fig. 6. The

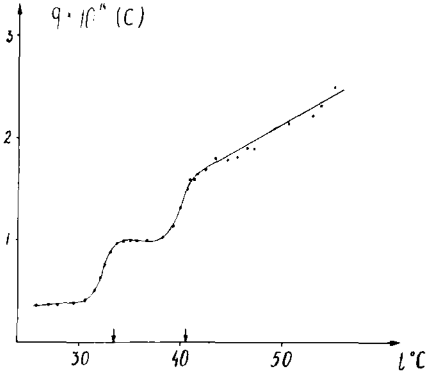

FIGURE 5 Charge-generation effect near phase transition points (T_{NA} = 33.5°C, T_{NI} = 40.5°C) in 8CB.

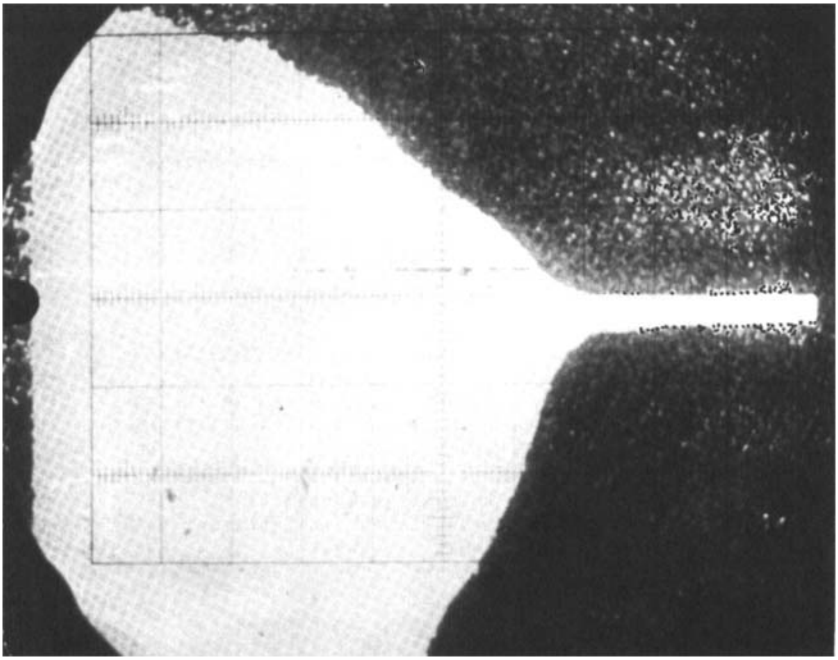

FIGURE 6 The oscilloscope trace for the charge generation effect during cooling a sample of a ferroelectric mixture in the vicinity of the transition from the smectic *A* to the chiral smectic *C* phase.

charge generation effect in the ferroelectric phase is two orders of magnitude more pronounced than in the isotropic or the nematic phases. The increase is due to the flow and pressure induced changes in the spontaneous polarization.

The Maxwell relaxation time and the thickness of the diffusion part of a double layer are the crucial factors for the appearance of the charge generation effect. Maximum signals are observed for the electrical conductivity of a liquid in the range of $\lambda \approx 10^{-14} - 10^{-10} \mathrm{Ohm}^{-1}\mathrm{cm}^{-1}$. However, we managed to detect signals even for water solutions with $\lambda \approx 10^{-3} \mathrm{Ohm}^{-1}\mathrm{cm}^{-1}$. Thus, our technique opens the possibilities to investigate such electrical effects in lyotropic liquid crystals and biologically active isotropic solutions.

In conclusion, we described a novel technique which allowed the detection of the electro-kinetic and direct flexo-electric (for the first time) effects in various liquid crystalline phases. The existence of the isoelectric points is shown for the electro-kinetic effect and a new sensitive method is proposed for the detection of any electrical changes at phase transition points.

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