

# Observation of the direct flexoelectric effect in nematic liquid crystals

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It is found that an electric current associated with flexoelectric and electrokinetic phenomena is generated in a pulsating flow of a liquid crystal in the nematic phase. The flexomodulus and the energy of binding of the liquid crystal with the substrate are calculated from the flexoelectric contribution.

1. In 1969, Meyer predicted the existence of flexoelectric polarization in a nonuniformly deformed layer of a liquid crystal (LC).<sup>1</sup> In contrast to the piezoelectric effect, which is associated with the relative displacement of the centers of mass of atoms or molecules at the lattice sites, the flexoelectric effect depends on the change in the mutual orientation of the molecules (LC). The experimental study of flexoelectric phenomena in nematic liquid crystals (NLC) began with Schmidt, Schadt, and Helfrich in 1972.<sup>2</sup> All later studies of flexoelectric phenomena, including those of Ref. 2, included the study of the deformation of the LC in an electric field, i.e., the inverse flexoelectric effect. All attempts to detect the direct flexoelectric effect in NLC were unsuccessful. The main difficulties in studying the direct flexoelectric effect are the screening of flexopolarization by impurity charges and coherent electrokinetic phenomena at the electrode-LC interface. We will discuss both phenomena in greater detail below.

2. We have obtained for the first time experimentally the temperature dependence of the induced polarization caused by the orientational deformation of the NLC.

We chose as the object of the study a mixture of two azoxy compounds (mixture *A*).<sup>3</sup> In the experiment the nonuniform deformation of the director of the NLC appeared during the flow of the LC under the action of a pulsating air flow. This was the key factor in the experimental arrangement shown in Fig. 1b. Under the combined action of the variable air pressure and capillary forces, the homeotropic layer of NLC pulsed around an equilibrium position. At the same time, a variable electric charge, which first entered a charge-sensitive amplifier ( $R_{in} = 1 \text{ G}\Omega$ ) and then an oscilloscope with storage, was induced.

Curve 1 in Fig. 1a shows the temperature dependence of the surface charge density generated by the LC with an excess-pressure amplitude  $P = 1.5 \text{ Pa}$ . The rise time of the excess pressure satisfies the inequality  $\tau < \tau_c$ , where  $\tau_c$  is the Maxwellian relaxation time (for the sample used in our experiment  $\tau_c = 0.1 \text{ s}$ ).

3. The liquid crystal generates a charge in the isotropic phase. This charge is associated with the so-called flow-induced current,<sup>4</sup> which is usually produced because the flow of the liquid removes the charge in the diffusion part of the double-layer formed at the liquid-phase-electrode interface. In the case of a laminar fluid flow (in our experiment the Reynolds number is  $Re = 0.1$ ) the electrokinetic polarization is

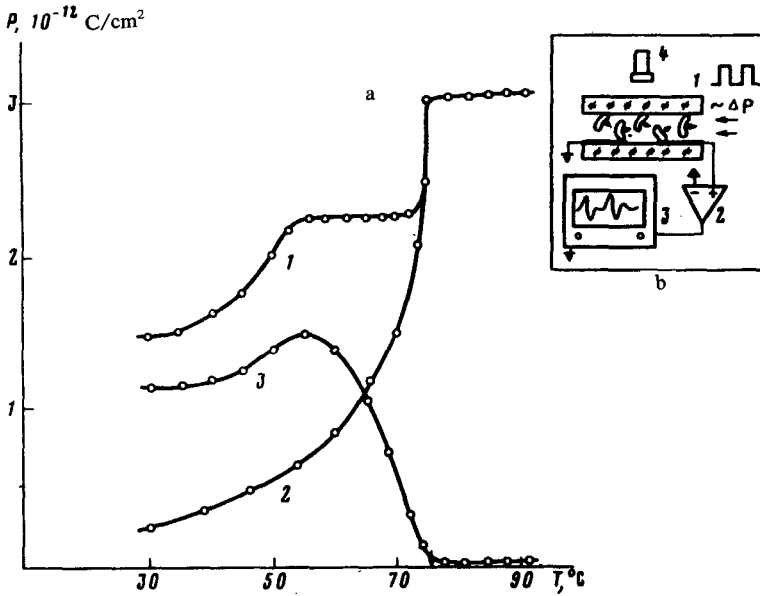


FIG. 1. (a) Homeotropic orientation of mixture *A* (the arrow marks the nematic–isotropic-phase transition) 1) Experimental curve of the surface charge density as a function of temperature, 2) electrokinetic polarization as a function of temperature, 3) flexoelectric polarization as a function of temperature; (b) the experimental arrangement: (1) liquid-crystal cell with planar electrodes, (2) charge-sensitive amplifier, (3) S8-1 oscilloscope, (4) Polam-311 polarization microscope.

related to the temperature-dependent constitutive parameters of the medium by the relation<sup>5</sup>

$$P_{\text{e.k.}} = A \epsilon \xi / \eta, \quad (1)$$

where *A* is a temperature-independent constant,  $\xi$  is the  $\xi$ -potential,  $\epsilon$  is the dielectric constant, and  $\eta$  is the viscosity of the liquid phase. Using the data in Ref. 3 on the temperature dependence of  $\epsilon_{\parallel}$  and  $\eta_1$  for the mixture *A* and taking advantage of the fact that the  $\xi$ -potential is nearly independent of temperature<sup>6</sup> [using Eq. (1) as a guide], we can analytically continue the isotropic branch of the electrokinetic effect into the temperature region of the nematic phase. We thus obtain curve 2 in Fig. 1a, which shows the temperature dependence of the electrokinetic polarization. The temperature dependence of the difference between the polarization measured in the experiment and the electrokinetic polarization is the sought flexopolarization (curve 3 in Fig. 1a).

4. To find the relationship between the flexoelectric charge and the parameters of the LC we solved the problem of the motion of a homeotropic layer of NLC under the action of a constant pressure drop. For a Poiseuille velocity profile in the LC, we obtained the following expression for the flexoelectric charge:

$$Q_{\text{f.e.}} = \frac{2 \text{Er} e_{33} l}{12 + 6 Wd / K_{33}}, \quad (2)$$

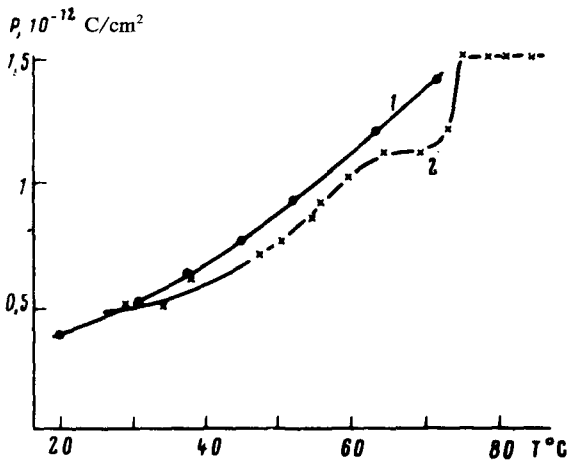


FIG. 2. Planar orientation of mixture *A* (the director of the LC is perpendicular to the direction of displacement) 1) Temperature dependence of the electrokinetic polarization, 2) experimental curve for the surface charge density as a function of temperature.

where  $E_r$  is Ericksen's number,  $e_{33}$  is the flexomodulus,  $l$  is the linear length of the meniscus of the LC,  $d$  is the thickness of the layer,  $K_{33}$  is the elastic constant, and  $W$  is the energy of binding of the liquid crystal with the substrate. Since the value of flexoelectric charge for two different thicknesses of the LC is known, we can calculate the value of the flexomodulus,  $e_{33} = 10^{-5}$  dynes<sup>1/2</sup>, and the energy of binding the LC with the substrate,  $W = 5 \times 10^{-5}$  dynes/cm. These values are in good order-of-magnitude agreement with the data obtained from studies of the inverse flexoelectric effect in related compounds.<sup>7</sup>

5. Figure 2 shows that for a flat layer of mixture *A* in a geometry such that the

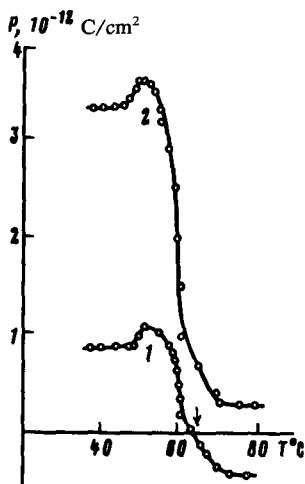


FIG. 3. Experimental temperature dependences of the surface charge density for a mixture with the compositions *A* and 8CB 1)  $d_1 = 60 \mu\text{m}$ , 2)  $d_2 = 110 \mu\text{m}$  ( $d_1$  and  $d_2$  are the thicknesses of the LC layers). The arrow marks the nematic-isotropic-phase phase transition.

director is perpendicular to the velocity of the LC the experimental curve for the temperature dependence of the surface charge density closely matches the curve for the electrokinetic effect. In this geometry there is no flexopolarization, since the liquid crystal is not strained nonuniformly.

6. In the isotropic phase, the electrical responses to a pressure increment for the mixtures *A* and 8CB (4-cyano-4'-octyldiphenyl) have opposite polarities; i.e., the diffusion parts of the double layers in these LC are oppositely charged. By mixing these crystals it is possible to achieve an isoelectric state, i.e., a state with zero charge in the diffusion part of the double layer, and thereby to decrease substantially the electrokinetic effect. The experimental curves for the surface charge density are plotted in Fig. 3 as a function of the temperature for an isoelectric mixture of the compositions *A* and 8CB. These curves reproduce the temperature profile of the flexopolarization (curve 3 in Fig. 1).

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