

## Dielectric quenching of the electric polar surface instability in a nematic liquid crystal

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The existence of polar electro-optic effect induced by a surface polarization of a nematic liquid crystal is proved using a simultaneous action of vertical dc and high-frequency ac electric fields on the homeotropically aligned nematic liquid crystal with positive dielectric anisotropy. The proposed method enables us to measure the surface electric polarization and the anchoring energy in the course of the same experiment. The evaluated surface polarization for the pentylcyanobiphenyl-silicone elastomere interface is 1 order of magnitude larger than the nematic flexoelectric constant.

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An external electric field  $\mathbf{E}$  applied to a uniformly oriented nematic liquid crystal can distort the mean direction of molecules (the director  $\mathbf{n}$ ) and thus alter the optical properties of the nematic liquid crystal. There are at least four different mechanisms of these electro-optic effects, namely, the dielectric, electrohydrodynamic, flexoelectric, and surface-polarization ones [1]. In the much studied Fréedericksz transition the distortions arise in the bulk of the sample. This effect is quadratic in field and thus nonsensitive to the field polarity. Alternatively, in the linear case of the flexoelectric or surface-polarization mechanism the distortions may be polar, and the destabilizing action of the electric field can be localized at the surface of the cell boundaries [2]. These polar surface instabilities were recently observed in homeotropic layers of the nematic liquid crystal with positive dielectric and electroconductivity anisotropies under the action of vertical dc or low-frequency ac electric field [3–5]. They possess the field threshold and are manifested by the appearance of rounded domains due to static deformations of the director near the cell anode [3–5] or cathode [4,5]. The polarity of the deformations depends on the nature of an orienting coating. This enables one to distinguish the flexoelectric and surface polarization origin of the effect [4,5]. Using the independent measurements of the surface anchoring at the cell boundary [6] (which tends to stabilize the initial homeotropic orientation) and the field threshold of instability, the surface polarization was estimated and shown to be an order of magnitude larger than the flexoelectric polarization [4,5].

However, with field increase, the static distortions begin to be accompanied by the electrohydrodynamic distortions with domains possessing similar optical textures but which have a completely different origin, namely, the Felici mechanism [7], brought about by charge injection (see [3–5] and also [8,9]). This additional voltage instability makes the observation of the field-induced static domains very delicate and demands significant effort for separation

[5].

In this paper, we report an idea and experimental realization of testing of the origin and instability parameters in the homeotropic nematic layer under the action of the vertical electric field. This test, first, proves the existence of the electric polar surface instability, which arises due to the surface polarization and, second, allows us to measure the values of the anchoring energy and surface destabilizing torque for the same cell in the course of the same experiment.

The idea is based on the dielectric quenching of the surface instability through the simultaneous action of the vertical dc and high-frequency ac electric fields on the homeotropically oriented nematic liquid crystal characterized by positive dielectric anisotropy  $\Delta\epsilon > 0$ . One can expect that the presence of an additional high-frequency ac field would not change the voltage threshold of the charge injection and thus the threshold of the electrohydrodynamic instability. On the other hand, if the origin of the direction distortions is the surface polarization or flexoelectricity, the corresponding field threshold should increase with the ac field. Let us consider this question in more detail.

Due to the usual dielectric mechanism the additional ac field should stabilize the initial vertical orientation of the director in the geometry mentioned above. Anisotropic dielectric response of the nematic bulk results in a torque exerted by the bulk on the surface. Following Ref. [3], the latter can be estimated for small angles  $\theta_s$  of the surface tilt of the director  $\mathbf{n}$  compared to vertical axis as  $K\theta_s/\xi_{ac}$ , where

$$\xi_{ac} = \sqrt{4\pi K/\Delta\epsilon}/|E_{ac}|$$

is the coherence length associated with stabilizing ac electric field and  $K$  is the nematic curvature modulus in a one-constant approximation. It will act with other stabilizing factors, namely, the analogical dielectric effect of the dc field with coherence length

$$\xi_{dc} = \sqrt{4\pi K/\Delta\epsilon}/|E_{dc}|,$$

and the surface anchoring torque  $K\theta_s/L$ , preventing the surface tilt. Here  $L = K/W$  is the de Gennes–Kléman extrapolation length characterizing the strength  $W$  of the surface anchoring. The  $L$  value is expected to be of a few micrometers [6]; thus  $L \leq \xi_{ac}, \xi_{dc}$ . In principle, one must also take into account the bulk curvature elasticity. However, in the large-field regime (which is of interest here)  $\xi_{ac}, \xi_{dc}$  are smaller than the cell thickness  $d$  and the corresponding stabilizing term can be neglected [3]. On the other hand,  $\xi_{ac}, \xi_{dc}$  are assumed to be larger than the coherence length of the nematic-isotropic transition; hence the quenching effect remains a bulk effect independent of possible changes of the order parameter close to the surface.

In spite of the similarity in the stabilizing dielectric effect of the ac and dc fields their destabilizing role is different. If the frequency of the additional ac field is sufficiently high (more than a few tens of Hz [3–5]), no ac terms will contribute to linear coupling energy  $-\mathbf{E}(\mathbf{P}_s + \mathbf{P}_f)$  of the flexoelectric  $\mathbf{P}_f$  and surface  $\mathbf{P}_s$  polarizations with the electric field. Thus this coupling will be determined only by the dc field and result in the destabilizing torque  $(-e^*E_{dc})\theta_s$ , where  $e^*$  represents the total flexoelectric and surface polarizations,  $e^* = e_1 + e_3 + |\mathbf{P}_s|$ ;  $e_1$  and  $e_3$  are the flexocoefficients in usual notation and the  $|\mathbf{P}_s|$  term is taken with a plus sign which corresponds to the so-called anode instability [5]. The surface torque equation with the assumption  $\xi_{ac}, \xi_{dc} < d$  reads as

$$\left( \frac{K}{\xi_{ac}} + \frac{K}{\xi_{dc}} + \frac{K}{L} - e^*E_{dc} \right) \theta_s = 0, \quad (1)$$

and results in the threshold of the surface dc-field-induced polar instability as the increasing function of the applied additional ac field,

$$E_{dc,th} = \frac{K}{e^* - (\Delta\epsilon K/4\pi)^{1/2}} \left( \frac{|E_{ac}|}{(4\pi K/\Delta\epsilon)^{1/2}} + \frac{1}{L} \right), \quad (2)$$

as it was expected. Hence, the behavior  $E_{dc,th}$  vs  $E_{ac}$  allows us to distinguish the static (i.e., due to surface and flexoelectric polarization) and dynamic (due to charge injection) mechanisms of the instabilities. The second important consequence of Eq. (2) is the possibility to evaluate  $L$  by comparison with the values of  $E_{dc,th}$  for some  $|E_{ac}| \neq 0$  and  $|E_{ac}| = 0$ ,

$$E_{dc,th}(ac \neq 0)/E_{dc,th}(ac = 0) = 1 + L/\xi_{ac}, \quad (3)$$

and then to extract from it the  $e^*$  value.

To perform the experiment, we have prepared homeotropic samples of pentycyanobiphenyl (5CB) with  $\Delta\epsilon > 0$  in transparent cells. The homeotropic orientation was established in two ways: by using clean untreated  $\text{In}_2\text{O}_3:\text{Sn}$  electrodes and by using the same electrodes coated with silicone elastomer of thickness amounting to several hundreds of nanometers [5]. The nematic layer thickness  $d$  was  $60 \mu\text{m}$ . A dc field was applied to the electrodes by a calibrated voltage source model No. P4108. The textural observations were carried out at room temperature ( $20^\circ\text{C}$ ) using a polarizing microscope.

The cell textures are similar to that described in Refs. [3–5] and depend on the applied dc field. One could identify three ranges of the voltage  $U_{dc}$ : (1) in the range  $0 \leq U_{dc} \leq U_{dc,1}$ , the initial homeotropic orientation was retained; (2) the static domain localized at cell anode were observed in the range  $U_{dc,1} \leq U_{dc} \leq U_{dc,2}$ , Fig. 1(a); (3) domains created by nematic vortex motion were observed when the voltage became  $U_{dc} \geq U_{dc,2}$ .

The applying of additional ac field results in the quenching of the static domains [Figs. 1(b) and 1(c)] and linear increase of their threshold  $U_{dc,1}$ , Fig. 2. It unambiguously confirms the conclusion of Refs. [3–5] about the flexoelectric and/or surface polarization origin of the instabilities in the range  $U_{dc,1} \leq U_{dc} \leq U_{dc,2}$ .

The slope of functions  $U_{dc,1}(U_{ac})$  in accordance with Eq. (2) depends on the nature of the surface coating and enables us to measure the  $e^*$  value as well as the surface anchoring length  $L$  and thus the coefficient  $W$  of the an-

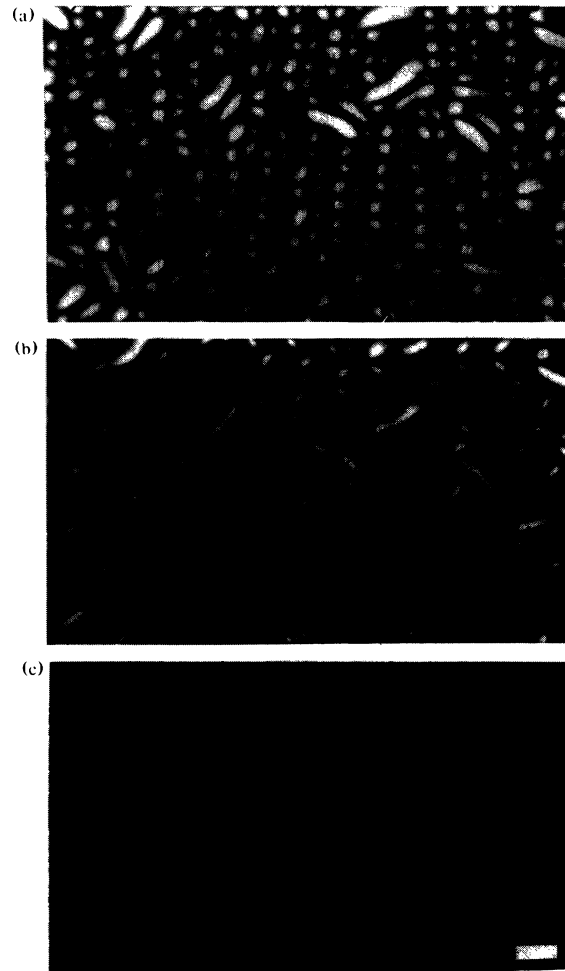


FIG. 1. Textures of a surface polar instability induced by the dc electric field ( $U_{dc} = 3 \text{ V}$ ) in a homeotropically oriented layer of 5CB which is subjected to additional action of the ac electric field: (a)  $U_{ac} = 0$ , (b) 2 V, and (c) 6 V. The increase of ac field results in the quenching of the domain structure. The cell electrodes are covered with silicone elastomere. Polarizers are crossed. Bar length is  $100 \mu\text{m}$ .

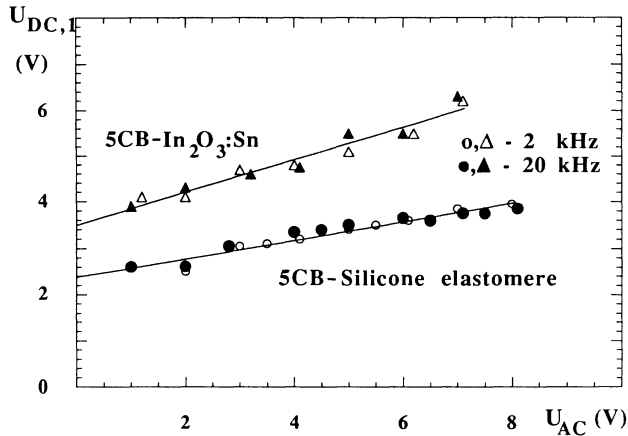


FIG. 2. The dc voltage threshold  $U_{dc,1}$  of the surface polar instability as a function of the applied ac field. The data correspond to two cells with different surface treatments and to two different ac field frequencies.

choring,

$$L = d \frac{(4\pi K/\Delta\epsilon)^{1/2}}{U_{ac}} \frac{\Delta U_{dc}}{U_{dc}^0} \quad (4)$$

and

$$e^* = \sqrt{\Delta\epsilon K/4\pi} \left( 1 + \frac{U_{ac}}{\Delta U_{dc}} \right), \quad (5)$$

where  $U_{dc}^0$  is the dc voltage threshold of the static domains for zero ac field,  $\Delta U_{dc} = U_{dc} - U_{dc}^0$ ,  $U_{dc}$  is the dc threshold corresponding to some nonzero ac voltage  $U_{ac}$ .

Taking the data of Fig. 2 and a typical value for 5CB at room temperature,  $\Delta\epsilon = 14$  and  $K = 0.8 \times 10^{-6}$  dyn [10], one finds from Eq. (4) for 5CB-In<sub>2</sub>O<sub>3</sub>:Sn interface  $L = 1.7$   $\mu\text{m}$  and  $W = 4.7 \times 10^{-3}$  erg/cm<sup>2</sup>, while for 5CB-silicone elastomere interface  $L = 1.3$   $\mu\text{m}$  and  $W = 6 \times 10^{-3}$  erg/cm<sup>2</sup>. The last value is close to one previously measured for the same interface by two independent testing techniques based on the light scattering and determination of parameters of the topological defects,  $(2 \pm 1) \times 10^{-3}$  erg/cm<sup>2</sup>; see Ref. [6]. Analogically, Eq. (5) gives the values of  $e^*$ :  $e^* = 4 \times 10^{-3}$  cgs for the 5CB-In<sub>2</sub>O<sub>3</sub>:Sn interface and  $e^* = 6 \times 10^{-3}$  cgs for the 5CB-silicone elas-

tomere interface. Thus, in both cases the total surface and flexoelectric polarization  $e^*$  is 1 order of magnitude larger than the known values of the corresponding flexocoefficients for cyanobiphenyls,  $e_1 + e_3 = 0.7 \times 10^{-3}$  cgs (Ref. [11]) and  $e_1 + e_3 = 0.15 \times 10^{-3}$  cgs (Ref. [12]). It means that the polar surface instability observed in the present and previous [3–5] experiments arises due to the surface polarization rather than to the flexoelectric ones, which confirms the conclusions of Refs. [4,5]. We should note the good agreement of the values estimated in the present paper with those earlier reported for  $|\mathbf{P}_s|$  at the 5CB-silicone elastomere interface:  $6 \times 10^{-3}$  cgs (Ref. [4]) and  $9 \times 10^{-3}$  cgs (Ref. [5]). Moreover, one can deduce the same magnitude of  $|\mathbf{P}_s|$  from a simple molecular model [4], taking into account the possibility of the ferroelectric ordering at the nematic surface due to the polar nature of the interaction between the nematic molecules and the ambient medium. In fact, this polar ordering has been observed for the cyanobiphenyls [13]. In principle, other sources of the surface polarization are also possible, for example, so-called order electric polarization and selective-ion adsorption; see, e.g., the discussion in Ref. [14]. Thus the forthcoming problem is to evaluate the effect of the different contributions to the total surface polarization. The corresponding experiments are in progress.

Our conclusion is as follows. Applying the vertical ac electric field to the homeotropic layer of 5CB, we have observed the dielectric quenching of the dc-electric-field-induced polar surface instability. It manifests in a linear increase of the instability threshold with the ac field. These data enable us to confirm the previous conclusion [4,5] that the origin of instability is the surface polarization of the nematic liquid crystal. The proposed testing method made possible the simultaneous measurements of the surface anchoring energy and the total surface electric polarization for the same cell in the course of the same experiment. The linear character of quenching shows that, in addition, the quadratic stabilizing bulk dielectric effect results in a linear surface torque.

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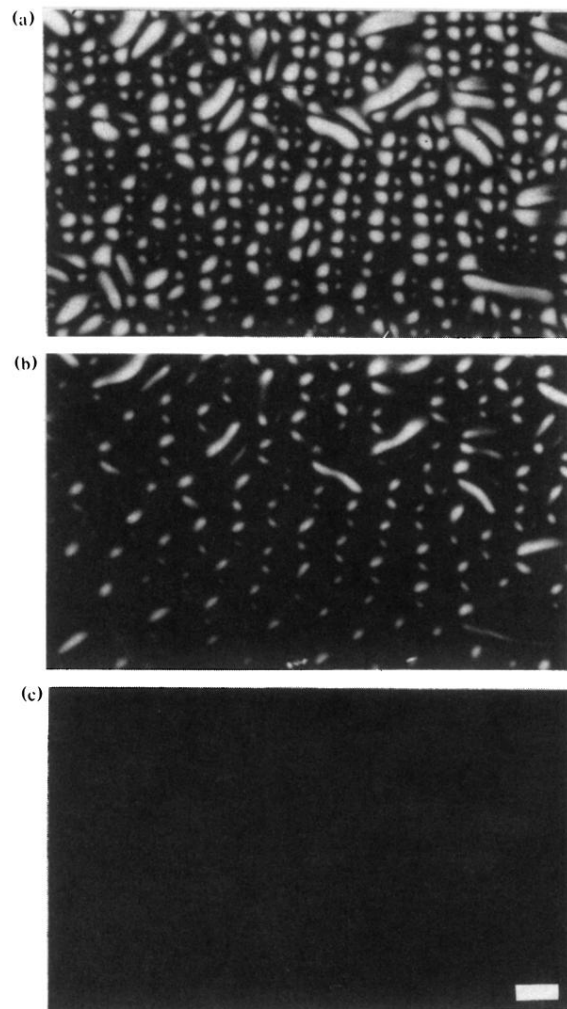


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