

Unipolar electro-optic effect in a nematic cell

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We report on a unipolar electro-optic effect in a nematic liquid crystal cell. The cell placed between two polarizers behaves as an "optical diode:" only one polarity of the applied voltage makes the cell transparent. The phenomenon is caused by a double symmetry breaking at the two plates that bound the nematic slab. [S1063-651X(98)50801-X]

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Uniform nematic materials are centrosymmetric even when they are formed by rodlike molecules with large longitudinal dipole moments. Molecular flip-flops and head-to-head overlapping establish centrosymmetric arrangement in which the bulk electric polarization averages to zero. However, real samples are always bounded. This spatial bounding breaks the central symmetry in different ways [1–10]. We present experimental evidence of a unipolar electro-optic effect as a particular consequence of the symmetry breaking.

We studied a nematic material pentylcyanobiphenyl (5CB) with positive dielectric anisotropy, $\epsilon_a = \epsilon_{\parallel} - \epsilon_{\perp} = 14$; the subscripts refer to the director \mathbf{n} . A 5CB molecule has a dipole moment $\mu \approx 4$ debye directed parallel to the long axis.

The cell is comprised of two glass plates separated by Mylar spacers (thickness 10–60 μm). Each plate as a transparent conducting layer ($\text{In}_2\text{O}_3:\text{Sn}$ or In_2O_3) and an alignment layer to orient the liquid crystal. The alignment layers must satisfy two requirements to make the unipolar optical response possible. First, both materials should orient \mathbf{n} perpendicularly to the plates (so-called homeotropic orientation). Second, the material at one plate should be different from the material at the opposite plate [Fig. 1(a)].

We used both organic and inorganic alignment layers: (a) silicon elastomer $(\text{CH}_3)_3\text{SiO}[(\text{CH}_3)_2\text{SiO}]_n\text{Si}(\text{CH}_3)_3$, $n \sim 25\,000$ (SE), coated over $\text{In}_2\text{O}_3:\text{Sn}$; (b) octadecyltrichlorosilane $[\text{C}_{18}\text{H}_{37}\text{SiCl}_3]$ (OTS) over $\text{In}_2\text{O}_3:\text{Sn}$; (c) In_2O_3 ; (d) $\text{In}_2\text{O}_3\text{-SiO}_2\text{-NiO}$; (e) lecithin *L*- α -phosphatidylcholine over $\text{In}_2\text{O}_3:\text{Sn}$; (f) *n*, *n*-dimethyl-*n*-octadecyl-3-aminopropyltrimethoxysilylchloride $\text{CH}_3(\text{CH}_2)_{17}(\text{CH}_3)_2\text{N}^+(\text{CH}_2)_3\text{Si}(\text{OCH}_3)_3\text{Cl}^-$ (DMOAP) over $\text{In}_2\text{O}_3:\text{Sn}$.

The sample placed between a pair of crossed polarizers was probed with normally incident modulated light of a He-Ne laser. When no field is applied, the system blocks the laser beam [Figs. 1(a), and 2–4]. Light incident, say, from below is linearly polarized by the bottom polarizer; since \mathbf{n} is parallel to the direction of propagation [Fig. 1(a)], this polarization does not change and light cannot get through the top polarizer. The situation changes when a dc voltage is applied across the cell. The dc voltage was incremented by steps in 20 mV each 5 s. Starting with some low-voltage threshold

$|U_{\text{th}}| \sim 1$ V the cell becomes transparent: The intensity of transmitted light increases with the voltage at $|U| \geq |U_{\text{th}}|$ [Figs. 1(b) and 2–4]. For the reversed polarity the clear state either does not appear at all or it appears at different threshold [Figs. 1(c) and 2–4].

Polarizing microscope inspection reveals that in the transparent state, \mathbf{n} deviates from the vertical orientation. To test the spatial location of these distortions we measured the intensity of light reflected from the nematic boundaries. Distortions at the surface change the effective refractive index and thus alter the reflection. To separate beams reflected from different interfaces, special wedge cells have been constructed.

The results show that the low-voltage threshold instabilities are caused by director distortions located either at the anode or cathode of the cell. In Fig. 3, for negative voltages, both transmitted and reflected intensities start to change at about (-2.6) V; the reflective surface is the anode. When the polarity is reversed (so that the reflective surface becomes the cathode), the reflected light intensity does not change even when $U > 4.5$ V and the transmitted intensity is increased due to the director distortions elsewhere in the cell.

No optical diode effect was observed in symmetric cells (both plates are treated identically): the reverse of polarity does not change $|U_{\text{th}}|$.

Polarity of the optical diode effect depends on both the alignment and nematic materials. For example, doping of 5CB with a small amount (0.15% by weight) of a chiral material CE1 $[\text{CH}_3\text{CH}_2\text{CH}(\text{CH}_3)\text{CH}_2(\text{C}_6\text{H}_4)_2\text{COO}(\text{C}_6\text{H}_4)\text{CN}]$ reverses the polarity of the cell (Fig. 4). Nematics with longitudinal dipoles and $\epsilon_a > 0$ other than 5CB show qualitatively the same effects as 5CB. In contrast, nematics such as diheptylazobenzene with $\epsilon_a > 0$ composed of centrosymmetric molecules with zero dipole moments do not respond to the field of either polarity.

The asymmetry of electrooptical effects has been previously reported by Petrov [3] and by Lee and Patel [7] for cells with in-plane (planar) director orientation. The difference from the optical diode effect in the homeotropic cell is twofold. First, in the absence of the field the planar cell is normally transparent. Second, if $\epsilon_a > 0$, the polarity of the planar cell is masked by a dielectric response and reorientation of \mathbf{n} along the field.

The parameters of the effect observed are close to the previously discussed instability in homeotropic cells with

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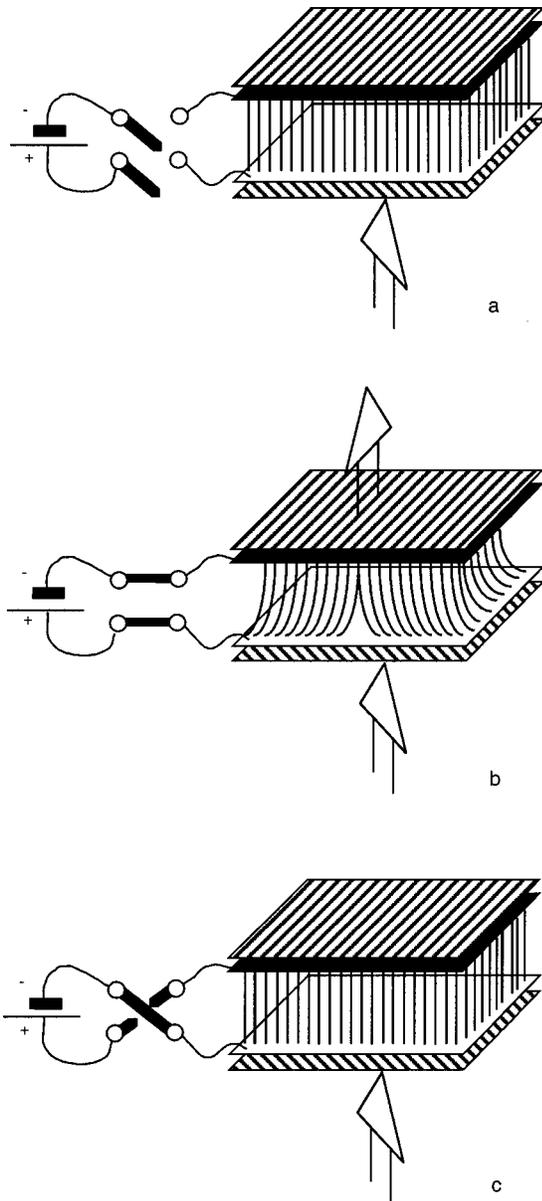


FIG. 1. A scheme of a nematic “optical diode.” The cell is bounded by two different plates (black and white) and placed between two crossed polarizers. The director is oriented normally to the plates. In the absence of the field, the system blocks light coming through (a). A vertical electric field of a particular polarity causes director distortions and allows the light to pass through the second polarizer (b), while the opposite field polarity does not (c).

identical boundary conditions [10,11], excluding the fact that the voltage of different polarity causes a different response or different threshold of the response. Below we briefly discuss the plausible mechanism of the effect.

A normal dielectric mechanism of coupling between a nematic with $\epsilon_a > 0$ and the field \mathbf{E} acting along \mathbf{n} should only stabilize the homeotropic orientation. The destabilizing mechanism can come from other effects, such as electrohydrodynamics, flexoelectricity, and surface polarization. Electrohydrodynamics can be ruled out since the director distortions are static and occur at low voltages. In addition, the observed distortions can be quenched by an ac field applied

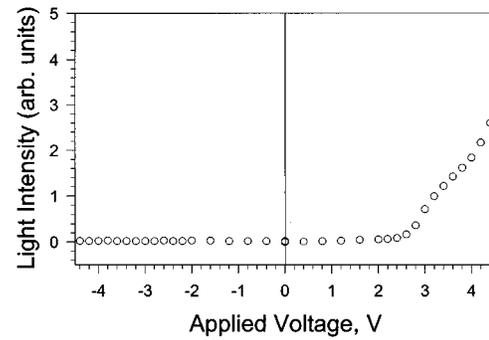


FIG. 2. Intensity of light transmitted through a nematic cell vs applied dc voltage. Nematic slab of thickness $23 \mu\text{m}$ is confined between OTS and DMOAP layers. The voltage is positive when the DMOAP-coated plate is the cathode.

simultaneously with the dc field [11]. The plausible mechanisms of the distortions are flexoelectricity and surface polarization [1–4].

Flexoelectric polarization comes from director distortions (an analogous piezoelectric effect is well known for solids). Surface polarization \mathbf{P}_s can be caused, for example, by ferroelectric ordering of nematic molecules with longitudinal dipoles [1–6]; nonzero \mathbf{P}_s has been directly observed in experiments on second-harmonic generation [5]. There is experimental evidence [10,11] that the surface polarization can cause director distortions in the applied field when \mathbf{P}_s and \mathbf{E} are antiparallel.

Both surface polarization and flexoelectric effects are linear in \mathbf{E} and result in surface torques $-2P_s E \theta$ and $-e E \theta$, respectively [4]. Here θ is the director deviation from the normal to the plate, e is the flexoelectric coefficient. The threshold field E_{th} of instability can be roughly determined as the point where the torques destabilizing normal orientation become larger than the stabilizing surface anchoring torque $W\theta$ and the stabilizing dielectric torque $\sqrt{\epsilon_0 \epsilon_a K} |E| \theta$; for more detailed theoretical models, see Refs. [3,6]. Here W is the so-called anchoring coefficient and K is the bend elastic constant. Surface anchoring is caused by anisotropic molecular interactions that keep \mathbf{n} oriented normally to the plate. The balance of torques ($\sqrt{\epsilon_0 \epsilon_a K} |E| + W - eE + 2P_s E$) $\theta = 0$ gives $E_{\text{th}} = W / (e - 2P_s - \sqrt{\epsilon_0 \epsilon_a K})$ (for the case when $E_{\text{th}} > 0$). Here and below we take $E_{\text{th}} > 0$ and $P_s > 0$ when the corresponding vectors are directed from the lower plate to

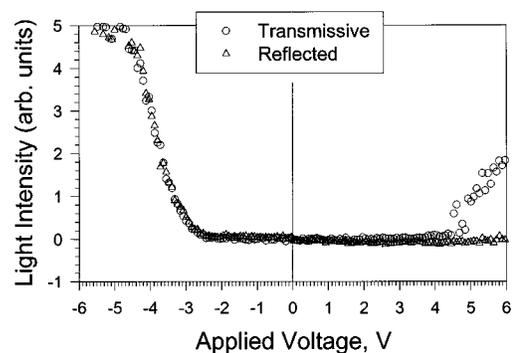


FIG. 3. Transmitted and reflected light intensities vs applied dc voltage. The nematic slab is confined between In_2O_3 and $\text{In}_2\text{O}_3\text{-SiO}_2\text{-NiO}$ layers.

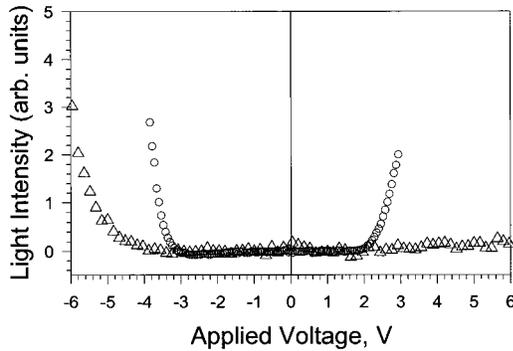


FIG. 4. Intensity of transmitted light vs applied dc voltage: pure 5 CB (circles) and 5CB doped with 0.15% (by weight) of CE1 (triangles). Nematic slab of thickness $36 \mu\text{m}$ is confined between DMOAP and SE plates. Note the polarity reverse of the effect.

the upper plate in Fig. 1. With typical $E_{\text{th}}=0.1 \text{ V}/\mu\text{m}$, $\epsilon_a=14$, $K=10^{-11} \text{ N}$, $W=10^{-5} \text{ J}/\text{m}^2$, one finds that $(e-2P_s)\sim 10^{-10} \text{ C}/\text{m}$. As measured for 5CB, $e\sim 10^{-11} \text{ C}/\text{m}$ [12]. Thus the surface polarization can be significant, $(-2P_s)\sim 10^{-10} \text{ C}/\text{m}$. The last estimate is reasonable: a ferroelectric layer of molecules with dipoles ~ 4 debye and area $\sim 0.2 \text{ nm}^2$ per molecule would result in $|P_s|\sim 10^{-10} \text{ C}/\text{m}$. Of course, factors such as electric double layers, finite conductivity of coating layers, etc., would modify the numerical estimates.

Neither surface polarization nor the flexoelectric effect are capable of causing the optical-diode effect if the cell is bounded by two identical plates. The effect appears only when the two homeotropic plates are different and thus $\mathbf{P}_{s,1}$

$\neq -\mathbf{P}_{s,2}$ and $W_1 \neq W_2$; the indices 1 and 2 correspond to, say, the lower and the upper plates, respectively. Following Derzhanski, Petrov, and Mitov [3] and extending the above model to the asymmetric cell, one finds two thresholds for director deviations at the plates: $E_{\text{th},1}=W_1/(e-2P_{s,1}\mp\sqrt{\epsilon_0\epsilon_a K})$ and $E_{\text{th},2}=W_2/(-e-2P_{s,2}\mp\sqrt{\epsilon_0\epsilon_a K})$, respectively. The sign “-” should be taken for $E>0$ and “+” for $E<0$. These equations clearly show the possibility of optical diode effect. For example, a positive field would cause $\theta_1 \neq 0$ if $P_{s,1}<(e-\sqrt{\epsilon_0\epsilon_a K})/2$ and $(-e-\sqrt{\epsilon_0\epsilon_a K})/2<P_{s,2}<(-e+\sqrt{\epsilon_0\epsilon_a K})/2$. However, the field of negative polarity would produce no effect at all: \mathbf{n} remains homeotropic everywhere provided the indicated inequalities hold. With $|e|\sim\sqrt{\epsilon_0\epsilon_a K}\sim 10^{-11} \text{ C}/\text{m}$ and $|P_s|\sim(10^{-10}-10^{-11}) \text{ C}/\text{m}$ these inequalities can be satisfied in experimental cells.

To conclude, we experimentally observed a unipolar electrooptic effect in a nematic cell with specific confinement. Two surface-mediated mechanisms seem to brake the central symmetry of the cell: locally at each plate due to the surface polarization and globally due to the difference in the coating materials at opposite plates. Further studies can bring better understanding of surface anchoring and surface polarization in liquid crystals. One possible direction might concentrate on alignment materials with controlled electric polarization, such as electrically poled polymers or Langmuir-Blodgett layers.

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