"Thresholdless" hysteresis-free switching as an apparent phenomenon of surface stabilized ferroelectric liquid crystal cells

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(Received 15 November 2001; revised manuscript received 8 April 2002; published 2 August 2002)

The thresholdless, hysteresis-free V-shape *electro-optical* switching in surface-stabilized ferroelectric liquid crystals, observed usually with a triangular voltage form, has been shown to be rather an apparent and not a real effect. Strictly speaking, it is observed only at one characteristic frequency f_i and is accompanied by an inversion of the electro-optical hysteresis direction from the normal to the abnormal one. The switching *of the director* in a liquid crystal layer at f_i, in reality, has a threshold and a normal hysteresis. Even the optical transmittance shows a hysteresis at f_i when it is plotted as a function of the voltage on the liquid crystal layer and not as a function of the total voltage on the liquid crystal cell which always includes the inner insulating layers. Due to these layers, a voltage divider is formed which includes the capacitance of the insulating layers and the dynamic impedance (capacitance and resistance) of the ferroelectric liquid crystal layer. The new explanation has been confirmed by experiments with different ferroelectric liquid crystal cells combined with external resistors and capacitors and by measurements of a strong dependence of f_i on the liquid crystal resistance which was varied over three orders of magnitude. A theoretical analysis of the problem has also been made using certain approximations for material parameters and the space dependence of the sine form of the electric field in the liquid crystal layer. The conclusions are qualitatively consistent with the experimental results. Finally, the dynamic problem has been solved numerically by taking into account of all the relevant parameters (in the absence of flow and irregularities in the cell plane) and the obtained results are in excellent correspondence with the experiment. This has been demonstrated for sets of material and cell parameters providing the best V-shape performance.

DOI: 10.1103/PhysRevE.66.021701 PACS number(s): 61.30.Cz, 61.30.Gd, 42.79.Kr

I. INTRODUCTION

Ferroelectric liquid crystals (FLC's) having a chiral smectic C^* structure, possess an intrinsic polarity, and can be driven to "on" and "off" states by an external ac voltage. In a confined geometry, when the FLC structure is stabilized by surfaces, the two ferroelectric states have memory and the electro-optical switching of the FLC's manifests a threshold behavior with a characteristic hysteresis [1]. However, in some applications the threshold is undesirable because it does not allow for the realization of gray scale. Several publications deal with a so-called "V-shape" or "thresholdless" switching mode of ferroelectric and antiferroelectric (AFLC) liquid crystals [2,3]. In this case, a cell with a bookshelf alignment of smectic layers is placed between crossed polarizers with the smectic layer axis along the electric vector of the incident light. When a triangular voltage waveform is applied to the cell, in a certain (narrow) range of frequencies, the field-induced optical transmission acquires the V-shape form without threshold and hysteresis.

The thresholdless switching has been shown to be accompanied by a change of the hysteresis direction from the normal to the abnormal one [4]. For the latter, the polarization and optical axis do not seem to lag behind the field, but run ahead of the field, in contradiction with common sense. In fact, the genuine V-shape switching is observed only at the hysteresis inversion frequency (f_i) and this regime arises typically at very low frequencies, depending on the thickness d_p of the aligning layers (typically, at room temperature $f_i = 0.01-10$ Hz for $d_p = 30-100$ nm). In fact, as shown below, the genuine V-shape switching is only observed just at the hysteresis inversion frequency.

The mechanism for V-shape switching is currently under discussion in the literature. Fukuda's group insists [4,5] that this regime is only observed in particular antiferroelectric, ferrielectric, or other (like SmX^*) phases, in which the azimuthal direction of the tilt is "fluctuating" from layer to layer (frustrated phases) and the electric field aligns all the individual layer polarizations into one direction (two-dimensional Langevin model). In this consideration, the insulating layers are important by their indirect role in influencing the inversion of hysteresis due to the ionic charges accumulated on them [4].

Experiments, however, do not support the Langevin model for the SmX^* phase [6]. Instead, it has been con-

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cluded that V-shape switching is only possible when a liquid crystal cell has a chevron structure, and even a question has been posed if conventional ferroelectric phases might manifest V-shape switching. Rudquist *et al.* [7] (see also [3]) answer this question positively: according to their "block model," any conventional FLC with a large self-interacting spontaneous polarization (P_s) automatically forms a block of uniformly oriented local P_s with kinks in the director field at both interfaces. Under external fields the whole block is switched and the kinks play the role of a surface lubricant for the director reorientation. The dielectric layers are supposed to play a crucial role in keeping charges necessary to stabilize such a block-kink structure. Very high P_s and screening of the field in the liquid crystal layer are necessary in this model [8]. It has been especially stressed that the switching of polarization is thresholdless due to the formation of "blocks" mentioned above [9]. Despite the fact that numerical calculations made for a certain set of parameters seem to support this model, it contradicts the fact that FLC's with low and even very low P_s manifest the same phenomenon.

Other models [10,11] relate the V-shape switching to strong polar anchoring of AFLC's to polymer aligning layers although some experiments point towards the opposite tendency [4]. Numerical calculations [12] have shown a positive role of a thick (1 μm) polymer layer in contact with a conductive FLC layer in promoting V-shape electro-optical response. However, no experimental data have been presented to verify these calculations. Finally, Čopič *et al.* [13] have found theoretically the conditions under which a V-shape electro-optical response can be realized in FLC layers with alignment layers. It was assumed that slow, zero-field diffusion of ions through a FLC cell plays the main role in the effect, that the polarization is large, and that the electric torque is zero. The zero torque is required either for the field $\mathbf{E}=0$ or for the polarization $\mathbf{P}$ normal to the cell electrodes. The ionic free carriers make the internal electric field in the FLC's inhomogeneous. This modifies the director distribution and the optical properties of the cell. Again, no experiments have been reported to support this consideration. In general, the analysis of the literature shows that the mechanism for "V-shape" switching is still under discussion and even regarded as "mysterious." Therefore, up to now, no correct approach has been found for the development of either new materials or the technology for proper cell preparation.

It is very strange that, in all of the listed works, one extremely important point has been missed. In fact, the conditions for the correct observation of the hysteresis are not fulfilled: the polarization or the optical response must be plotted as a function of the voltage on the FLC layer and not as a function of the voltage on the two layer system including the FLC layer attached to a thick polymer layer.

Our experiments with a number of FLC's have shown that true V-shape switching is always observed only at the frequency (f_i) of hysteresis inversion, which is mainly determined by the parameters of the voltage divider formed by the aligning (or/and dielectric) layer and the FLC layer. Without such a divider the V-shape regime does not exist at all, and recent experiments of Seomun *et al.* [14] on cells prepared without aligning layers confirm this unequivocally. Besides the impedance, other properties of a FLC (P_s, tilt angle, viscosity, anchoring conditions, etc.) are also important factors for influencing both f_i and the particular form of the V-shape response (for example, the saturation voltage). The aim of the present paper is to demonstrate the experimental proofs of our new approach, and on the basis of theoretical consideration and computer modeling, to discuss the prospects for the development of new FLC materials (or probably even AFLC's) with a V-shape electro-optical response.

The paper is organized as follows. First we discuss the importance of the hysteresis inversion frequency f_i and coercive field, which are the fundamental parameters relevant to the problem. Then we consider the equivalent electric circuit of a FLC cell and the role of the voltage divider between the aligning (or additional dielectric) layer and the FLC layer. After this we demonstrate how easily one can manipulate the frequency f_i by varying the external capacitors and resistors. On this basis, we show the best performance characteristics of the real FLC cells. Then we theoretically consider the problem of hysteresis-free switching using an approximation of relatively small values of the spontaneous polarization and the resistivity of a FLC and find the distribution of the polarization and the electric field over the bulk of the FLC layer. Finally, we demonstrate the two particular results of our computer simulations of the problem for a set of material and cell parameters, providing the best performance characteristics of the V-shape electro-optical switching.

II. EXPERIMENT

In our experiments we used different FLC cells. All cells were of the sandwich type, consisting of two glass plates covered by transparent conductive films of indium-tin-oxide (ITO). On top of the ITO layers, aligning polyimide layers of different thickness d_p were deposited by spin coating. Their thickness was determined by atomic force microscopy (AFM), and as a rule, only one aligning layer was buffed. The gap between the plates was between 1 and 2 μm and installed with nanoscopic polymer balls located along the perimeter of the electrode overlapping area A. The thickness of the gap d was measured by the capacitance technique, and afterwards the cell was filled with a liquid crystal in the isotropic phase.

All measurements have been carried out only in the smectic C^* phase of the FLC's as shown in Table I.

A particular material and cell were chosen according to the idea of the experiment. The textures and phase transitions were studied under a polarizing microscope. The kinetics of the repolarization currents and of the optical response of the FLC's to triangular voltage form were investigated using a setup with an automatically rotating hot stage, a He-Ne laser, a photomultiplier, and a digital oscilloscope HP54815A.

III. HYSTERESIS INVERSION FREQUENCY

Figure 1 illustrates a change in the hysteresis direction for cell No. 1 with the following parameters: two alignment

TABLE I. FLC materials used in the present work ($T=25\,^{\circ}\mathrm{C}$).

Name	Phase sequence	P_s ($\mathrm{nC/cm^2}$)	Tilt angle Θ (deg)	Viscosity γ_φ (Pa s)[b]	Pitch (μm)
FLC-422 (LPI[a])	Cr-10 °C-SmC^*-58 °C-SmA-80 °C-Iso	100	23.5	0.7	∞
CS-1025 (Chisso)	Cr-−3 °C-SmC^*-62 °C-SmA-84 °C-N-90 °C-Iso	16.4	21	0.4	10
FLC-36 (LPI)	Cr→3 °C→SmC^*→35 °C→SmA→62 °C→Iso	21	15	0.3	∞
PBH-13 (LPI)	Cr-20 °C-SmC^*-87 °C-SmA-100 °C-N-101 °C-Iso	130	35	0.4	0.2

[a]Lebedev Physical Institute (Moscow).
[b]Rotational viscosity defined as $\gamma_\varphi=\gamma_0\sin^2\Theta$.

layers each of thickness $d_p=80$ nm, FLC layer thickness $d=1.7\,\mu$m, area $A=14\times18\,\mathrm{mm}^2$, and pitch-compensated liquid crystal mixture FLC-422 (see Table I). The optical transmittance of the cell induced by a triangular voltage form of amplitude ±8.3 V shows a typical thresholdless electro-optical behavior at a frequency of 7 Hz [Fig. 1(b)]. Below and above this frequency an abnormal and normal hysteresis is observed [see transmittance diagrams in Figs. 1(a) and 1(c), respectively] for 1 and 50 Hz (arrows show the route of the transmittance variation). It should be noted that there is another change of the hysteresis direction at a much higher frequency at which the shape of the transmittance-voltage curve is more similar to the Greek letter Λ than to the Latin V and the switching contrast is very low. We shall not discuss this case here.

The voltage difference between two transmission minima seen in Figs. 1(a) and 1(c) is the doubled threshold voltage $2U_{\mathrm{th}}$ for the FLC switching. The genuine V-shape switching corresponds to $U_{\mathrm{th}}=0$. In other words, at this condition the coercive field vanishes. The frequency dependence of the threshold voltage for the same cell was measured very carefully over the range of more than five orders of magnitude and is shown in Fig. 2, curve 1. In the curve, the negative (positive) values of U_{th} correspond to anomalous (normal) hysteresis. The curve is quite different from that predicted by a simple FLC model [15]. One can see that, instead of a finite frequency range usually reported in the literature, genuine (ideal) thresholdless switching is observed at only one frequency f_i. It is evident that such a “magic” frequency observed for all FLC’s studied must be related to a certain characteristic time $\tau_i=1/2\pi f_i$ dependent on material parameters.

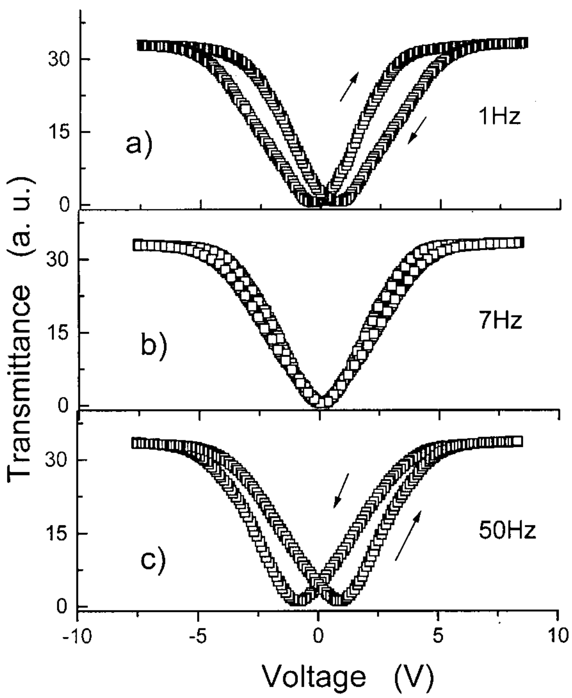

FIG. 1. Transmittance of cell No. 1 (FLC-422, $d=1.7\,\mu$m, two 80-nm-thick polyimide layers) controlled by a triangular voltage form ±8.3 V at different frequencies (from top to bottom): $f=1$, 7, and 50 Hz. Temperature 25 °C.

IV. VOLTAGE DIVIDER

Our principal idea is that the “thresholdless, hysteresis-free” V-shape switching is rather an apparent and not a real

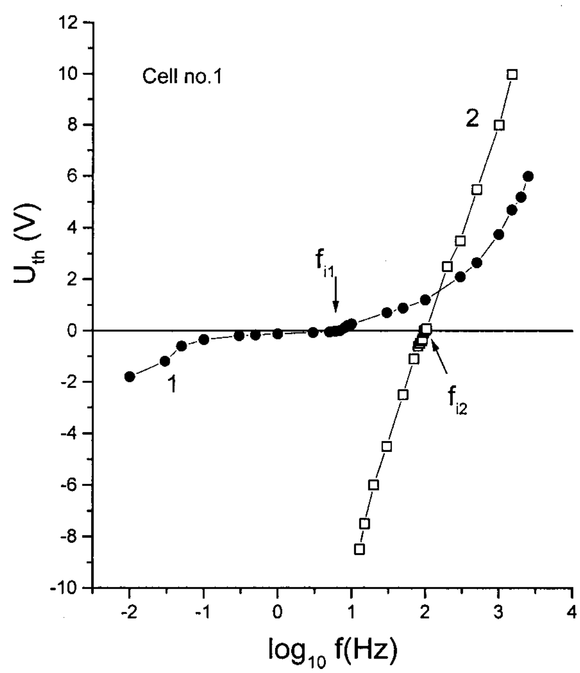

FIG. 2. Threshold voltage for optical transmittance as a function of frequency for cell No. 1 (FLC-422, $d=1.7\,\mu$m, two 80-nm-thick polyimide layers). Curve 1: triangular voltage ±9 V is applied directly to the cell electrodes. Curve 2: triangular voltage ±18 V is applied to the cell through a capacitor $C=22$ nF in series and resistor $R=180\,\mathrm{k}\Omega$ in parallel with the cell as shown in Fig. 3(c). Temperature 21 °C.

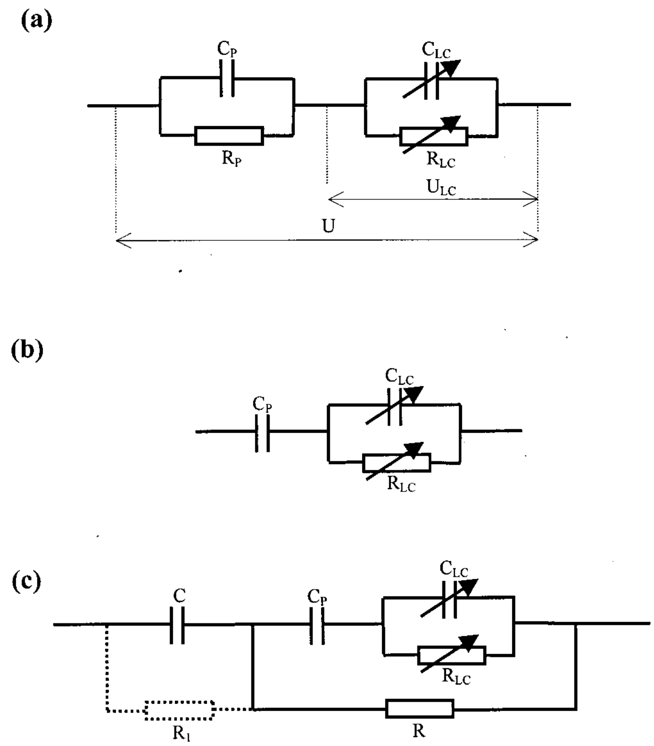

FIG. 3. Equivalent circuits. (a) C_p and R_p are the permanent capacitance and resistance of the polymer layer, C_{LC} and R_{LC} correspond to the dynamic capacitance and resistance of the FLC layer. U is total voltage applied to the cell, and U_{LC} is the voltage "seen" by the liquid crystal. (b) The same cell with resistance of polymer layer neglected. (c) Equivalent circuit of the cell with external elements C, R, and R_1.

effect. The switching of the director of the liquid crystal layer, in reality, always has a threshold and manifests the normal hysteresis if the optical transmittance is plotted as a function of the *voltage on the liquid crystal layer* and not as a function of the *total voltage on the liquid crystal cell*, which always includes inner aligning-insulating layers. Due to these layers, a voltage divider forms between the insulating layers and the FLC layer and the liquid crystal suffers a reduced voltage with a considerable change in its form with respect to the triangular voltage applied from a function generator.

Consider an equivalent electric circuit shown in Fig. 3(a). A typical cell with a V-shape response contains two thick polymer aligning layers, each about 50 nm thick, and a 2-μm-thick FLC layer. With a cell area of 1 cm^2, a dielectric constant of the polyimide $\varepsilon_p = 3.8$ and specific resistance $\rho_p \approx 10^{14}\ \Omega\,\mathrm{cm}$, the polymer layer capacitance and resistance are about $C_p \approx 35\,\mathrm{nF}$ and $R_p \approx 10^9\ \Omega$, respectively. The characteristic time constant is $\tau_p = R_p C_p \approx 35\,\mathrm{s}$. If we are not interested in frequencies $f \ll (2\pi\tau_p)^{-1} \approx 5\,\mathrm{mHz}$, the polymer resistance can be ignored and the circuit becomes simpler [see Fig. 3(b)]. For the sine form of the voltage, $U = U_0 \sin \omega t$, applied to the circuit as shown in Fig. 3(b) (with fixed parameters), the amplitude and phase δ of the voltage across the FLC layer can be calculated easily as

$$U_{\mathrm{LC}} = -\frac{\omega R_{\mathrm{LC}} C_p}{[1+\omega^2 R_{\mathrm{LC}}^2 (C_p + C_{\mathrm{LC}})^2]^{1/2}} U_0 \cos(\omega t + \delta),$$

where $\tan \delta = \omega R_{\mathrm{LC}}(C_p + C_{\mathrm{LC}})$. The characteristic time of the voltage divider is $\tau_d = R_{\mathrm{LC}}(C_p + C_{\mathrm{LC}})$ and is this time which mainly predetermines the hysteresis inversion frequency $\tau_d = \tau_i = 1/2\pi f_i$.

With increasing field the dielectric constant of the FLC's and therefore C_{LC} decrease by more than one order of magnitude due to the suppression of the Goldstone mode. In thin cells, the FLC resistance is also voltage dependent. Therefore, both C_{LC} and R_{LC} are dynamic components of the FLC impedance. It is also of principal importance that both of them, especially R_{LC}, are strongly dependent on temperature. This explains why V-shape switching is much easier observed at enhanced temperatures.

V. DEPENDENCE ON EXTERNAL ELEMENTS

In a FLC cell with V-shape response, the polymer layer plays several roles: it aligns the liquid crystal, forms a shoulder for the voltage divider, and is also capable of discharging any surface charges accumulated at the liquid-crystal–polymer interface. The liquid crystal plays the role of a switcher and also contributes to a change in the dividing ratio. In order to prove the unique role of the voltage divider, we can liberate the polymer from the second two functions and transfer them to the external electronic elements. The resistance of the liquid crystal can generally be controlled either by doping the material with ionic impurities or by temperature. However, we can also mimic these variations using external resistors parallel to the FLC resistance.

Figure 3(c) shows the same model cell as before with additional C, R, and R_1 elements. Two of them C and R play a crucial role in increasing the hysteresis inversion frequency. The resistor R_1 is optional: it provides a possibility to apply a dc bias voltage to the liquid crystal or to discharge the liquid crystal cell (in case of some asymmetry) from the electric charges accumulated on the capacitance C_p or the capacitor C.

Now we can show how the hysteresis inversion frequency is easily changed by the variation of the external elements. First, we consider cell No. 1 without external elements: $f_i = 1.5\,\mathrm{Hz}$ at $T = 20\,^\circ\mathrm{C}$ (note that f_i is extremely sensitive to temperature). With external C and R elements, as shown in Fig. 3(c), but without R_1, the inversion frequency increases dramatically. The optical transmittance voltage forms are shown in Fig. 4. With an additional capacitor $C = 22\,\mathrm{nF}$ alone, the frequency is shifted from 1.5 to 8.9 Hz. When resistors either $R = 180$ or 82 kΩ are connected in parallel to the cell, the inversion frequency is further shifted up to 99 and 159 Hz, respectively. f_i is increased by two orders of magnitude, e.g., 106 times, and the role of the RC constant is evident. The frequency dependence of the threshold voltage for the latter combination is displayed as curve 2 in Fig. 2.

The second example is even more demonstrative. Cell No. 2 is a standard 2-μm-thick (area $A = 0.16\,\mathrm{cm}^2$) cell filled with a Chisso CS-1025 mixture (see Table I) having a low value of spontaneous polarization. For such a cell no V-shape

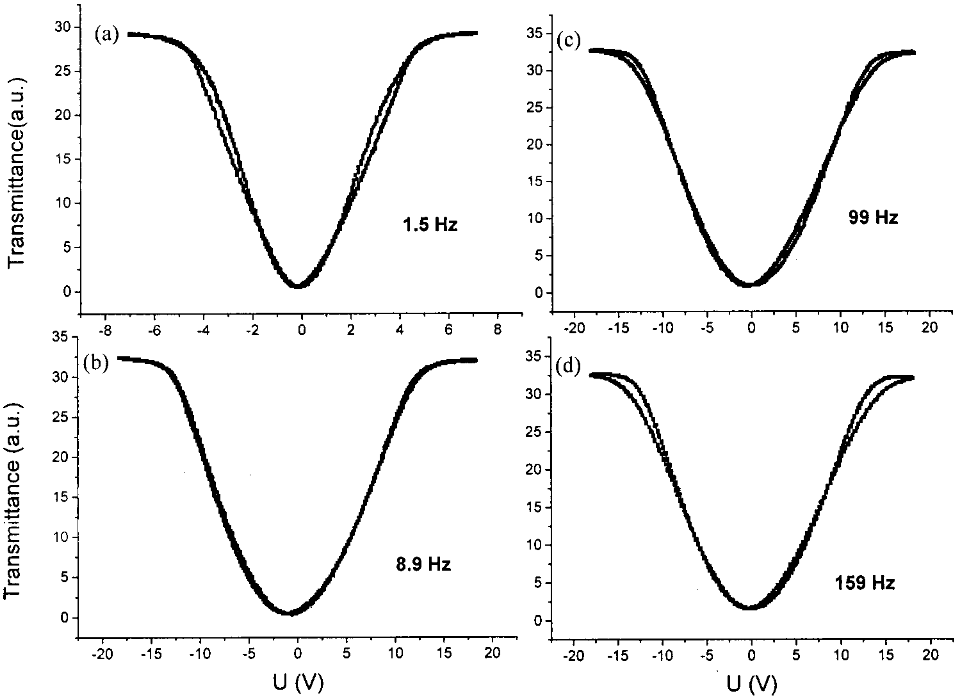

FIG. 4. V-shape optical transmittance of cell No. 1 (FLC-422, $d=1.7\ \mu$m, two 80-nm-thick polyimide layers) in different regimes: (a) without external elements, $f_i=1.5$ Hz; (b) with external capacitor $C=22$ nF, $f_i=8.9$ Hz; (c) with external capacitor $C=22$ nF and external resistor $R=180\ \mathrm{k}\Omega$, $f_i=99$ Hz; (d) with external capacitor $C=22$ nF and external resistor $R=82\ \mathrm{k}\Omega$, $f_i=159$ Hz. Temperature 20 °C.

switching is expected from any model discussed in the literature. But in fact, when the Chisso material is fresh the hysteresis inversion frequency was found to be at a very low frequency $f_i=0.7$ Hz. However, when cell No. 2 was filled with an aged and a more conductive material (small R_{LC}), as expected from the above consideration, the V-shape switching is observed at a higher frequency 3.5 Hz. With an external capacitor $C=2.7$ nF, the hysteresis inversion frequency increases 150 times and reaches 530 Hz, a value that has never been reported in the literature. The frequency dependences of the coercive field for both cases, with and without the capacitor, are very similar to those shown in Fig. 2.

The experiments with external capacitors allow measurements of the voltage on the FLC layer and follow the electro-optical hysteresis as a function of the voltage across that layer. Indeed, if a cell has no insulating layers at all and the voltage divider shoulder necessary for V-shape switching is formed by an external capacitance, we can measure the same electro-optical response as a function of two different arguments U and U_{LC} as shown in Fig. 3(a). However, to observe an electro-optical response we need at least one very thin aligning polyimide layer. In this case we used a 2-μm-thick cell No. 3 with a thin polyimide layer ($d_p \approx 40$ nm) on one of the plates. The cell was filled with the pitch compensated mixture FLC-36 (see Table I). In this cell the inversion of hysteresis takes place at frequency $f_i=0.25$ Hz. When an external capacitor $C=22$ nF is connected in series with the same cell, a typical V-shape switching is observed at an almost 100 times higher frequency $f_i=21$ Hz. At this frequency the aligning layer impedance may be neglected and the only reason for the V-shape electro-optical response is a change in the voltage on the FLC cell, which is controlled solely by the external capacitor.

In Fig. 5 the form of the external voltage, U, the form of the repolarization current oscillogram, I_p, and the form of the voltage on the liquid crystal cell, U_{LC}, are presented. It is well seen that the shape of U_{LC} is quite different from U. This difference modifies the hysteretic behavior of the FLC layer and the shape of the current oscillograms. For example, flat peaks of I_p seen in Fig. 5 are very similar to those reported, e.g., by Chandani *et al.* [4]. Now we can plot the same optical transmission in two different coordinates: namely, as a function of the total voltage U and as a function of the voltage on the liquid crystal cell, U_{LC}: see Fig. 6. As seen, a typical V-shape form is observed only when the optical transmittance is plotted as a function of the total voltage

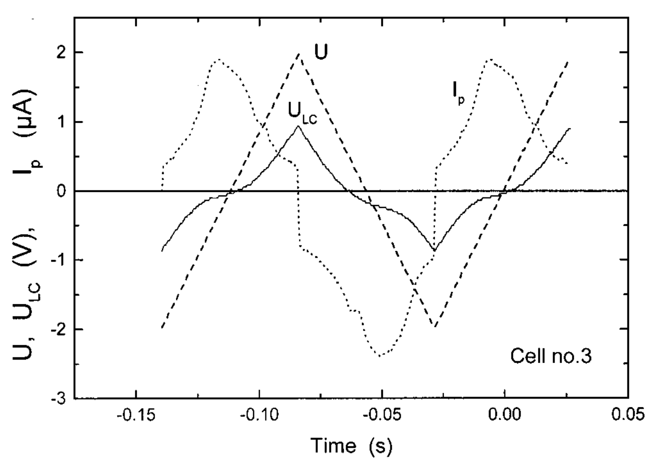

FIG. 5. Wave form of the external voltage U, repolarization current I_p, and voltage U_{LC} on FLC cell No. 3 (FLC-36, $d=2\ \mu$m, one 40-nm-thick polyimide layer) connected to a generator in series with external capacitor $C=22$ nF.

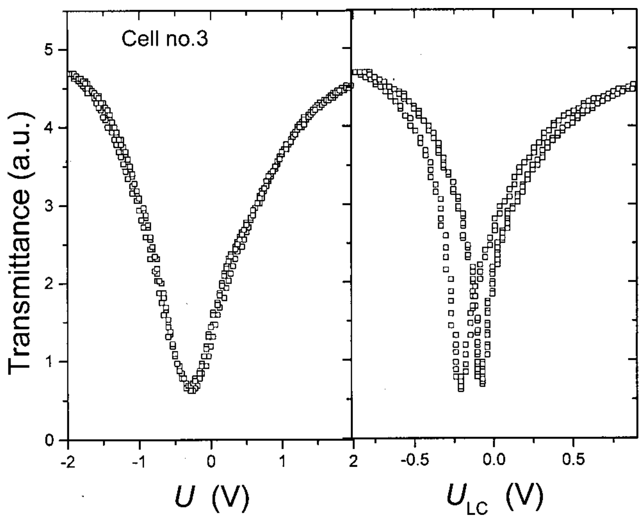

FIG. 6. Same optical transmittance of cell No. 3 (FLC-36, $d=2\ \mu$m, one 40-nm-thick polyimide layer) in different coordinates: namely, as a function of the total voltage $T(U)$ and as a function of the voltage on the cell, $T(U_{\mathrm{LC}})$.

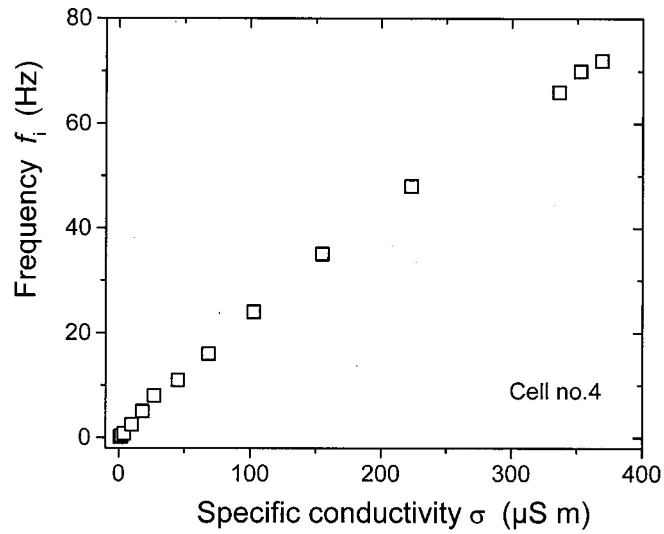

FIG. 7. Hysteresis inversion frequency f_i as a function of the specific conductivity σ for cell No. 4 (PBH-13, $d=1.4\ \mu$m, two 40-nm-thick polyimide layers).

$T(U)$ applied to the circuit including a capacitor C. However, a hysteresis typical for a conventional FLC's is clearly seen in curve $T(U_{\mathrm{LC}})$. The latter is slightly asymmetric because the cell has only one alignment layer. Note that the voltage on the FLC layer is considerably reduced with respect to the total voltage and this is an inevitable price we pay for the hysteresis-free V shape of the electro-optical switching. The same result has been obtained for several other cells having insulating layers of different thickness.

The possibility to control f_i over an enormous frequency range without any change in orientation and anchoring conditions for FLC materials shows that the latter conditions are factors of minor importance for performance of V-shape cells. Therefore, our experiments do not confirm earlier models [10,11], but are in accordance with more adequate computer modeling of FLC cells with polymer layers described in the last section.

VI. TEMPERATURE DEPENDENCE OF THE INVERSION FREQUENCY

As follows from Fig. 4, a resistor connected in parallel to a FLC cell dramatically increases f_i. Therefore, the same effect should be achieved by a change in the resistance of the FLC layer. This can be done using a variation of temperature without an external capacitance, just in the FLC configuration typical of the V-shape switching (i.e., with aligning-insulating layers). For this evaluation we have made cell No. 4 with $A=1.3\ \mathrm{cm}^2$ and two aligning polyimide layers of thickness $d_p=40$ nm, one of which was buffed. The thickness of the cell was $d=1.4\ \mu$m and the FLC used was PBH-13 (see Table I).

At each temperature in the range of the smectic C^* phase we determined R_{LC} and f_i. The dynamic resistance of the FLC's was measured using the repolarization current oscillogram *at the hysteresis inversion frequency* with the same triangular voltage form and amplitude as used for f_i measurements. For this purpose we determined the characteristic slope of the pedestal in the current oscillogram which is proportional to the FLC conductivity. It can easily be shown that the correct values of R_{LC} can be obtained for large enough C_p (for details see [16]). Upon increasing temperature from 24 to 85 °C the FLC layer conductance $G_{\mathrm{LC}}=1/R_{\mathrm{LC}}$ increased from 0.01 to 4.4 μS. Correspondingly, the hysteresis inversion frequency increased from 0.12 to 72 Hz. Evidently, f_i follows the increase in G_{LC} over the whole smectic C^* phase. This is seen in Fig. 7 where f_i is plotted as a function of specific conductivity $\sigma=G_{\mathrm{LC}}d/A$.

The dependence $f_i(\sigma)$ is intermediate between the linear one and the square root dependence predicted from the theoretical consideration: see Eq. (12) in Sec. VII. The deviation from the theory can be explained by violation of the assumption of large enough specific conductivity at low temperature. However, our results unequivocally show that the main process which controls the hysteresis inversion frequency, at which the true electro-optical V-shape switching is only observed, is just a specific voltage distribution between the aligning layers and the FLC layers. The voltage divider is mostly determined by the capacitance of the insulating layer C_p and the resistance of the FLC layer R_{LC}.

VII. THEORETICAL CONSIDERATION OF THE ELECTRO-OPTICAL HYSTERESIS INVERSION

From the experiments described above we can see that the apparent V-shape electro-optical effect is controlled by two sets of material parameters. The first group includes only the real and imaginary dynamic impedance components of both the aligning-insulating layers and the FLC layers. These parameters mainly determine the hysteresis inversion frequency at which the genuine V-shape switching is observed. The other group of parameters define the shape of the V curve (e.g., the optical contrast and the saturation voltage are controlled by the tilt angle Θ and the polarization P_s, respectively) and the speed of response (rotational viscosity γ_φ). In addition, the quality of the optical texture is also of great importance for good optical contrast. Leaving aside the technical problems, we may analyze theoretically a role of FLC material parameters responsible for a particular frequency and voltage at which the V-shape electro-optical switching occurs.

Since it has been found that the key parameter of a whole FLC cell is its *RC* constant, we should focus our attention on a possibility to have a proper correspondence between the different characteristic frequencies of the FLC itself (in addition to the period of the external electric field T_E or angular frequency ω). These frequencies are the space charge relaxation frequencies $\omega_{sc}=\sigma/\varepsilon_0\varepsilon$, orientational relaxation frequency $\omega_{or}=(Kq^2/\gamma_\varphi)$, inverse switching time $a=(P_sE/\gamma_\varphi\Theta^2)$, and another inverse switching time due solely to P_s ($\sim P_s^2/\varepsilon_0\gamma_\varphi\Theta^2$), where ε is the FLC dielectric constant, K is the elastic modulus, $P_s=\mu\Theta$, μ is the piezomodulus, and $2\pi/q$ is the characteristic length for the variation of the director orientation. In general, for the description of the switching process in an alternating electric field, we should take into account the following system of equations: (i) an equation for the distribution of director azimuthal orientation $\varphi(z,t)$ under the influence of an electric field $E_z(z,t)$ inside the LC layer, (ii) an equation for the distribution of $E_z(z,t)$ which follows from the conservation law for the external charge density ρ_{ion} and the Maxwell equation $\partial D_z/\partial z=\rho_{ion}$ with electric displacement $D_z=\varepsilon_0E_z+P_{sz}$, and (iii) a continuity equation for the concentration c_{ion} reflecting the mass conservation law which reduces to the diffusion equation in the absence of an electric current.

In reality, both the electrochemical and diffusion processes are of importance near the electrodes. These processes change the distribution of the electric potential near the cell boundaries, where a space electric charge exists of the order of the Debye length $r_D\approx(\varepsilon\varepsilon_0k_BT/e_{ion}^2c_{ion})^{1/2}$. Therefore, they modify the boundary conditions. Due to diffusion, an ionic distribution $c_{ion}(z,t)$ should be taken into account as has been done, e.g., in [13]. Thus the problem is very complicated, and in order to simplify it, an effective field E_i at distances of the order of r_D from the electrodes may be defined and included into the equations. The diffusion can be neglected in the equation for the bulk of the FLC layer if r_D is much smaller than the layer thickness d. For an analytic approach and for estimates, it is convenient to use an approximation of the form $E_z(z,t)=E_0\cos\omega t+\Xi(z,t)$, where the perturbation $\Xi(z,t)$ is small if the polarization P_s is not large. Hence we write

$$\gamma\Theta^2\frac{\partial\varphi}{\partial t}=K\Theta^2\frac{\partial^2\varphi}{\partial z^2}+P(E_0\cos\omega t+\Xi)\sin\varphi, \tag{1}$$

$$\frac{\partial\Xi}{\partial t}+\frac{\sigma}{\varepsilon_0}\Xi-\frac{P}{\varepsilon_0}\sin\varphi\frac{\partial\varphi}{\partial t}=0, \tag{2}$$

and will show that the phase shift in distributions $E_z(z,t)$ and $\varphi(z,t)$ is responsible for the hysteresis phenomena during the switching process. The perturbation Ξ will be found from Eq. (2) into which the magnitude φ will be substituted as a solution of Eq. (1). Although Ξ is assumed to be smaller than E_0, our qualitative results seem to be valid even at $\Xi\sim E_0$.

For estimation, we assume that the parameter a is larger than frequency ω and, correspondingly, $\varphi(z,t)$ smoothly changes over the FLC layer except in the narrow surface layers. Then the solution of Eq. (1) takes the form

$$\varphi(z,t)\approx\arctan\frac{1}{\sinh\left(-\frac{a}{\omega}\sin\omega t\right)}. \tag{3}$$

Near the cell boundaries, the φ value can be estimated by using an expansion

$$\varphi(z,t)=Bqz+(qz)^3g_1(t)+(qz)^5g_2(t)+\cdots, \tag{4}$$

where B is a constant, which shows that $\varphi=0$ at $z=0$ for strong anchoring conditions. The length q^{-1} is a result of the competition between electric and elastic forces near the electrodes. For example, in the static situation and in the presence of field E_i, from Eq. (1) we obtain that $\varphi=2\arcsin[\tan(qz)]$, where $q=(P_sE_i/K\Theta^2)^{1/2}$, φ being equal to zero at $z=0$ and $\varphi=\pi$ if q^{-1} is much less than z. The length q^{-1} is of the order of r_D if r_D is much smaller than the other lengths. The substitution of expression (4) into Eq. (1) allows the determination of magnitudes g_k, for instance,

$$g_1=-\frac{Ba}{6\omega_{or}}\cos\omega t. \tag{5}$$

In the present approximation we obtain the bulk value of Ξ from Eqs. (2), (3), and (4),

$$\Xi\approx\frac{4aP}{\varepsilon_0[(\sigma\varepsilon_0^{-1}-2a)^2+\omega^2]}[(\sigma\varepsilon_0^{-1}-2a)\cos\omega t+\omega\sin\omega t], \tag{6}$$

and the magnitude of Ξ near the surface is

$$\Xi\approx\frac{BaP\omega(qz)^4}{6\varepsilon_0\omega_{or}(\sigma^2\varepsilon_0^{-2}+\omega^2)}(\sigma\varepsilon_0^{-1}\sin\omega t-\omega\cos\omega t). \tag{7}$$

We see from Eq. (7) that Ξ is a rapidly increasing function of z. At $zq\sim1$, the parameters of Eqs. (6) and (7) should be approximately equal. Thus we get a condition which relates the characteristic frequencies in order to ensure the polarization reversal:

$$\omega^2\approx\sigma\varepsilon_0^{-1}(2a-\sigma\varepsilon_0^{-1}). \tag{8}$$

This condition is necessary for the existence of solutions to Eqs. (6) and (7) and a corrected solution for angle φ in the bulk with the maximum value of $\varphi\approx\pi$. Due to the boundary conditions and arbitrary values of a and ω, the values of φ in the bulk can be less than the maximum one.

The analytic estimation of transmitted light intensity $I(t)$ through a cell with distribution $\varphi(z,t)$ can be made for small tilt angles Θ and a corresponding weak change in the director orientation in the cell plane. In this case, we obtain, for the bulk,

$$I(t) \propto \sin^2(2\psi) \approx 4\Theta^2 \cos^2 \varphi. \tag{9}$$

Here $\psi \approx \Theta \cos\varphi$ is the angle which the light polarization vector forms with the orientation of the director in the cell plane: the phase retardation multiplier is discarded. To calculate the function $\varphi(t)$, we should substitute Ξ into Eq. (1) and solve the latter for the new field value E. This leads to substitution of the term $-(a/\omega)\sin\omega t$ in Eq. (3) by the term $-(a/\omega)\sin\omega t - (P/\gamma\Theta^2)\int \Xi\, dt$. From Eqs. (3) and (9) we then obtain the intensity

$$I(t) \propto \tanh^2\left[\frac{a}{\omega}\sin(\omega t - \delta)\right], \tag{10}$$

with $0 < \delta < \pi/2$. The minimum intensity $I_{\min}=0$ satisfies the condition

$$\tan\omega t = \tan\delta \approx \frac{\omega}{(\sigma\varepsilon_0^{-1} - 2a) + \tau[(\sigma\varepsilon_0^{-1} - 2a)^2 + \omega^2]}, \tag{11}$$

with $\tau^{-1} \equiv (4P_s^2\varepsilon_0\gamma\Theta^2)$. Equations (10) and (11) correspond to a phase shift in intensity $I(t)$ with respect to the *voltage* $U_{\mathrm{LC}}(t)$ *applied to the FLC layer*. Therefore, the absence of hysteresis in the optical transmission *with respect to the total voltage applied to the cell* $U = -U_0 \sin(\omega t - \delta^*)$, with $\tan\delta^* = \omega R_{\mathrm{LC}}(C_p + C_{\mathrm{LC}})$, corresponds to the condition $\tan\delta = \tan\delta^*$.

Equation (11) determines the magnitude of the hysteresis inversion frequency ω_{inv}. For the case $\tau\sigma/\varepsilon_0 \gg 1$ and C_p being not large, we obtain the following relationships from Eqs. (8) and (11):

$$\delta \approx \sqrt{\frac{R_{\mathrm{LC}}(C_{\mathrm{LC}} + C_p)}{\tau}} \ll 1, \quad a_{\mathrm{inv}} \approx \frac{\sigma}{2\varepsilon_0},$$

$$\frac{a_{\mathrm{inv}}}{\omega_{\mathrm{inv}}} \propto \sqrt{\frac{\tau\sigma}{\varepsilon_0}} \gg 1, \quad U_{\mathrm{LC}}(U_{0,\mathrm{inv}})/d \propto \tau\sigma P_s/\varepsilon_0,$$

$$\omega_{\mathrm{inv}} \approx \sqrt{\frac{1}{\tau R_{\mathrm{LC}}(C_{\mathrm{LC}} + C_p)}},$$

$$U_{0,\mathrm{inv}} \approx \frac{d\sigma}{2\varepsilon_0}\frac{\gamma\Theta^2}{P_s C_p}\sqrt{\frac{\tau(C_{\mathrm{LC}} + C_p)}{R_{\mathrm{LC}}}}. \tag{12}$$

Thus, in order to have hysteresis-free electro-optical switching, the characteristic frequencies mentioned above must be tightly related to each other and the hysteresis inversion frequency is an increasing function of the decreasing capacitance C_p of the orienting layers and the resistance of the FLC layer (in agreement with Figs. 4 and 7). Then ω_{inv} increases with $\tau^{-1/2}$—that is, with increasing P_s and decreasing viscosity γ. This is also in accordance with a general tendency found experimentally and by modeling. Besides, Eqs. (12) shows that, in general, the hysteresis-free situation only occurs when the frequency and amplitude of the external voltage have unique values at given parameters of an FLC and alignment layers (or external circuit elements). This is confirmed by numerical calculations presented in the next section. In fact, Eqs. (12) describe the most suitable conditions for the effect to be observed. A deviation from those conditions—for example, a change in U_0—must lead to less-pronounced V-shaped switching. In reality, the distribution $\varphi(z,t)$ has a weak spatial change in the bulk and, therefore, Eqs. (3), (6), (9), and (10), corresponding to an approximation for $a \gg \omega$, have a qualitative character.

VIII. COMPUTER MODELING

As was shown in the experimental part, the hysteresis inversion frequency f_i and the particular form of the optical transmission curve $T(U)$ depend on the parameters of the FLC's and aligning layers of capacitance C in a very complicated way. To simulate this effect one has to solve two sets of equations: (i) dynamic equations for the director $n(t,z,U)$ coupled with electric current equations like Eqs. (1) and (2) of the previous section, but more general, and (ii) Maxwell equations in order to find the optical transmission $T(t,U)$ for different polarization, wavelength, and coherency of light incident onto a cell.

The main difficulties in the simulation result from the fact that the voltage on a FLC layer U_{LC} cannot be fixed. For a circuit shown in Fig. 3(b) one can only fix the voltage on the cell electrodes U and let voltage U_{LC} change due to cell switching. In this case, it is simpler to operate not with an electric field $E(z)$, but with the electric displacement independent of z. This procedure has been carried out in the present work. The problem has been simplified in the sense that the director distortion has been treated as unidimensional (along the z axis) and flows were disregarded. The following contributions have been taken into account in the Lagrange-Euler equations for the chiral SmC^* phase: (i) a viscoelastic term including the molecular tilt angle Θ, rotational viscosity coefficients γ_i, Frank moduli K_{ii}, layer compressibility B_c, and spontaneous twist and bend coefficients, (ii) an electric term including spontaneous polarization P_s, dielectric tensor component ε_{ii}, and applied voltage of arbitrary form, and (iii) a surface term including variable pretilt angles ϑ_s, azimuthal W_a, and zenithal W_z anchoring energy. In the equations for the electric current, the electric properties (capacitance and conductivity) of aligning layers and FLC's were taken into account (the diffusion current in an FLC was neglected). $T(t,U)$ was calculated using the Berreman matrix method and a new algorithm described in [17]. At this stage, the properties of all optical elements (light source, polarizers, conductive glasses, aligning layers, and FLC layer) were taken into account.

The details related to the software and numerical procedure are out of the scope of the present paper. Before simulating the V-shape electro-optical regime in the cells with rather thick aligning (dielectric) layers, we have tested the program on simpler model cells and always had excellent

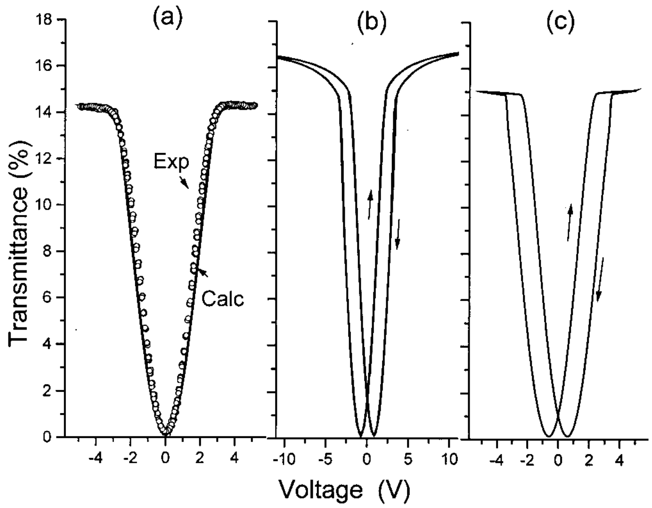

FIG. 8. Result of the computer modeling of cell No. 4 (PBH-13, $d=1.4\ \mu$m, two 40-nm-thick polyimide layers, 40 °C). (a) $U_m=5$ V, $G=0.07\ \mu$S; (b) $U_m=15$ V, $G=0.07\ \mu$S; (c) $U_m=5$ V, $G=1\ \mu$S (see text for other parameters).

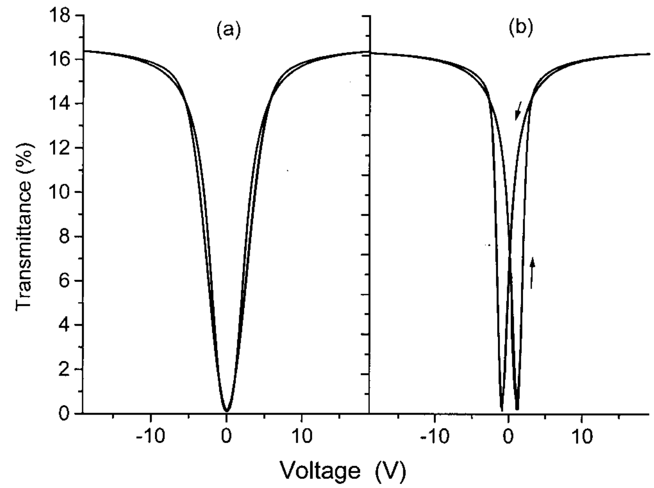

FIG. 9. Result of the computer modeling of the cell with the highest frequency (100 Hz) for V-shape electro-optical switching. Aligning layer capacitance $C=26$ nF (a) and $C=50$ nF (b) (see text for other parameters).

agreement with experiments. Here we show two examples related to V-shape switching which confirm the most important conclusions of the experimental and theoretical work presented in the previous sections.

The result of modeling made with parameters of cell No. 4 at 40 °C is shown in Fig. 8. The most important parameters (group 1) were taken from independent experiments: the others (group 2) were taken from the literature, e.g., [3]. The parameters used are the following.

Group 1. (a) FLC material PBH-13 (40 °C): $P_s=1.2\times10^{-3}$ C/m^2, $\Theta=35°$, $\gamma_0=0.6$ Pa s, $\varepsilon_{ii}=3$, $K_{ii}=1$ pN, $G=1/R_{\mathrm{LC}}=0.07\ \mu$S, helical pitch $p_0=0.2\ \mu$m, $n_\perp=1.521$, and $n_\parallel=1.726$. (b) Cell parameters and voltage: $d=1.4\ \mu$m, $A=1.3$ cm^2, $d_p=40$ nm (2 polyimide layer capacitance $C=50$ nF), triangular voltage $U_m=5$ V, and $f=2.5$ Hz.

Group 2. Pretilt angles 4° on both sides, $W_a=0.05$ mJ/m^2 and $W_z=0.5$ mJ/m^2 on both sides, and compression modulus $B_c=100$ kPa [in the Landau expansion very close to the phase transition T_c it is equal to $a(T-T_c)$; far from the transition it has to be measured].

In Fig. 8(a) the solid line corresponds to the numerical calculation with the listed parameters and the curve with symbols is the experimental one obtained in the course of experiment described in Sec. VI. Note that there was no fitting parameter (parameters from group 2 only slightly influence the result of calculations). There is discrepancy between the cell conductivity $G=0.07\ \mu$S used in the calculation and the experimental value taken from the current oscillogram at 40 °C ($G_{\mathrm{expt}}=0.12\ \mu$S). Since G_{expt} is a very steep function of temperature, the coincidence of the two curves may be considered as excellent. We have repeated the same procedure for 80 °C with independently measured $P_s=0.6\times10^{-3}$ C/m^2, $\Theta=20°$, $\gamma_0=0.3$ Pa s and $G=6.5\ \mu$S and without any fitting parameters obtained complete coincidence of the calculated and measured $T(U)$ curves at hysteresis inversion frequency $f_i=65$ Hz [not shown in the figure, because they are almost the same as the curves in Fig. 8(a)].

The calculated curve in Fig. 8(b) differs from that of Fig. 8(a) only by the amplitude of the applied triangular voltage (now $U_m=15$ V, parameters again taken for $T=40$ °C). In this case the hysteresis inversion frequency f_i increases to 20 Hz and at the operating frequency of 2.5 Hz we can see a typical ferroelectric loop with *anomalous* hysteresis (marked by arrows). Thus the voltage dependence of f_i is evident, as predicted by Eq. (12). The calculated curve of Fig. 8(c) differs from the initial curve (a) only by the value of the cell conductivity, $G=1\ \mu$S. Increasing G also shifts f_i to the new frequency of 25 Hz, and because $f<f_i$, once again we have a pronounced *anomalous* hysteresis. This demonstrates the importance of precisely tuning of the cell conductivity to get V-shape switching at a particular frequency.

The second example of the calculation is shown in Fig. 9, corresponding to our best experimental cell showing V-shape switching at frequency 100 Hz ($T=40$ °C). The FLC material is PBH-13, the cell thickness and the electrode areas are now 1.1 μm and 1.3 cm^2, respectively, and the applied voltage is $U_m=20$ V. A higher voltage is necessary because the aligning layers are thicker ($d_p=80$ nm). Correspondingly, their capacitance $C=26$ nF was taken for the calculations. Figure 9(a) shows the calculated V-shape curve coinciding with the experiment when the cell conductivity is fitted to $G=1.3\ \mu$S. This value of G is higher than that of the previous example, but is still quite reasonable and corresponds to the specific resistance of $10^{11}\ \Omega$ cm. The role of the capacitance is seen from Fig. 9(b): with all the other parameters kept as in Fig. 9(a), but for $C=50$ nF (40-nm-thick aligning layers), the hysteresis inversion frequency decreases down to 49 Hz and at the operating frequency of $f=100$ Hz a pronounced *normal* hysteresis is observed, as predicted by Eq. (12).

From the above consideration, it follows that for the optimization of the cells operating in the V-shape mode one should first choose an FLC material providing a high optical transmission in the on state, which means having a large tilt angle and a proper thickness of an FLC layer in order to reach an optimum value of $\Delta nd \approx k\lambda/2$, where $k=1$ (transmittive displays) or $k=1/2$ (reflective displays). Then it is necessary to optimize the capacitance C_p and increase the conductivity of the material (e.g., by doping or by increasing the temperature) to maximize the hysteresis inversion frequency. The spontaneous polarization may be varied within rather large limits, but large P_s and low viscosity increases f_i. The elastic properties and the anchoring conditions seem to be important only from the viewpoint of the dark off-state texture which is necessary for high optical contrast.

IX. CONCLUSION

The electro-optical and the polarization switching experiments have been carried out with FLC surface-stabilized cells having different aligning-insulating layers as well as with the same cells combined with external resistors and capacitors. The experiments allowed us to conclude that the thresholdless, hysteresis-free V-shape electro-optical switching in ferroelectric liquid crystals is observed, strictly speaking, only at the hysteresis inversion frequency f_i and is an apparent and not a real effect. The switching of the director in a liquid crystal layer at f_i has a well-pronounced threshold and a normal hysteresis. The latter can be easily observed if the optical transmittance is plotted as a function of the voltage on the liquid crystal layer and not as a function of the total voltage on the liquid crystal cell which always includes the inner insulating layers. Due to these layers, a voltage divider forms which includes the capacitance of the insulating layers and the dynamic impedance (capacitance and resistance) of the ferroelectric liquid crystal layer. Hence the liquid crystal layer suffers a reduced voltage with a considerable change of the voltage form as compared to the triangular voltage applied from a function generator.

This explanation of the phenomenon has been confirmed by experiments on different FLC's with variable thickness of aligning layers and by measurements of a strong dependence of f_i on liquid crystal resistance varied over three orders of magnitude. Despite all measurements being performed in the smectic C^* phase, we believe that the same principle is also valid for the antiferroelectric or other liquid crystal phases (like SmX^*) dynamically switched between two ferroelectric states. The theoretical consideration resulted in an analytical expression for the hysteresis inversion frequency and is completely consistent with observed tendencies. Numerical calculations of the optical transmittance of FLC cells made for operation in the V-shape regime show excellent agreement with experimental data.

ACKNOWLEDGMENTS

The authors are grateful to D. Ganzke, V. M. Shoshin, Yu. P. Bobylev, and A. L. Andreev for technical help and discussions. W. H. acknowledges financial support from the Volkswagen Foundation, S.A.P. and S.P.P. acknowledge support from Russian Foundation for Basic Research (Project Nos. 00-02-17801 and 01-02-16287, respectively), and S.A.P. also appreciates support from the Alexander von Humboldt Stiftung.

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