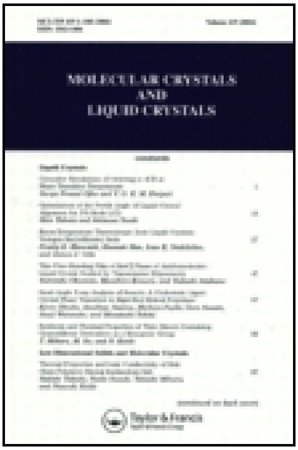

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Modeling electrooptical effects in Ferroelectric Liquid Crystals. 2. V-Shape Switching in the SmC* phase

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MODELING ELECTROOPTICAL EFFECTS IN FERROELECTRIC LIQUID CRYSTALS. 2. V-SHAPE SWITCHING IN THE SmC* PHASE

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The thresholdless, hysteresis-free V-shape electrooptical switching in surface stabilized ferroelectric liquid crystal (FLC) cells has been modeled and the results compared with experimental data. High frequency V-shape switching (at $f > 100\,Hz$) was experimentally demonstrated and simulated numerically.

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The key role of the capacitance C of FLC aligning layers has been underlined: thin layers with large C promote the well known bistability effect, thick layers, on the contrary, are necessary for V-shape switching. The results of modeling are in quantitative agreement with experiment.

Keywords: ferroelectricity; liquid crystals; simulation; V-shape effect

I. INTRODUCTION

When ferroelectric liquid crystals (FLC) with a chiral smectic C^* structure are stabilized by surfaces, the two ferroelectric states have memory and electrooptical switching of the FLC manifests a threshold behavior with a characteristic hysteresis (bistability phenomenon) [1]. Several publications deal with a so-called "V-shape" or "thresholdless" switching mode of ferroelectric and antiferroelectric (AFLC) liquid crystals [2]. That switching of transmission Tr by triangular voltage U_{tr} at frequency $f_{\mathrm{i.}}$ was shown to be accompanied by a change of the hysteresis direction from the normal to the abnormal one when the voltage across the cell electrodes lags behind the electrooptical response! ([3]. This regime arises typically at very low frequencies, dependent on the thickness d_{p} of the aligning layers. Several mechanisms were suggested for explanation of the phenomenon [3–10], however the problem is still far from the final solution. Recently we have experimentally shown that the capacitance of aligning (or insulating) layers and conductivity of the FLC layer play the key role in the appearance of the V-shape effect [11]. Some experimental results were confirmed by computer modeling based on numerical procedure developed by Palto [12,13]. A brief description of the basic equations and experimental tests of the computing procedure have been presented in the previous paper [14]. Here we apply the modeling procedure to the experimentally observed features of the V-shape switching. All important material parameters used in calculations were measured independently for the same FLC used in experiments. We have obtained excellent agreement between the calculated and experimental results.

2. MATERIALS AND CELLS

All measurements were carried out in the smectic C^* phase of pyrimidine compound based mixtures shown in Table 1. For these materials the temperature dependencies of spontaneous polarization, P_{s}, tilt angle Θ, rotational viscosity γ_{φ} (or γ_2) and helical pitch p are available.

All our cells were of sandwich type consisting of two glass plates covered by transparent conductive films of Indium-Tin-Oxide (ITO). When PBH-13

TABLE 1 FLC Materials Used in the Present Work (Parameters are Given at $T = 25°C$)

Name	Phase sequence	P_s (nC/cm^2)	Tilt angle Θ (deg)	Viscosity[b] γ_φ; γ_2 (Pa·s)	Pitch p (μm)
PBH-13 (LPI[a])	Cr-20°C-SmC*-87°C-SmA -100°C-N-101°C-Iso	130	35	0.4; 1.2	0.2
ZhK-438 (LPI)	Cr-12°C-SmC*-82C*-SmA -103°C-Iso	85	28	0.27; 1.22	0.2

[a]Lebedev Physical Institute (Moscow).
[b]Rotational viscosity is defined as $\gamma_\varphi = \gamma_2 \sin^2\Theta$.

was used, on the top of ITO were deposited 70–90 nm thick polyimide (PI) aligning layers. In case of ZhK-438, insulating Al_2O_3 (thickness 70 nm) and additional 30 nm thick aligning PI layers were deposited. In both cases only one PI layer was buffed. The gap d between the plates was installed using calibrated glass balls and measured by a capacitance technique before the cells were filled with a FLC in the isotropic phase.

3. EXPERIMENTAL RESULTS AND MODELING

a) Dependence of Hysteresis Inversion Frequency on FLC Layer Thickness

For high optical contrast in the V-shape regime, the optimum thickness has to be about 1.4–1.5 μm and 0.7–0.8 μm for displays operating in the transmission and reflection modes, respectively. In paper [11] we have presented some computation results for a 1.4 μm thick cell. At room temperature the hysteresis inversion frequency f_i in such thick cells is between 1 and 10 Hz depending on PI layer thickness and FLC conductivity. Our experiments carried out with wedge form cells have shown that, with decreasing thickness from 2.5 down to 0.8 μm, the frequency f_i increases one order of magnitude. We can only model FLC cells with uniform gap. The result is shown in Figure 1 for material PBH-13 and applied triangular voltage $\pm$ 10 V. As cell conductivity G depends on d the specific conductivity of the material was fixed at $\sigma = 10^{-9}$ (curve 1) and $\sigma = 2{\cdot}10^{-8}$ Ohm^{-1}m^{-1} (curve 2). The other measured parameters were as follows: cell area S = 1 cm^2, $P_s = 1.2{\cdot}10^{-3}$ C/m^2, $\Theta = 34$ deg, $\gamma_2 = 0.6$ Pa.s, $p = 0.2$ μm, optical anisotropy $\Delta n = 0.195$ ($\lambda = 633$ nm), at $T = 40°C$, capacitance of two aligning layers $C = 35$ nF.

However, in addition to the listed parameters some others have been taken for the calculations: Frank elastic moduli $K_{1,2,3} = 15$, 3 and 6 pN,

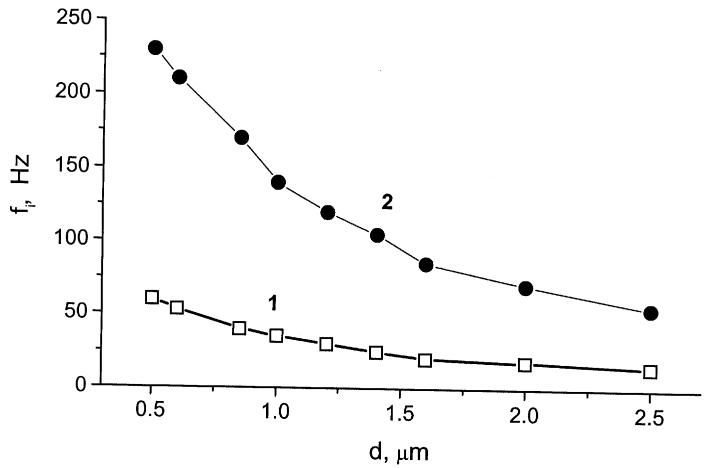

FIGURE 1 Calculated dependencies of hysteresis inversion frequency on FLC layer thickness for two different specific conductivity of PBH-13 (40°C) mixture, $\sigma = 10^{-9}$ and $2 \times 10^{-8}\,\Omega^{-1}\mathrm{m}^{-1}$. Other parameters are specified in the text.

respectively; compression modulus $K_4 = 5\,\mathrm{MPa}$ (values higher than 5 MPa only weakly influence the results of calculations but increase the calculation time); the viscosity ratio $R = \gamma_{1,3}/\gamma_2 = 3$ in accordance with the results of [14]; ordinary refraction index $n_0 = 1.53$ (typical value for these materials); background dielectric constants $\varepsilon_i = 3$ and typical values of azimuthal and zenithal anchoring energy $W_a = 0.05$ and $W_z = 0.5\,\mathrm{mJ/m^2}$. The smectic layer normal was assumed parallel to the substrates (bookshelf structure) because tilted layers and chevrons are hardly compatible with good V-shape form of the $Tr(U_{tr})$ curve.

From Figure 1 it is well seen that high frequency V-shape transmission should be observed in thin cells filled with high conductive material. Indeed, thin cells filled with ZhK-438 mixture manifest thresholdless behavior at frequencies of the order of 100 Hz. An example is shown in Figure 2 (curve 1). In this case, the voltage is $\pm 5\,\mathrm{V}$ ($f = 95\,\mathrm{Hz}$), the cell thickness and temperature are 0.85 μm and 30°C. The experimental curve has been modeled with the ZhK-438 parameters taken at 30°C: $S = 1\,\mathrm{cm^2}$, $P_s = 0.82 \cdot 10^{-3}\,\mathrm{C/m^2}$, $\Theta = 26\,\mathrm{deg}$, $G = 4\,\mu\mathrm{S}$, $\gamma_2 = 1.2\,\mathrm{Pa.s}$, $p = 0.2\,\mu\mathrm{m}$, optical anisotropy $\Delta n = 0.195$ ($\lambda = 633\,\mathrm{nm}$), capacitance of two aligning layers $C = 35\,\mathrm{nF}$ and other parameters listed in the previous paragraph. The only fitting parameter is the maximum transmission of the experimental cell (at level 0.046 predicted by modeling). The result of calculation is shown in Figure 2 (curve 2). The two curves are similar, however, due to some texture inhomogeneities the full darkness is not reached at the minimum of the experimental curve (the contrast is not very high). Below we

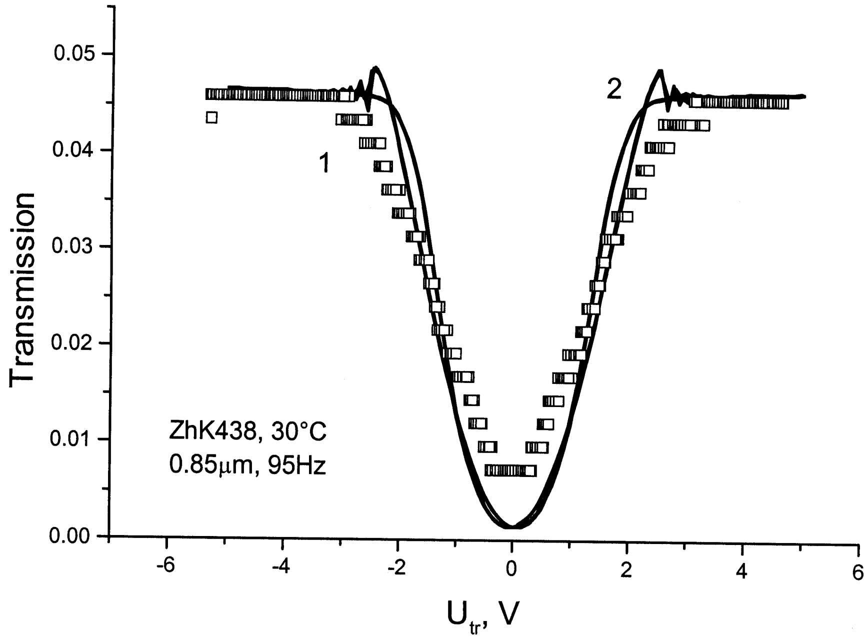

FIGURE 2 Experimental (1) and calculated (2) oscillograms of the optical transmission of 0.85 μm thick cell filled with Zk-438 (30°C) as function of triangular voltage ±10 V, f = 95 Hz. Other parameters are specified in the text. [Note, that small oscillations in curve (2) have no physical sense and should be disregarded; in fact, they disappear when the number of z-points along the cell thickness increases from 300 to 900, however, such a procedure results in the 30 times increase in calculation time].

shall vary separately the most important parameters of our calculations. First, consider the role of the capacitance of insulating-aligning layers (all the remaining parameters are fixed).

B) Dependence of Threshold Voltage and Inversion Frequency on Aligning Layer Thickness

This dependence was calculated for PBH-13 material for which the V-shape switching is found in the range 100–200 Hz and even higher depending on conductivity. All parameters are the same as in Figure 1, but the cell conductivity is assumed to be $G = 5\,\mu S$. At f_i the distance between the two transmission minima, that is the double threshold voltage $2U_{th}$ is exactly zero. In our case, for C = 35 nF the frequency $f = 200$ Hz is close to the genuine inversion frequency, therefore, if we fix the frequency at 200 Hz and vary C, the function $2U_{th}(C)$ would pass through the zero point at some value of C close to 35 nF. In fact, it happens at $C_0 = 44$ nF. It is moreinstructive, however, to plot $2U_{th}$ as a function of the thickness of a PI aligning layer deposited on each of the two ITO surfaces and having $\varepsilon = 3.75$. Evidently such a curve should have a minimum at $d_p(0) = \varepsilon\varepsilon_0 S/2C_0 = 37$ nm. Indeed we see the zero point in curve (1) plotted in Figure 3. At the same time the genuine inversion frequency ($U_{th} \equiv 0$ at f_i) grows systematically with increasing thickness d_p, see Insert to Figure 3. With thick layers it is possible to reach very high values of f_i at the cost of the enhanced voltage across a cell because the voltage on a FLC layer dramatically decreases with d(curve 2 in the main plot). It is interesting that for $d_p > d_p(0)$ and fixed $f = 200$ Hz $< f_i$ the threshold voltage U_{th} is of the order of 0.1 V over a wide thickness range and the $Tr(U_{tr})$ curve still has a shape similar to the V-letter with some *abnormal* hysteresis hardly seen. For $d_p < d_p(0)$ the *normal* hysteresis is observed.

C) Dependence of the Threshold Voltage on FLC Conductivity

Next, we analyze the role of FLC conductivity with fixed thickness of a PI layer at $d_p = 60$ nm ($C = 27$ nF). In the main plot of Figure 4 the cell conductivity is varied from 10^{-3} to 10^{2} μS (it corresponds to specific conductivity of a FLC of $\sigma \approx 10^{-11} - 10^{-6}\,\Omega^{-1}\mathrm{m}^{-1}$). The distance between the two

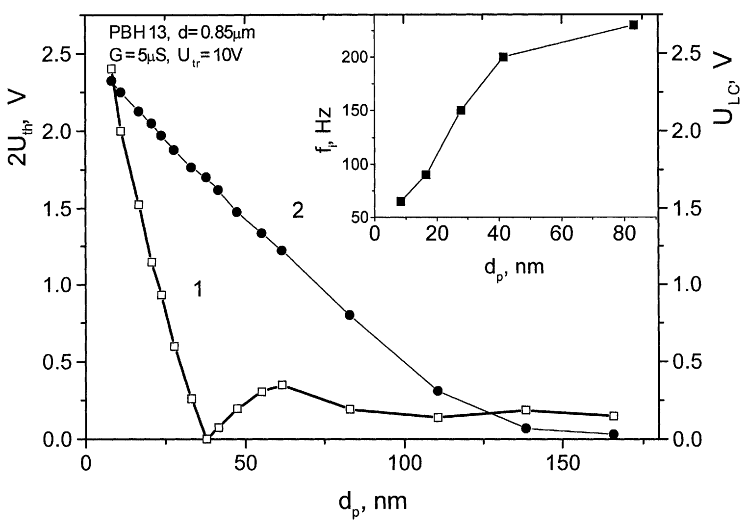

FIGURE 3 Calculated dependencies of the double threshold voltage applied to the cell $2U_{th}$ (curve 1) and the voltage on the PBH-13 (40°C, $G = 5$ μS) layer (curve 2) as functions of aligning PI layer thickness d_p. Insert: dependence of the hysteresis inversion frequency on d_p.

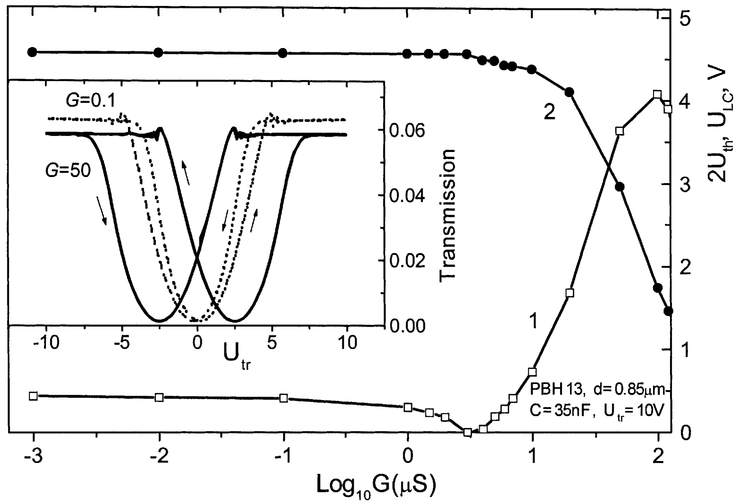

FIGURE 4 Calculated dependencies of the double threshold voltage applied to the cell $2U_{th}$ (curve 1) and the voltage on the PBH-13 (40°C) layer (curve 2) as functions of FLC conductivity G, C = 35 nF/cm^2. Insert: two loops showing a normal (for $G = 0.1\,\mu$S) and abnormal (for G = 50 μS) hysteresis.

transmission minima passes through the zero point at about $G(0) = 3\,\mu$S ($\sigma = 2.5 \times 10^{-8}\Omega^{-1}\text{m}^{-1}$, close to our experimental value at $T = 40°$C). For $G < G(0)$ and $G > G(0)$ the *normal* and *abnormal* hysteresis is observed, respectively. This is shown by arrows at the hysteresis loops for $G = 0.1$ and $50\,\mu$S in the Insert to Figure 4. As expected, for very large G the voltage across the FLC layer dramatically decreases, see curve 2 in Figure 4.

It is of great interest to see the shape of the voltage across an FLC layer for different operating frequencies, satisfying conditions $f = f_i, f > f_i$ and $f < f_i$. This situation is very difficult to realize experimentally (in [11] very thin aligning layers and external capacitors were used). The result of modeling is shown in Figure 5. Curve 1 corresponds to very thin layers ($d_p = 17\,\text{nm}, f_i = 90\,\text{Hz}, f = 200\,\text{Hz} > f_i$) and the shape of the voltage on a FLC layer (U_{LC}) almost repeats the triangular voltage $U_{tr} = \pm 10\,\text{V}$ (not shown). Curve 2 corresponds to the V-shape regime ($d_p = 47\,\text{nm}$, $f_i = 200\,\text{Hz}, f = f_i$): the voltage across FLC is approximately twice as low as the total voltage U across the cell and has a very characteristic features, namely, small zero-voltage "shelves" in vicinity of $U = 0$. Namely these zero-field shelves provide "thresholdless" optical switching as *was assumed* in earlier calculations [6,11]. With further increase in layer thickness (for curve 3 $d_p = 110\,\text{nm}, f_i = 250\,\text{Hz}, f < f_i$) the voltage across FLC

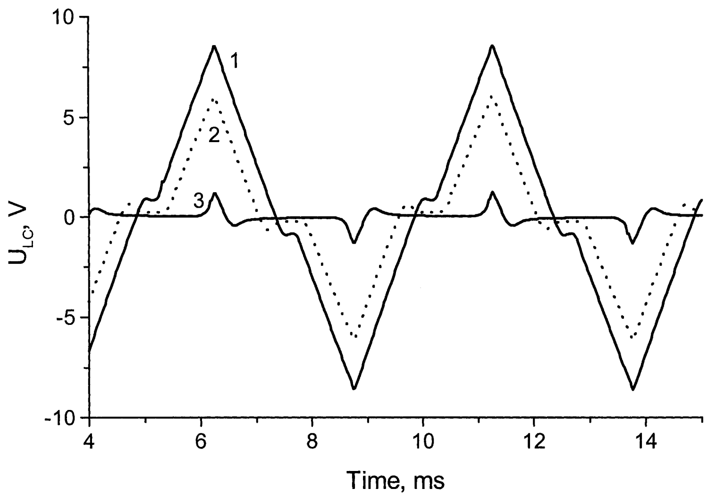

FIGURE 5 Calculated form of the voltage on PBH-13 (40°C) layer for a triangular voltage $\pm 10\,\mathrm{V}$ ($f = 200\,\mathrm{Hz}$) applied to the cell. Capacitance of aligning layers is varied: $C = 100$ (1), 35(2, V-shape regime) and $15\,\mathrm{nF/cm^2}$(3).

becomes very small and this regime would require for a considerable increase of the external voltage.

D) Response to Polar Pulses with High Duty Ratio

It has been found experimentally that FLC cells with thick aligning layers never show bistable response to polar pulses of high duty ratio. Such a case is shown in Figure 6. The experimental curve 1 is obtained on a 0.85 µm thick cell with ZhK-438 at 30°C. The polarizer is parallel to the rubbing direction and the optical response to positive and negative pulses is the same, as expected. What is quite specific is a fast relaxation of the field off state (no memory effect). For modeling the same parameters were used as before (Fig. 2) with the same capacitance ($C = 35\,\mathrm{nF}$) of the aligning layers. The calculated curve 2 in Figure 6 decays slower than curve 1, however its main feature is the same (no memory). It looks like the back relaxation of FLC is accelerated by a field of opposite sign with respect to the pulse polarity. Such a field should be provided by a discharge of capacitance C through the external circuit after the end of the pulse from a function generator. This is exactly what out modeling shows, see Figure 7.

We increase the capacitance C from 27 to $1000\,\mathrm{nF/cm^2}$ (curves 1–5) and follow a change in the cell transmission (top) and voltage on the FLC layer (bottom). This increase of capacitance is equivalent to adecrease of the

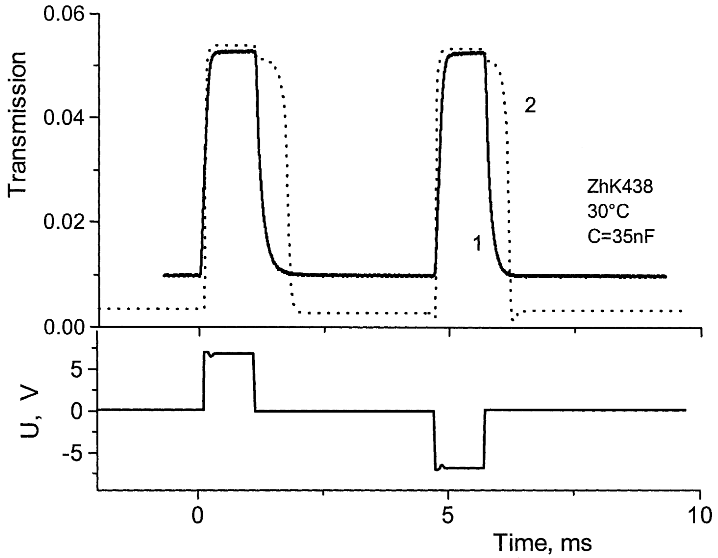

FIGURE 6 Experimental (1) and calculated (2) oscillograms of electrooptical response of 0.8 μm thick cell filled with ZhK-438 (30°C, $G = 1\,\mu S$) to polar voltage pulses shown at the bottom.

thickness of an aligning PI layer from 60 nm to 1.6 nm. Curve 1 in the bottom plot clearly indicates that, after the positive voltage pulse, an inverse (negative) voltage remains on the FLC layer for about $t_{inv} = 0.5$ ms. This inverse voltage forces the optical transmission to vanish very fast, within the same 0.5 ms (curve 1 on the top plot). With increasing C(curves $1 \rightarrow 5$) the t_{inv} time increases, the delay in the optical transmission pulse become longer, and finally, in the limit of infinitely thin aligning layers ($C \rightarrow \infty$) the voltage on the FLC layer repeats the shape of the external voltage pulse and the optical response manifests the true bistability (curves 5 on both plots). Experimentally we have checked this trend using a standard 2 μm thick cell with thin aligning layers and several *external* capacitors connected in series with the cell. The results of our calculations have completely been confirmed.

Therefore, our modeling has resulted in a better understanding of the very nature of two relevant phenomena, the V-shape switching and bistability. In fact, they are two sides of the same coin. With a large capacitance and/or high conductivity of aligning layers the inverse field is very low and true bistability is observed. On the contrary, with a small capacitance and low conductivity of aligning layers (and also with enhanced conductivity of an FLC) the V-shape electrooptical switching is observed. Note,

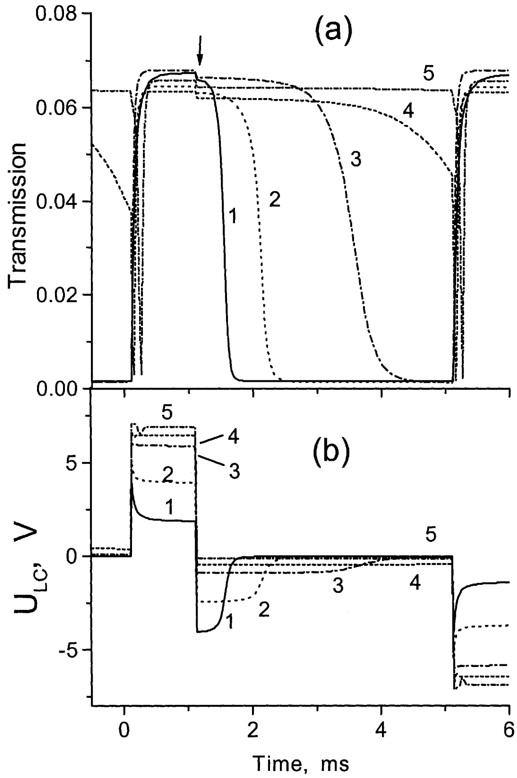

FIGURE 7 Calculated oscillograms of optical transmission (a) and voltage on the FLC layer (b) for 0.8 μm thick cell filled with ZhK-438 (30°C, $G = 1\,\mu S$). Capacitance of aligning layers is varied: $C = 27(1)$, 35(2), 100(3), 200(4) and $1000\,nF/cm^2$ (5).

that electrooptical hysteresis would still be observed, if the transmission is plotted as a function of the voltage on the FLC layer and not of the voltage on the cell as discussed earlier [11].

4. CONCLUSION

The V-shape electrooptical switching in FLC cells has been modeled and the results are in quantitative agreement with experiment. The switching threshold and hysteresis inversion frequency strongly depend on the conductivity of FLC layers. The key role of the capacitance C of FLC aligning layers has been underlined: thin layers with large C promote the well known bistability effect, thick layers, on the contrary, are necessary for V-shape switching.

REFERENCES

[1] Lagerwall, S. T. (1999). *Ferroelectric and antiferroelectric liquid crystals*, Wiley-VCH: Weinheim, 390.
[2] Inui, S., Iimura, N., Suzuki, T., Iwane, H., Miyachi, K., Takanishi, Y., & Fukuda, A. (1996). *J. Mater. Chem.*, *6*, 671.
[3] Chandani, A., Cui, Y., Seomun, S. S., Takanishi, Y., Ishikawa, K., Takezoe, H., & Fukuda, A. (1999). *Liq. Cryst.*, *26*, 151 and *26*, 167.
[4] Takezoe, H. & Takanishi, Y. (2001). In: *Chirality in Liquid Crystals*, Kitzerow, H.-S. & Bahr, C. (Eds.), Springer, 251.
[5] Rudquist, P., Lagerwall, J. P. F., Buivydas, M., Gouda, F., Lagerwall, S. T., Clark, N. A., Maclennan, J. E., Shao, R., Coleman, D. A., Bardon, S., Link, D. R., Natale, G., Glaser, M. A., Walba, D. M., Wang, M. D., & Chen, X.-H. (1999). *J. Mat. Chem.*, *9*, 1257.
[6] Clark, N. A., Coleman, D., & Maclennan, J. E. (2000). *Liq. Cryst.*, *27*, 985.
[7] opi, M., Maclennan, J. E., & Clark, N. (2002). *Phys. Rev. E.*, *65*, 021708.
[8] Panarin, Yu., Panov, V., Kalinovskaya, O. E., & Vij, J. K. (2000). *Ferroelectrics*, *246*, 35.
[9] opi, M., Maclennan, J. E., & Clark, N. (2001). *Phys. Rev. E.*, *63*, 031703.
[10] Seomun, S. S., Panov, V. P., Vij, J. K., Fukuda, A., & Oton, J. M. (2001). *Phys. Rev. E.*, *64*, 040701(R).
[11] Blinov, L. M., Pozhidaev, E. P., Podgornov, F. V., Pikin, S. A., Palto, S. P., Sinha, A., Yasuda, A., Hashimoto, S., & Haase, W. (2002). *Phys. Rev. E*, *66*, 021701.
[12] Palto, S. P. (2001). *JETP*, *92*, 552.
[13] Palto, S. P. (2003). *Kristallografia*, *48*, 145 (to be translated from Russian).
[14] Palto, S. P., Blinov, L. M., Podgornov F. V., & Haase, W. *Mol. Cryst. Liq. Cryst.* (the previous paper).