Fast (submillisecond) Switching of Nematic Liquid Crystals and Effects of Dielectric Dispersion: Theory and Experiment

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Abstract

We demonstrate that the finite rate of dielectric relaxation in liquid crystals which has been ignored previously causes profound effects in the fast dielectric reorientation of the director. The phenomenon is relevant for sub-millisecond switching of the director when the switching rate approaches the rate of dielectric relaxation through the reorientation of the molecular dipoles. A submillisecond switching (15– 400 μs for an optical retardation shift 0.3 – 2.2 μm in 10-15 μm thick cells) is demonstrated for dual frequency nematic cells with high pretilt that maximizes the dielectric torque acting on the director. We propose the theory of dielectric response in which the electric displacement depends not only on the present (as in the standard theory) but also on the past values of electric field and director. We design an experiment in which the standard "instantaneous" model and our model predict effects of opposite signs; the experimental data support the latter model.

1. Introduction

Orientational dynamics of nematic liquid crystals (NLCs) in the electric field caused by dielectric anisotropy of these materials is a fundamental physical phenomenon that is at the heart of numerous modern technologies. The director **n** that is the direction of the average molecular orientation and simultaneously the optic axis of the NLC, reorients under the action of the dielectric torque of density $\mathbf{M}(t)=\mathbf{D}(t)\times\mathbf{E}(t)$, where **E** is the electric field and **D** is the electric displacement at the moment of time *t*. In the widely accepted standard approach, the director dynamics is described assuming that the dielectric response is instantaneous, i.e., the displacement $\mathbf{D}(t)$ is determined by the electric field at the very same moment t. Such an approximation is certainly valid when the switching time of the NLC device is much longer than the time of dielectric relaxation. The characteristic relaxation time of the dipole reorientation in NLCs might be rather slow, of the order of 0.1-1 ms, which is close to the desired range for the modern nematic display modes [1-7]. As we demonstrated recently, the nematic cells can be switched very quickly, within 0.1 ms for a cell of a thickness 10-15 μm [7]. Further development in the field of fast switching NLC is hindered by the fact that the model of an instantaneous response becomes inaccurate for submillisecond response times. In this paper, we propose a general model to describe the time-dependent dielectric response of NLCs. The corresponding theories exist for isotropic fluids and solid crystals: in both cases, the dielectric properties of the medium do not change with time [8]. In the NLC, however, the situation is more complex, as the electric field causes director reorientation which in its turn changes the dielectric coupling between the field and the medium. We present an experiment that is well described by the proposed theory but which cannot be described within the classic model of an instantaneous response. The next section describes the basic experimental setup that allows one to achieve a fast switching, followed by the theory and then by verification of the theory and the experiment.

2. Fast switching: Experiment

We use the so-called dual-frequency nematic materials in anti-parallel cells with a high pretilt angle ($\alpha\approx$30-70, preferably close to 40-50 degrees) driven by a sequence of electric pulses of different frequency and amplitude. The high pretilt angle is achieved by

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an oblique deposition of SiO layers (Fig.1), although some polyimides can be used, too. High value of pretilt angle has several advantages. First, the dielectric torque of the applied field, which scales as $\sim \sin\theta\cos\theta \sim \sin 2\theta$, is maximized when the angle θ between the director and the field is about 45 degrees, so that $\sin 2\theta \approx 1$. Second, there is no threshold for director reorientation. Third, high pretilt guaranties strong restoring surface torques that facilitates reorientation from both the homeotropic and the planar states.

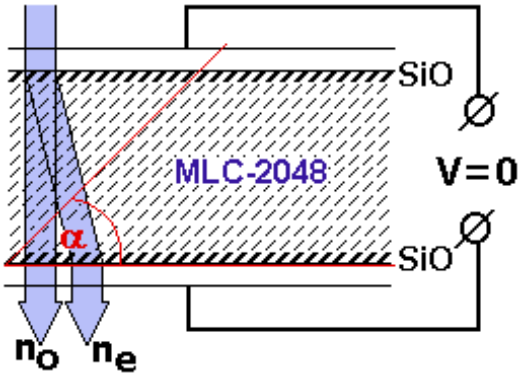
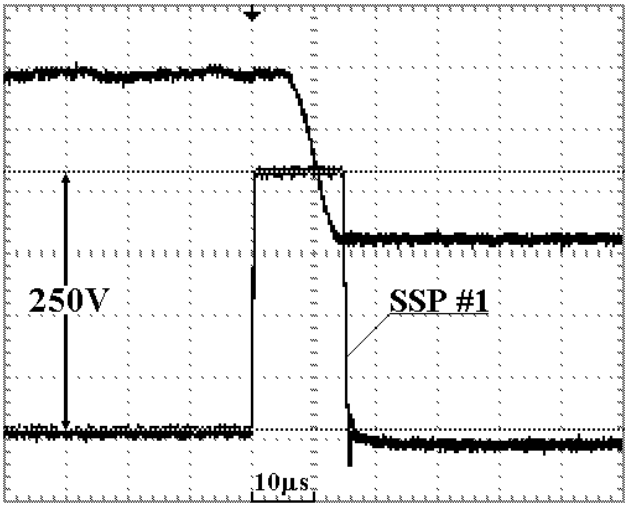


Figure 1. Initial tilted orientation of dual frequency nematic cell.

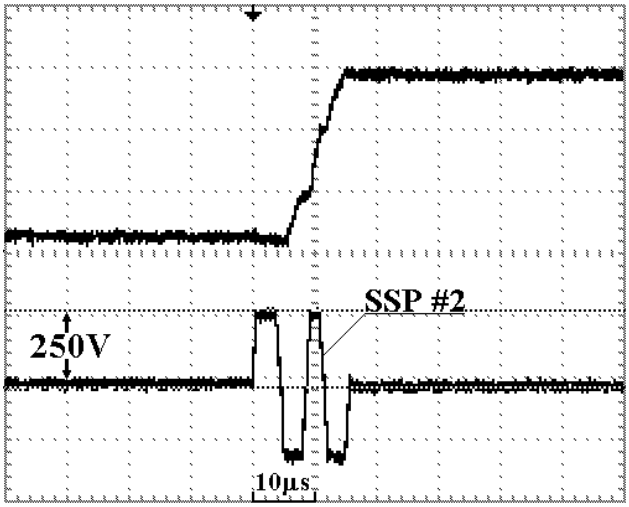
Depending on the frequency of the applied voltage, the director reorients towards homeotropic state or towards the planar state (Fig.1), because the dielectric anisotropy $\Delta\varepsilon$ of the dual-frequency nematic changes the sign at crossover frequency. For example, in the nematic material MLC-2048 that we used, Δε=3.22 at 1 kHz and Δε=-3.5 at 100 kHz at the room temperature.

The speed of switching of the cell can be determined by measuring the intensity of laser light (HeNe 632 nm) passing through the cell; the light is linearly polarized under the angle 45 degrees with respect to the "easy" axes direction in the plane of the cell. The intensity is determined as $I(\Delta\varphi) = I_0 \sin^2 \frac{\Delta\varphi}{2}$, where I_o is the intensity of incident light (we neglect small corrections due to the reflection of light at interfaces, scattering at director fluctuations, etc.) and $\Delta\varphi$ is the phase shift acquired by the light passing through the cell; in the homeotropic state, $\Delta\varphi$=0.

The dual-frequency nematic allows us to control the switching dynamics in **both** directions by the applied voltage. We start with the special short pulse (SSP) of large amplitude to speed up the switching. The low-and high-frequency SSPs are used for switching toward the homeotropic and planar states, respectively. Amplitude and duration of the SSP were adjusted to minimize the transition time to the final state, which is controlled by the following "holding" voltage of appropriate frequency and amplitude. The scheme allows us to achieve 2 μm optical retardation shift during 0.4 ms; 0.3 μm switches can be achieved much faster, approximately within 15 μs.



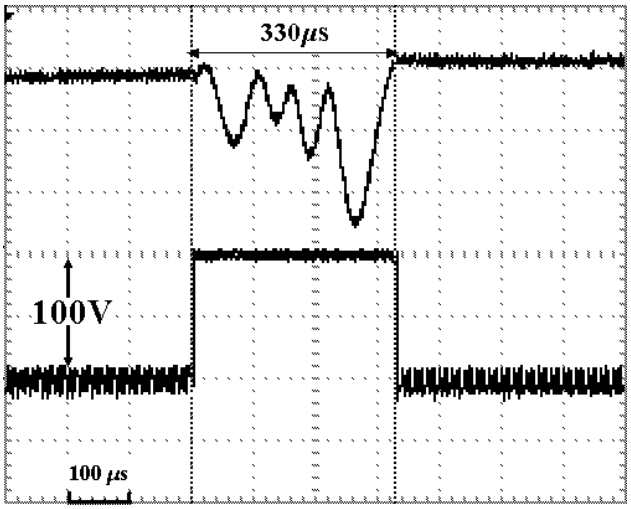
a) 10 μs/square;



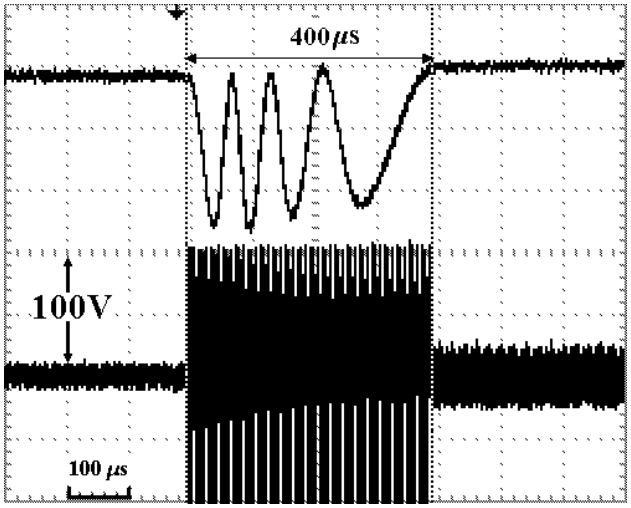
b) 10 μs/square

Figure 2. Fast switching of phase shift *ΔL ≈0.3μ*m by (a) square pulse; (b) high frequency pulse. The top trace shows the light intensity change and the bottom shows the voltage profile.

An example of the applied voltage profile with two SSPs is presented in Fig.2. The first square SSP (duration 15 μs) produces fast reorientation of phase shift $\Delta L \approx 0.3\mu$m. Fig.2b shows how the cell is switched back to the original state; we used the second SSP (duration 16 μs, frequency 125 kHz). The dual-frequency cell demonstrates 0.4 ms of the switching time for the optical retardation shift by $\Delta L \approx 2.2\mu$m, which is close to the complete reorientation between homeotropic and planar states, see Fig 3:



(a) 100 μs/square;



(b) 100 μs/square

Figure 3. Fast switching of $\Delta L \approx 2.2\ \mu$m by (a) square pulse (b) high frequency pulse. The top trace shows the light intensity change and the bottom shows the voltage profile.

3. Dielectric dispersion and dielectric torque: Theory

The superposition rule in the classical electromagnetic theory results in the following dependence:

$$\mathbf{D}(t) = \varepsilon_0 \mathbf{E}(t) + \varepsilon_0 \int_{-\infty}^{t} \boldsymbol{\alpha}(t,t')\mathbf{E}(t')du \qquad (1)$$

where $\boldsymbol{\alpha}(t,t')$ is the step response tensor describing the contribution of the electric field at the moment $t' \le t$. To analyze the dynamics, it is useful to split $\boldsymbol{\alpha}(t,t')$ into a fast $\boldsymbol{\alpha}_f(t,t')$ and a slow $\boldsymbol{\alpha}_s(t,t')$ contributions with respect to the director rotation and to the rate of electric field change, $\boldsymbol{\alpha}(t,t') = \boldsymbol{\alpha}_f(t,t') + \boldsymbol{\alpha}_s(t,t')$, so that Eq. (1) reads

$$\mathbf{D}(t) = \varepsilon_0 \boldsymbol{\varepsilon}_f(t)\mathbf{E}(t) + \varepsilon_0 \int_{-\infty}^{t} \boldsymbol{\alpha}_s(t,t')\mathbf{E}(t')dt', \qquad (2)$$

The slow part $\boldsymbol{\alpha}_s(t,t')$ is caused by reorientation of the permanent molecular dipoles in the NLC. Even when the director angular velocity is high and approaches the relaxation rate of polarization, the director-imposed slow rotation of all molecules should not affect substantially the fast individual flip-flops responsible for the dielectric relaxation. The latter assumption implies that $\boldsymbol{\alpha}_s(t,t')$ can be expressed through the step response tensor component $\alpha_{\parallel}(t-t')$ along the director, when the director is fixed:

$$\boldsymbol{\alpha}_s(t,t') = \alpha_{\parallel}(t-t')\hat{\mathbf{n}}(t) \otimes \hat{\mathbf{n}}(t'). \qquad (3)$$

Here $\otimes$ stands for the external product of two vectors, which is the tensor with the components $\left[\hat{\mathbf{n}}(t) \otimes \hat{\mathbf{n}}(t)\right]_{ij} = n_i(t)n_j(t)$. The classical Debye's theory of relaxation predicts the exponential decay of $\alpha_\nu(t-t')$ and the Lorenzian behavior for $\alpha_\nu(\omega)$ [9-11]:

$$\alpha_\nu(t-t') = \frac{\varepsilon_{l\nu} - \varepsilon_{h\nu}}{\tau_\nu} \exp\left(-\frac{t-t'}{\tau_\nu}\right) \qquad (4)$$

$$\alpha_\nu(\omega) = \frac{\varepsilon_{l\nu} - \varepsilon_{h\nu}}{1 - i\omega\tau_\nu} \qquad (5)$$

where ε_{lv} and ε_{hv} are the dielectric permittivity components at low and high frequencies, respectively; we remind that $v = \|, \perp$. In this case the resulting dielectric torque density for the uniaxial NLC is:

$$\mathbf{M}(t) = \varepsilon_0 \mathbf{n}(t) \times \mathbf{E}(t) \{ (\varepsilon_{h\|} - \varepsilon_h) \mathbf{n}(t) \cdot \mathbf{E}(t) + \frac{\varepsilon_{l\|} - \varepsilon_{h\|}}{\tau_\|} \int_{-\infty}^{t} \exp\left(-\frac{t-t'}{\tau_\|} \right) \hat{\mathbf{n}}(t') \cdot \mathbf{E}(t') dt' \}. \quad (6)$$

The dielectric memory effect is described by the integral term of Eq. (6), which is absent in the standard approach.

4. Dielectric dispersion and dielectric torque: Experiment vs. Theory

Figure 3 shows the transmitted intensity (top trace) versus the applied voltage (bottom trace) at two frequencies (100 kHz and 1 kHz) when the voltage applied to a hybrid aligned nematic cell varies slowly with the rate 2.4 V/s. For such a slow rate, the dielectric behavior can be regarded as a quasi-static dielectric response, where the standard description with an instantaneous relation between the displacement and the field is valid.

The dielectric memory effect described by the last term in Eq.(3) becomes evident when the voltage changes abruptly. The behavior of light intensity recorded for 100 kHz pulses in Fig. 5(a) is in agreement with the quasi-static behavior in Fig. 4. However, the initial response to a step-like pulse of a low frequency, Fig. 5(b), is exactly opposite to what is expected from the quasi-static model and experiment in Fig. 4. Namely, Fig. 4 suggests that the light intensity should increase towards point A when the voltage is increased at 1 kHz, while Fig. 5(b) demonstrates that the voltage pulse actually **decreases** the light intensity (towards point Y in Fig.5. (b)) at the beginning of director reorientation. This anomalous decrease is not related to the possible parasitic effects such as light scattering losses: The insert in Fig. 5(b) demonstrates that the trend is reversed when an additional π phase retarder (Soleil-Babinet compensator SB-10 purchased from Optics for Research) is inserted between the cell and the polarizer. Therefore, the reason for the different response of the director to the quasi-static, Fig.4, and abrupt, Fig.5 (b), voltage increase at 1 kHz is not related to the parasitic effects and might be caused by the dielectric memory effect, i.e., by the fact that $\mathbf{D}(t) \neq \varepsilon_0 \boldsymbol{\varepsilon} \mathbf{E}(t)$.

To verify this hypothesis, we simulated the transmitted light intensity using Eq.(6). The polar angle $\theta(z,t)$ between $\hat{\mathbf{n}}$ and the normal to the cell is described by the Erickson-Leslie equation; we neglected the backflow effects as we are interested in the very beginning of field-induced reorientation; the initial condition is $\theta(z,\ t=0) = \theta_0 = 45^0$ at $M(t<0) = 0$. We independently measured the rotation viscosity $\gamma = 0.3\ kg \cdot m^{-1} \cdot s^{-1}$ [12], and the elastic constants $K_1 = 17.7$ pN and $K_3 = 21.4$ pN, the cell thickness $d = 10\ \mu$ m; the extraordinary and ordinary indices of refraction: $n_e = 1.705$ and $n_o = 1.495$ (both at $\lambda =$ 633 nm). The experimental light intensity curves in Fig.5 are compared to the two models: the model developed in this work, Eq. (6) (dashed lines), and the standard model (dotted line) with an instantaneous relationship $\mathbf{D}(t) = \varepsilon_0 \boldsymbol{\varepsilon} \mathbf{E}(t)$. The new model agrees well with the experiment, while the standard model contradicts it. The standard model, as compared to the experiment, shows the opposite direction of intensity changes and thus the opposite direction of the director reorientation when the amplitude of 1 kHz voltage changes abruptly, Fig.5(b).

5. Conclusion

We propose a general model to quantitatively describe the orientation dynamics of dispersive liquid crystals in which the assumption of the instantaneous relationship between the electric displacement and the electric field is lifted. This time scale is typically in the sub-millisecond range which is of great interest for modern fast-switching devices. The proposed model expresses the electric displacement $\mathbf{D}(t)$ as the function of the static dielectric properties of the NLC, the present and past electric field and the present and past director. The proposed model should be applicable to dynamic reorientation of other LC phases; in the case of ferroelectric LCs, the theory should be supplemented by the consideration of spontaneous electric polarization. We demonstrated that the phenomenon of dielectric dispersion should

be taken into account in the development of fast-switching LC devices. The next step is to use the theory outlined above in the optimization of the driving voltage pulses to achieve the fastest possible response times.

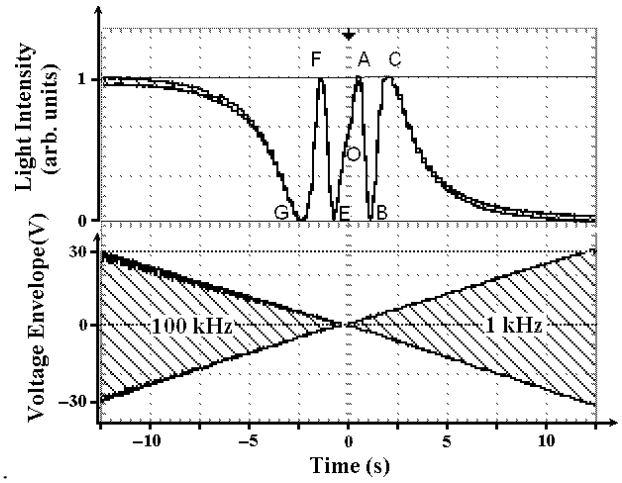


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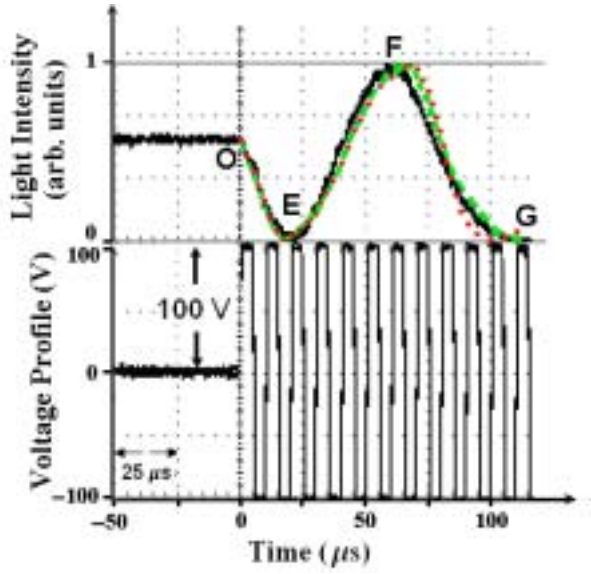
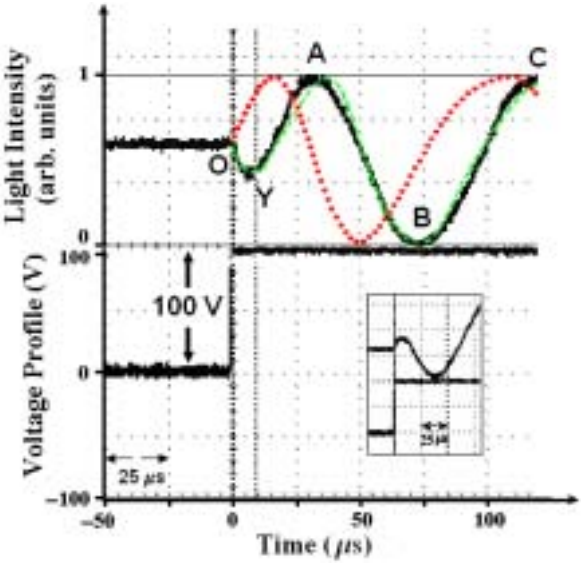
Figure 4. Transmitted light intensity modulated by the changes of optical retardation (the top curve) *vs*. slowly changing sinusoidal voltage (the bottom trace that shows the envelope of sinusoidal signal) applied at two different frequencies 100 kHz (left part) and 1 kHz (right part) to the MLC2048 cell of thickness $d = 10\ \mu m$. Point O corresponds to light transmittance at zero voltage; points A, B, etc. are the extrema of the light intensity curve where $\Delta\varphi = k\pi;\quad k$ is an integer.



(a)



(b)

Figure 5. Transmitted light intensity modulated by the changes of optical retardation for the same DFN cell as in Fig. 2, but driven by steep changes of the applied voltage at 100 kHz (a) and 1 kHz (b). The voltage profile is shown by the lower traces. The time scale is $25\mu s/sqr$. In the top parts, the solid lines are the oscilloscope's trace for the experimentally determined light transmittance, the dashed lines represent the transmitted intensity as calculated from our model (6), and the dotted lines represent the standard approach with $\mathbf{D}(t) = \varepsilon_0 \varepsilon \mathbf{E}(t)$. Point "Y" corresponds to the maximum director reorientation in the "wrong" direction. The insert is the optical transmission for the DFN cell driven by a 1 kHz pulse when a π phase retarder is inserted between the polarizer and the cell.

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7. References

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