Polarity-Dependent Dielectric Torque in Nematic Liquid Crystals

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The dielectric dispersion in the uniaxial nematic liquid crystals affects the switching dynamics of the director, as the dielectric torque is determined by not only the present values of the electric field and director but also by their past values. We demonstrate that this ''dielectric memory'' leads to an unusual contribution to the dielectric torque that is linear in the present field and thus polarity sensitive. This torque can be used to accelerate the ''switch-off'' phase of director dynamics.

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The reorientation of nematic liquid crystal (NLC) molecules by an electric field $\mathbf{E}$ is used in many electro-optical applications, most notably liquid crystal displays (LCDs) [1]. Tremendous efforts have been put into improving the performance of LCDs, especially their switching time. There are two phases in electric switching of a LCD. The fast ''active'' phase of ''switch on'' is driven by an applied voltage U with the characteristic time $\tau_{\rm on} \approx \gamma_1 d^2/(\varepsilon_0|\Delta\varepsilon|U^2)$ (here ε_0 is the electric constant, d is the cell thickness, γ_1 is the rotational viscosity, $\Delta\varepsilon = \varepsilon_\parallel - \varepsilon_\perp$ is the dielectric anisotropy, $\varepsilon_\parallel$ and $\varepsilon_\perp$ are the principal dielectric permittivities referred to the director $\hat{\mathbf{n}}$) [1]; $\tau_{\rm on}$ can be decreased by increasing U. Director reorientation in the ''switch off'' phase is a ''passive'' process driven by relaxation of elastic distortions with switch-off time $\tau_{\rm off} \approx \gamma_1 d^2/(\pi^2 K)$ [1] that depends on the NLC properties such as γ_1 and the elastic constant K, but not on the (pre)applied electric field. This consideration is based on a classic picture of a NLC as a medium with no dielectric dispersion and instant dielectric response; the dielectric torque $\mathbf{M}_d = \varepsilon_0\Delta\varepsilon(\mathbf{E}\cdot\hat{\mathbf{n}})\mathbf{E}\times\hat{\mathbf{n}}$ is quadratic in $\mathbf{E}$ and is determined by the present values of $\mathbf{E}$ and $\hat{\mathbf{n}}$ [1]. Frequency dependence of dielectric permittivity changes this picture, as $\mathbf{M}_d$ becomes dependent not only on the present $\mathbf{E}$ and $\hat{\mathbf{n}}$, but also on their past values [2,3]. This ''dielectric memory effect'' (DME) has been studied for so-called dual frequency NLCs in which $\Delta\varepsilon$ changes sign with the frequency f [2–4]. In this work, we demonstrate a very unusual consequence of DME, namely, an existence of a ''memory'' dielectric torque that is *linear* (rather than quadratic) in the present $\mathbf{E}$. The direction of the torque can be controlled by the polarity of $\mathbf{E}$, regardless of the sign of $\Delta\varepsilon$. This feature allows one to design a situation when the switch-off phase can be accelerated by a properly chosen back edge of the electric pulse and is no longer a passive process.

Theory.—Because of dielectric relaxation, electric displacement $\mathbf{D}(t)$ and the torque $\mathbf{M}_d(t) = \mathbf{D}(t)\times\mathbf{E}(t)$ depend on both the present $\mathbf{E}(t)$, and the past field $\mathbf{E}(t')$, $-\infty < t' \le t$ [2,3]. Many NLCs experience only a single relaxation process at $f < 10$ MHz that can be described by the Debye model:

$$\varepsilon_\parallel(f) = \varepsilon_{h\parallel} + \frac{\varepsilon_{l\parallel} - \varepsilon_{h\parallel}}{1 - i2\pi f\tau}, \qquad \varepsilon_\perp(f) = \varepsilon_\perp = \text{const}, \tag{1}$$

where ''l'' and ''h'' refer to the low and high f, τ is the dielectric relaxation time. We focus on two Debye type materials with different (but f independent) signs of $\Delta\varepsilon$ with dielectric relaxation in the kHz region convenient for experimental studies (although the consideration is applicable for other parts of the spectrum). The negative $\Delta\varepsilon < 0$ NLC was obtained by mixing 60.6 wt % MLC-7026-100 (EM Industries) and 39.4 wt % 2F-3333 (Rolic Technologies); the positive $\Delta\varepsilon > 0$ NLC was a mixture of 20.0 wt % of pentylcyanobiphenyl (5CB, EM Industries) and 80.0 wt % 2F-3333, Fig. 1. The dielectric permittivities were measured using Schlumberger 1260 impedance/gain-phase analyzer.

The director dynamics is determined by the balance of dielectric $\mathbf{M}_d$, viscous $\mathbf{M}_v$, and elastic $\mathbf{M}_e$ torques through the Ericksen-Leslie equation $\mathbf{M}_d + \mathbf{M}_v + \mathbf{M}_e = \mathbf{0}$. For a flat cell with plates along the x-y plane, $\mathbf{E}(t) = E_z(t)\hat{\mathbf{z}}$, $\hat{\mathbf{n}}(t)$ in the x-z plane depending only on z, the only nonzero components of torques are along the y axis. The dielectric torque is [2]:

$$M_d(t) = \varepsilon_0 E(t)\sin\theta(t)\Big[\Delta\varepsilon_h E(t)\cos\theta(t) + \frac{\varepsilon_{l\parallel} - \varepsilon_{h\parallel}}{\tau} \times \int_{-\infty}^{t} \exp\Big(-\frac{t-t'}{\tau}\Big)E(t')\cos\theta(t')\,dt'\Big], \tag{2}$$

where $\theta(t)$ is the angle between $\hat{\mathbf{n}}(t)$ and $\mathbf{E}(t)$. For $\varepsilon_{l\parallel} = \varepsilon_{h\parallel} = \varepsilon_\parallel$, Eq. (2) recovers the classic ''instantaneous'' dielectric response theory.

Consider the torque balance in response to a voltage changes over a short time interval between $t = 0$ and $t \sim \tau$. This interval is short enough to assume the changes of $\theta(t)$ small. This allows us to approximate as time independent the following three quantities: (a) M_d with $\theta(z, t) \approx \theta(z, t = 0) = \theta_0(z)$ [provided $\theta_0(z) \neq 0, \pi/2$], (b) the elastic torque $M_e(z, t) = M_e(z)$, and (c) the spatial nonuniformity of the electric field, $E_z(z, t) = g(z)U(t)$, where $g(z)$ is a proportionality factor. We neglect the backflow effect, thus $M_v(t) = \gamma_1 d\theta(t)/dt$. Under these assumptions,

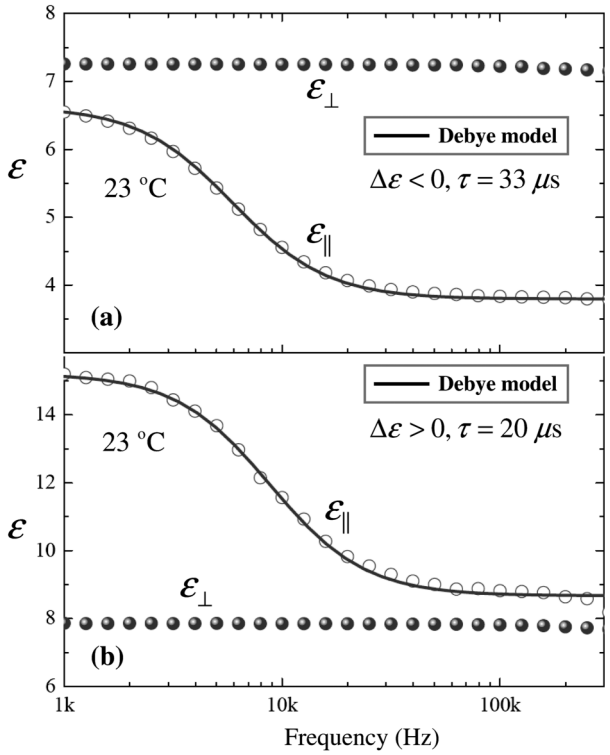

FIG. 1. Dielectric dispersion of two NLCs, with $\Delta\varepsilon < 0$ (a) and $\Delta\varepsilon > 0$ (b). The data for $\varepsilon_{\parallel}$ are fitted by Eq. (1) with $\varepsilon_{l\parallel} = 6.60$, $\varepsilon_{h\parallel} = 3.91$, $\varepsilon_{\perp} = 7.31$, and $\tau = 33$ μs (a); $\varepsilon_{l\parallel} = 15.21$, $\varepsilon_{h\parallel} = 8.67$, $\varepsilon_{\perp} = 7.88$, and $\tau = 20$ μs (b).

the solution of Ericksen-Leslie equation $\gamma_1 d\theta(t)/dt = -M_d - M_e$ that describes the time evolution of the system, is $\theta(t) = \theta_0(z) - \Theta(z)Q + M_e(z)t$, where $\Theta(z) = \varepsilon_0(\varepsilon_{l\parallel} - \varepsilon_{h\parallel})\tau U_0^2 g^2(z)\sin 2\theta_0(z)/\gamma_1$ and Q is the normalized integrated dielectric torque:

$$Q = \tau^{-1}\int_0^t \left\{\xi u^2(t') + u(t')\left\{u_{\rm mem}\exp(-t'/\tau) + \tau^{-1}\int_0^i \exp[-(t'-t'')/\tau]u(t'')dt''\right\}\right\}dt'. \quad (3)$$

Here $\xi = (\varepsilon_{h\parallel} - \varepsilon_{\perp})/(\varepsilon_{l\parallel} - \varepsilon_{h\parallel})$, $u(t) = U(t)/U_0$ is the normalized voltage with $U_0 = U(t \to 0^-)$ being the voltage that acts just before the switch-off moment $t = 0$, $u_{\rm mem} = \tau^{-1}\int_{-\infty}^0 \exp(t'/\tau)u(t')dt'$ is the memory term caused by the "past" field.

To optimize Q, we apply a direct variational method. Integrating Eq. (3) with a decaying exponential probe function $u(t) = a\exp(-\Gamma t/\tau)$, where a and $\Gamma > 0$ are two variational parameters, one obtains

$$Q = \frac{a[2u_{\rm mem}\Gamma + a(1+\xi+\xi\Gamma)]}{2\Gamma(1+\Gamma)} - \frac{a^2(1+\xi-\xi\Gamma)}{2\Gamma(\Gamma-1)}e^{-2\Gamma t/\tau} + \frac{a[u_{\rm mem}(\Gamma-1)+a]}{\Gamma^2-1}e^{-(1+\Gamma)t/\tau}. \quad (4)$$

The last expression will be used to fit the data below; it is instructive to qualitatively discuss its basic feature. The first term dominates for large $t > 5\tau$ and determines the saturated value of Q. Its extremum $Q_e = u_{\rm mem}^2\{[(1+\xi)\xi]^{1/2} - \xi - 1/2\}$ is reached for $\Gamma_e = (1+\xi^{-1})^{1/2}$ and $a_e = u_{\rm mem}(1-\Gamma_e)$. It is easy to see that $Q_e < 0$ for the positive NLCs in which $\xi > 0$, while $Q_e > 0$ for the negative NLCs in which $\xi < -1$. The Q_e is opposite in sign to the dielectric torque in the switch-on phase, thus it can accelerate the director relaxation in the switch-off phase.

The theory can be qualitatively explained as follows. Consider first a NLC with $\Delta\varepsilon > 0$ in a planar cell. A positive dc field $E_z > 0$ reorients $\hat{\mathbf{n}}$ towards the z axis. E_z also induces a dipole moment density $\mathbf{p}$ with the components $p_{\perp} = \varepsilon_{\perp}E_z\sin\theta$ and $p_{\parallel} = \varepsilon_{h\parallel}E_z\cos\theta + p_{\rm mem}$, perpendicular and parallel to $\hat{\mathbf{n}}$, respectively. Here $p_{\rm mem} \propto u_{\rm mem}$ is the memory contribution that saturates to the value $p_{\rm mem} = (\varepsilon_{l\parallel} - \varepsilon_{h\parallel})E_z\cos\theta$ after the dc field E_z acted for a sufficiently long time $>\tau(u_{\rm mem} \to 1)$ (note that $p_{\rm mem} > 0$ and $E_z > 0$ are of the same sign). When the field is switched off at $t = 0$, $p_{\rm mem} > 0$ does not disappear instantaneously, but decays with a characteristic time τ. If within the interval $0 < t \le \tau$, one applies a new electric pulse of the opposite polarity $E_z < 0$, then this field would interact with the decaying $p_{\rm mem} > 0$ to assist the reorientation towards the planar state $\theta \to \pi/2$.

In the homeotropic cell with a negative NLC, the field $E_z > 0$ at $t < 0$ also induces $p_{\rm mem} > 0$ (of the same polarity). If within the interval $0 \le t \le \tau$ one applies a new voltage pulse of *the same* polarity, $E_z > 0$, then this field will couple to $p_{\rm mem} > 0$ to assist the director reorientation into the homeotropic state $\theta \to 0$.

If the NLC were not dispersive, any field-induced polarization would relax instantaneously, $p_{\rm mem} = 0$, $\varepsilon_{l\parallel} = \varepsilon_{h\parallel}$, and $\tau = 0$, and the effect would not be observed. Therefore, the classic theory with an instantaneous dielectric response cannot predict the phenomenon we describe. Below we present experimental verifications of the effect.

Experiment.—We used homeotropic and planar (with a small pretilt of $\sim 1°$) cells (EHC Ltd.) composed of glass substrates with indium tin oxide electrodes of area 10×10 mm^2; $d = 14.4$ μm for the homeotropic cell and $d = 20.6$ μm for the planar cell. The field-induced director dynamics was monitored by measuring the He-Ne laser ($\lambda = 633$ nm) light transmission $I(t) = A\sin^2(\Phi/2)$ through the cells placed between two polarizers [1]. The phase retardation Φ depends on θ; for small variations of θ, the retardation change is linear in Q, $\Phi - \Phi_0 \approx \rho Q$, where

$$\Phi_0 = \frac{2\pi n_o}{\lambda}\int_0^d \left\{\frac{n_e}{\tilde{n}(z)} - 1\right\}dz, \qquad \rho = \frac{\pi n_o n_e(n_e^2 - n_o^2)}{\lambda}\int_0^d \frac{\Theta(z)\sin 2\theta_0(z)}{\tilde{n}(z)^3}dz, \quad (5)$$

and $\tilde{n}(z) = [n_e^2\sin^2\theta_0(z) + n_o^2\cos^2\theta_0(z)]^{1/2}$. The coeffi-

cient A is close to the intensity I_0 of the impinging light for the planar cell when the rubbing direction is at 45° with respect to the polarizers [1]. In the homeotropic cell, $A \approx I_0/2$ because the applied field creates random azimuthal orientation of $\hat{\mathbf{n}}$ with numerous umbilics, 10–100 within the area probed by the beam. These defects randomize the director field in the plane of the cell thus assuring reproducibility of the experiment. The umbilics relax much slower (seconds and minutes [5]) than the duration of our experiments (<0.5 ms). We used TIA-500S-TS photodetector (Terahertz Technologies) and Tektronix TDS 210 oscilloscope to measure $I(t)$. The driving pulses were produced by WFG500 waveform generator (FLC Electronics); the maximum rate was 240 V/μs. To test the switch-off dynamics, we used two different profiles for the pulse's back edge: (i) an instantaneous back edge (in practice ~1 μs in duration because of the finite voltage change rate); (e) an exponentially decaying back edge $u(t) = a\exp(-\Gamma t/\tau)$.

To drive the homeotropic cell with a negative NLC, we first apply a square 100 V dc pulse of duration 225 μs, much longer than $\tau = 33$ μs, Fig. 1(a), to produce the saturated memory dipole moment ($u_{\text{mem}} \rightarrow 1$). This pulse is switched off by an instantaneous back edge (i) or by three different exponential edges with $\Gamma = 0.45$ and (e1) positive polarity, $a = 0.5$; (e2) $a = 0.87$; (e3) negative polarity, $a = -0.5$; Fig. 2. The optical response is different in all four cases. In the case (i), $\hat{\mathbf{n}}$ reorients slowly toward the homeotropic state $\theta \rightarrow 0$, as evidenced by the decrease in $I(t)$ in the Fig. 2 inset. The pulse (e1) produces much faster reorientation (r-e1), despite the fact that U decreases less abruptly as in case (i). The shape of the pulse (e1) is close to the optimum, as any departure from the preselected $a = 0.5$ and $\Gamma = 0.45$ causes a slower or even a nonmonotonous response, as in (e2) case. The linear $\mathbf{E}$ dependence of the memory torque is well illustrated by the response to pulses (e1) and (e3) that differ only in polarity: (e1) drives $\hat{\mathbf{n}}$ toward $\theta = 0$ while pulse (e3) continues to drive $\hat{\mathbf{n}}$ toward $\theta = \pi/2$. After a sufficiently long time, the NLC relaxes to the same homeotropic state with $I = 0$ for all pulses, Fig. 2.

The different scenarios can be fitted by the model above, using the approximation $\Phi - \Phi_0 \approx \rho Q(t)$ and Eq. (5). The only fitting parameter is ρ, as $Q(t)$ is determined by the experimental values of a and Γ, Eq. (4). We first fit the response curve (r-e1) using $a = 0.5$, $\tau = 33$ μs, and $\Gamma = 0.45$ and find $\rho = 2.33$. With this ρ, and with the experimental τ, a, and Γ, the model (4) reproduces response curves (r-e2) and (r-e3) very well with no fitting parameters, Fig. 2.

For the positive NLC in a planar cell, we used a 100 V dc pulse of duration 310 μs, Fig. 3(a). The back edge was

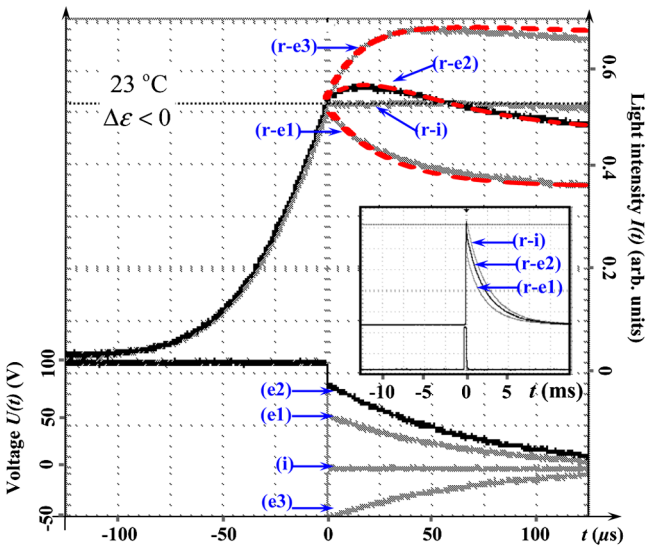

FIG. 2 (color online). Electro-optic response $I(t)$ of a $\Delta\varepsilon < 0$ NLC in a homeotropic cell driven by dc pulses with an instantaneous (i) and exponentially decaying back edges (e1–e3); "r" stands for "response." The dashed lines show $I(t)$ simulated using Eqs. (4) and (5). The inset shows $I(t)$ over a large time scale.

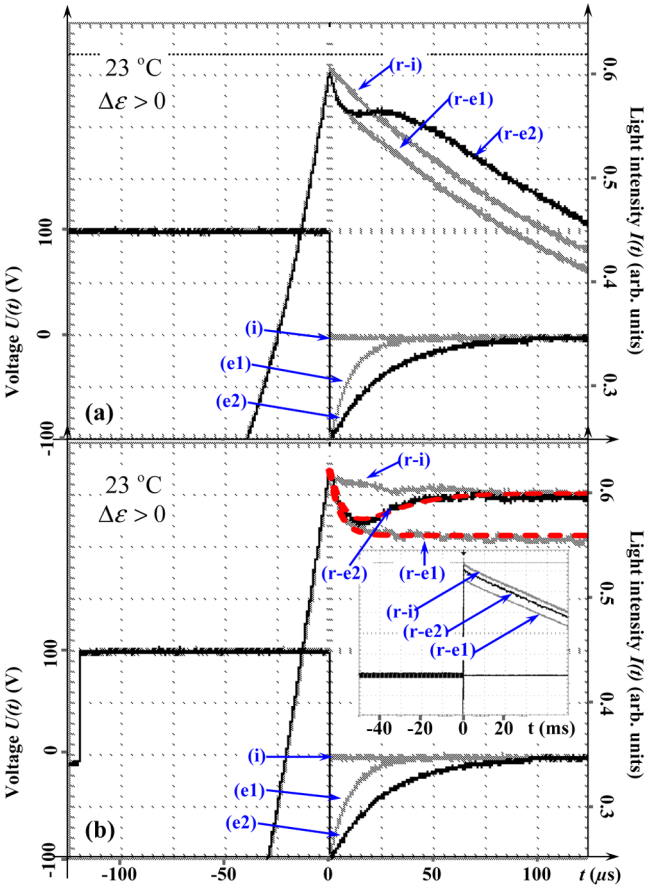

FIG. 3 (color online). Electro-optic response $I(t)$ of a $\Delta\varepsilon > 0$ NLC in a planar cell driven by dc pulses with an instantaneous (i) and exponentially decaying back edges (e1, e2). (a) The cell is driven by a dc pulse of duration 310 μs; (b) the cell is driven by a 5 V, 1 kHz ac pulse of duration 180 ms, followed by a dc pulse of duration 120 μs; the inset shows a larger time scale. The dashed lines show $I(t)$ simulated using Eqs. (4) and (5).

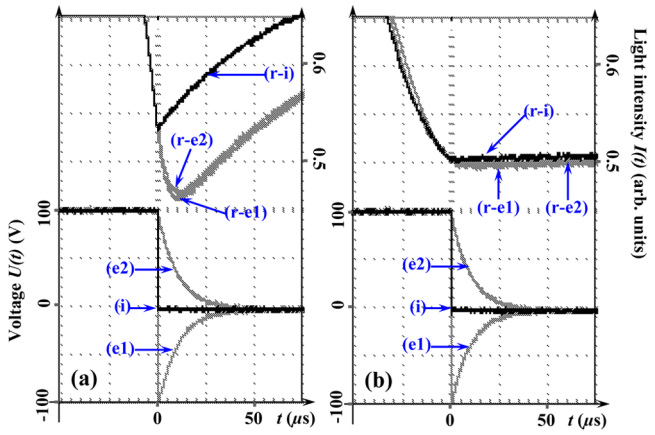

FIG. 4 (color online). Electro-optic response $I(t)$ of 5CB in a planar cell driven by dc pulses with an instantaneous (i) and exponentially decaying back edges (e1, e2) of opposite polarity. (a) The cell is driven by a dc pulse of duration 121 μs; (b) the cell is driven by a 5 V, 1 kHz ac pulse of duration 180 ms (not shown), followed by a dc pulse of duration 80 μs.

either (i) instantaneous or (e) exponential $u(t) = a\exp(-\Gamma t/\tau)$, with $a = -1$, $\tau = 20$ μs, and two different rates, $\Gamma = 2$ (e1) and (e2). A properly timed exponential "tail" (e1) produces the fastest optical response (r-e1). However, all response curves in Fig. 3(a) share one common feature that prevents a direct comparison with the theory: a universal (independent of the details of the back edge) decay, with a characteristic time 0.5 ms that is much shorter than the elastic relaxation time, $\tau_{\text{off}} \approx 0.4$ s for a typical $\gamma_1 \sim 0.1\ \text{kg m}^{-1}\,\text{s}^{-1}$ [1], but is close to the characteristic times of the backflow effect [6], i.e., coupling of the director reorientation and mass flow [1]. The other possible reason, ionic currents, does not seem plausible, because the 0.5 ms universal decay behavior does not change when we replace the dc pulse with two subsequent pulses of opposite polarities and half duration.

Although we do not know the precise mechanisms behind the universal decay and can only tentatively attribute it to the backflow, we have found empirically that this decay can be reduced while preserving the value of phase retardation at which the switch-off stage starts. This reduction is achieved by modifying the "switch-on" pulse, namely, by applying first a long (180 ms) low amplitude (5 V) ac pulse followed by a shorter dc pulse (120 μs instead of 310 μs, still longer than $\tau = 20$ μs), Fig. 3(b). Backflow dynamics depends on the combination of NLC viscosities, $B = \alpha_3\sin^2\theta - \alpha_2\cos^2\theta$, where $\alpha_3/\alpha_2 \sim 10^{-2}$ for a typical LC [7]. The ac pulse pretilts $\hat{\mathbf{n}}$ from the planar orientation $\theta \approx \pi/2$, thus stabilizing the sign of B during the action of the dc pulse. In addition, this pretilt reduces the needed angle of director reorientation during the dc pulse; the latter thus can be shorter than in the experiment in Fig. 3(a), but still long enough to produce p_{mem}. The empirically found regime in Fig. 3(b) allows us to try to describe the experimental data with the model derived in the backflow-free approximation, Eq. (4). The difference in response to pulses (i), (e1), and (e2) in Fig. 3(b) is very pronounced: pulse (e1) produces a faster reorientation than (i), while pulse (e2) results in a nonmonotonous behavior. Moreover, response curves (r-e1) and (r-e2) in Fig. 3(b) are well fitted with Eq. (4) as explained above (in the backflow-free approximation), using the experimental a, $\tau = 20$ μs, and Γ and the single fitting parameter $\rho = 0.496$.

We performed control experiments with a planar cell filled with 5CB, in which the relaxation time of $\varepsilon_{\parallel}$ is $\tau \approx 50$ ns [8]. Such a short τ should not cause a DME if the typical time of voltage changes is 1 μs. Indeed, as shown in Fig. 4, two (e) pulses with the same amplitude $|a| = 1$ and duration $\tau/\Gamma = 10$ μs, but of *opposite* polarity, produce *the same* positive torque. This feature is observed regardless of whether the cell is addressed by a dc pulse only or is additionally preaddressed with an ac pulse. The behavior is consistent with the nondispersive character of 5CB in the kHz range and with the classic model of dielectric torque quadratic in $\mathbf{E}$; it is clearly different from the behavior of dispersive NLCs that are sensitive to the polarity of the driving pulses.

Conclusion.—We demonstrate a polarity-sensitive dielectric response in NLCs with dielectric dispersion. The effect is caused by a memory term in the dielectric torque $\mathbf{M}_d(t)$ that is linear in the present field $\mathbf{E}(t)$, in contrast to a regular term quadratic in $\mathbf{E}(t)$. The polarity-dependent dielectric effect is of practical importance, as it can be used to accelerate the switch-off phase of electro-optic devices and it is also of basic interest, not only for NLCs, but generally for all soft materials with anisotropic and dispersive dielectric properties.

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