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Electronic Supplementary Information (ESI)

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Derivation of Equations 2 and 3 from the main text

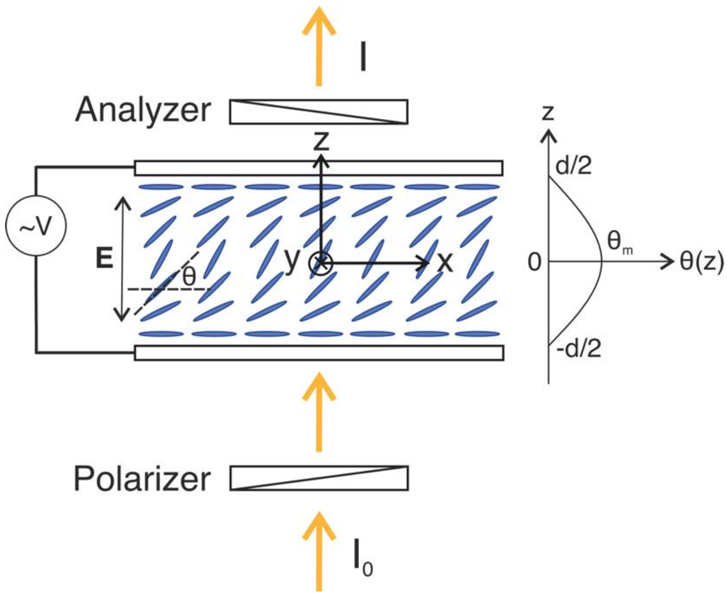

Fig. 1. Schematic of a cell placed between crossed polarizers in the presence of the electric field.

The schematic of a standard sandwich cell with planar boundary conditions is shown in Fig. 1. To find equations for the $I(t)$ curves, one needs first to describe the relaxation of the director field, characterised by the deflection angle $\theta(z,t)$ (see Fig. 1), (i) after the application and (ii) after removal of the electric field. Let us first consider the case of the electric field application.

If one ignores the director inertia and back-flow effects and considers the limit of small director field deformations, then the equation of the torque balance takes the following form [1,2]:

$$M = M_{electric} + M_{elestic} + M_{viscous} =$$

$$= -\varepsilon_0 \Delta\varepsilon E^2 \left(\theta - \frac{1}{2}\theta^3\right) - K_{11}\frac{\partial^2\theta(z)}{\partial z^2} + \gamma_1 \frac{\partial\theta}{\partial t} = 0, \quad (1)$$

where $M_{electric}$, $M_{elastic}$, and $M_{viscous}$ are the torques exhibited on the director by the electric, elastic, and viscous forces, respectively, ε_0 is the vacuum permittivity, $\Delta\varepsilon$ is the dielectric anisotropy, E is the applied electric field, K_{11} is the splay elastic constant, γ_1 is the rotational viscosity.

The solution of this equation can be approximated as $\theta = \theta_m(t)\cos\frac{\pi z}{d}$ where θ_m is the deflection angle in the middle of the cell, and d is the cell gap. After substituting this expression into the torque balance equation and solving it, one obtains:

$$\theta_m(t) = \sqrt{\frac{{\theta_{\mathrm{on}}}^2}{1 + \left[\frac{{\theta_{on}}^2}{{\theta_{off}}^2} - 1\right] e^{-\frac{2t}{\tau_r}}}} \tag{2}$$

where θ_{off} is the deflection angle in the centre of the cell before applying the electric field (which is close to zero), θ_{on} is its equilibrium value after the field application, and the rise time, τ_r, is equal to:

$$\tau_r = \frac{\gamma_1}{\varepsilon_0 \Delta\varepsilon E^2 - \frac{\pi^2}{d^2} K_{11}}. \tag{3}$$

$\theta(t, z)$ can now be found by substituting the derived $\theta_m(t)$ into $\theta = \theta_m(t)\cos\frac{\pi z}{d}$.

After the removal of the electric field, only the elastic and viscous torques are left. Under the same assumptions as for the previous case, the torque balance can be written as:

$$M = M_{elestic} + M_{viscous} = -K_{11}\frac{\partial^2\theta(z)}{\partial z^2} + \gamma_1\frac{\partial\theta}{\partial t} = 0. \tag{4}$$

The solution of this equation can be searched for in the form of $\theta = \theta_m(t)\cos\frac{\pi z}{d}$ where:

$$\theta_m(t) = \theta_{on} e^{-\frac{t}{\tau_d}}, \tag{5}$$

and after substituting this into the torque balance equation, one gets the expression for the decay time:

$$\tau_d = \frac{\gamma_1 d^2}{K_{11}\pi^2}, \tag{6}$$

and $\theta(t, z)$ becomes fully described.

The intensity of light passing through the system I can be describe as [1–3]:

$$I \sim \left[\sin\left(\frac{\Delta\varphi}{2}\right)\right]^2, \qquad (7)$$

where $\Delta\varphi = \frac{2\pi}{\lambda} n_e d - \frac{2\pi}{\lambda} n_o d$ is the phase difference between the extraordinary and ordinary waves, n_e and n_o are the corresponding refractive indices, and λ is the wavelength of light. $\Delta\varphi$ is related to the deflection angle in the middle of the cell θ_m. Let us find this relation.

During a relaxation process, the director field is different from the homogeneous configuration, and therefore, the extraordinary wave experiences different values of the refractive index at different z-coordinates. The total phase shift for the extraordinary wave can be calculated by considering the liquid crystal medium as the stack of birefringent layers with a fixed optic axis and thickness dz:

$$\varphi_e = \frac{2\pi}{\lambda} \int_{-\frac{d}{2}}^{\frac{d}{2}} n_e(z) dz \,. \qquad (8)$$

From the geometrical consideration of the dielectric ellipsoid for a birefringent layer at a given z-coordinate follows the equation for $n_e(z)$ [1,2]:

$$n_e(z) = \frac{n_\perp n_\parallel}{\sqrt{n_\parallel{}^2 (\sin\theta(z))^2 + n_\perp{}^2 (\cos\theta(z))^2}} = \frac{n_\parallel}{\sqrt{1 + \left(\frac{n_\parallel{}^2 - n_\perp{}^2}{n_\perp{}^2}\right)(\sin\theta(z))^2}} \qquad (9)$$

where $n_\parallel$ and $n_\perp$ are the refractive indices experienced by an electro-magnetic wave with the polarization parallel and orthogonal to the director, respectively.

In the limit of small θ values, this equation can be simplified to:

$$n_e(z) = n_\parallel \left(1 - \frac{1}{2}\left(\frac{n_\parallel{}^2 - n_\perp{}^2}{n_\perp{}^2}\right)\theta(z)^2\right). \qquad (10)$$

Approximating $\theta(z)$ by $\theta(z) = \theta_m \cos\frac{\pi z}{d}$ and substituting $n_e(z)$ into the equation for φ_e, one obtains:

$$\varphi_e = \frac{2\pi}{\lambda}\left(n_\parallel d - \frac{1}{2}\left(\frac{n_\parallel{}^2 - n_\perp{}^2}{n_\perp{}^2}\right) n_\parallel \theta_m{}^2 \frac{d}{2}\right) \qquad (11)$$

which under the assumption of $n_\perp \approx n_\parallel$ (which is usually justified), can be rewritten as:

$$\varphi_e = \frac{2\pi d}{\lambda}\left(n_{\parallel} - \frac{1}{2}(n_{\parallel} - n_{\perp})\theta_m{}^2\right). \tag{12}$$

Now we can write the expression for the total phase difference:

$$\begin{aligned}\Delta\varphi = \varphi_e - \varphi_o = \frac{2\pi d}{\lambda}\left(n_{\parallel} - \frac{1}{2}(n_{\parallel} - n_{\perp})\theta_m{}^2\right) - \frac{2\pi d}{\lambda} n_{\perp} = \\ = \frac{2\pi d}{\lambda}(n_{\parallel} - n_{\perp}) - \frac{\pi d}{\lambda}(n_{\parallel} - n_{\perp})\theta_m{}^2,\end{aligned} \tag{13}$$

or:

$$\Delta\varphi = \Delta\varphi_{off} - \frac{\pi d}{\lambda}(n_{\parallel} - n_{\perp})\theta_m{}^2 \tag{14}$$

where $\Delta\varphi_{off} = \frac{2\pi d}{\lambda}(n_{\parallel} - n_{\perp})$ is the phase difference in the absence of the field.

Substituting Equation 2 and Equation 5 found earlier for $\theta_m(t)$ into the expression for $\Delta\varphi$ and then substituting the result into Equation 7 for the light intensity, it is:

$$\begin{aligned}I \sim \left[\sin\left(\frac{\Delta\varphi_{off}}{2} - \frac{1}{2}\frac{\pi d}{\lambda}(n_{\parallel} - n_{\perp})\frac{\theta_{on}{}^2}{1 + \left[\frac{\theta_{\text{on}}{}^2}{\theta_{\text{off}}{}^2} - 1\right]e^{-\frac{2t}{\tau_r}}}\right)\right]^2 = \\ = \left[\sin\left(\frac{\Delta\varphi_{off}}{2} - \frac{(\Delta\varphi_{off} - \Delta\varphi_{on})}{2}\frac{1}{1 + \left[\frac{\theta_{\text{on}}{}^2}{\theta_{\text{off}}{}^2} - 1\right]e^{-\frac{2t}{\tau_r}}}\right)\right]^2\end{aligned} \tag{15}$$

for the intensity relaxation upon the electric field application, and

$$I \sim \left[\sin\left(\frac{\Delta\varphi_{off}}{2} - \frac{1}{2}\frac{\pi d}{\lambda}(n_{\parallel} - n_{\perp})\theta_{on}{}^2 e^{-\frac{2t}{\tau_d}}\right)\right]^2 = \left[\sin\left(\frac{\Delta\varphi_{off}}{2} - \frac{(\Delta\varphi_{off} - \Delta\varphi_{on})}{2}e^{-\frac{2t}{\tau_d}}\right)\right]^2 \tag{16}$$

for the intensity relaxation upon the electric field removal where $\Delta\varphi_{on} = \Delta\varphi_{off} - \frac{\pi d}{\lambda}(n_{\parallel} - n_{\perp})\theta_{on}{}^2$ is the equilibrium phase difference in the presence of the field. By fitting the measured $I(t)$ curves with the derived expressions, the response times could be found. Fitting was applied to the beginning of the relaxation curve in the case of the electric filed application and to the final part of the curve in the case of the electric filed removal since the small angle approximation used to derive the formulas is valid only for those stages of the relaxation.

References

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