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Time-resolved switching analysis of a ferroelectric liquid crystal by Snapshot Mueller Matrix Polarimetry

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An experimental snapshot Mueller Matrix polarimeter based on wavelength polarization coding is used to get a time-resolved description of electric field-induced fast transition within a ferroelectric liquid crystal cell. The parameters extracted from experimental Mueller matrices are linked to the molecule director distribution to further determine the average trajectory and the collective behavior of these molecules while they switch over to another state. © 2009 Optical Society of America.

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Mueller matrix (MM) polarimetry is usually employed to characterize samples that show depolarization, birefringence and dichroism. For a more complete characterization, MMs are coupled with variables such as wavelength, space coordinates, wave vector. To our knowledge,

little has been done with time as variable, though a temporal monitoring of MMs could permit the study of fast polarization dynamics and widen the scope of MM polarimetry. Such measurements require a device allowing the acquisition of a full MM in a very short time.

The principle of the Snapshot Mueller Matrix Polarimeter (SMMP) developed by our team and based on wavelength polarization coding [1,2] is that the polarization states are encoded in the spectral domain through use of a broadband source and high-order retarders [3]. On condition to use a well-suited retarder-thickness configuration, the full MM of a sample is available in a single spectrum, $I(\lambda)$, measured with a dispersive detection system (spectrometer and CCD camera). As the acquisition time only depends on the aperture time of the CCD camera, it can be very short (about $1\,\mu s$, here). This Letter is aimed at demonstrating the potentiality of the SMMP to give a time-resolved description of electric field-induced fast switching in a ferroelectric liquid crystal (FLC) cell. To our knowledge, only two teams have performed time-resolved MM polarimetry on liquid crystals [4,5]. But, the polarimeters in use were sequential, so the MMs were reconstructed from various input and output polarization states and multiple synchronized acquisitions. The SMMP can acquire a full MM in a single shot.

The SMMP described in Fig. 1 is composed of a broadband source (SLD from B&W Tek, Inc) around $\lambda_0 = 830\,nm$ with $\Delta\lambda = 15\,nm$, a linear polarizer oriented at 0°, two calcite retarders ($\Delta n = 0.166$) of thickness $e = 2.08\,mm$ respectively oriented at 45° and 0°, two calcite retarders of thickness $5e = 10.4\,mm$ respectively oriented at 0° and 45°, a linear polarizer oriented at 90°, a diffraction grating (1200 grooves/mm covering 10 nm) and a CCD camera (512 x 512 pixels). The signal $I(\lambda)$ is periodic and composed of several frequencies. With this retarder-thickness configuration ($e,e,5e,5e$), 13 frequencies are generated on the analysis

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window. The coefficients of a MM (m_{ij}) are retrieved through application of a Fourier transform to $I(\lambda)$ since they are linked to the magnitudes of the Fourier peaks through relationships that only depend on the retarder-thicknesses configuration [1]. By application of the calibration procedures described in [2], the accuracy on the m_{ij} coefficients (normalized by m_{00}) is below 0.03 for measurements of well-known media (polarizer, wave-plate).

The chiral liquid crystal (LC) material under study is Felix 015/100, which has a birefringence around 0.16 in the visible/NIR. Two glass plates with transparent ITO electrodes and with a thin film of rubbed polyimide, produced by spin coating, are used to fabricate the cell. Its gap, obtained by spraying of spacers, is about 1.6 µm. The cell is then filled by capillary suction and the LC is confined between both plates in planar orientation which means that the LC molecules are parallel to the substrate. At room temperature (25°C), this LC is in the smectic C* phase, where the molecular layers are perpendicular to the rubbing direction. The average orientation of molecule is specified by the unit vector $\vec{n}$, called the director (Fig. 2). The director is tilted by an angle θ with respect to the layer normal and in SC* bulk material, it rotates, forming a helical structure with the axis perpendicular to the layers. However, as the LC thickness is far below the helix pitch, and due to the planar anchoring conditions, the helical structure is suppressed so LC exhibits ferroelectricity properties. The spontaneous polarization, $\vec{P_S}$, lies in the smectic layers and is perpendicular to the director. This structure is called SSFLC (Surface-Stabilized Ferroelectric Liquid Crystal) [6]. Application of an alternative electric field, $\vec{E}$, to this cell (Fig. 2) makes $\vec{P_S}$ aligns with $\vec{E}$ and the molecules move between two stable states ("up" and "down"). The experiments were made with the FLC cell set perpendicular to the incident light.

The parameters issued from experimental MMs *(M)* were extracted by the Lu and Chipman decomposition [7], which consists in assuming that the medium under study is composed successively of a diattenuator, M_D, a retarder, M_R, and a depolarizer, M_Δ, so that $M = M_\Delta M_R M_D$. For a FLC cell in normal incidence, no diattenuation is expected, and the values obtained are only associated to the experimental noise. In that case, the parameters used for the characterization are the depolarization index, P_D, the retardance, R, the fast axis-orientation, α_R, and -ellipticity, ε_R [8]. They are linked to the director-orientation and -distribution in the FLC cell. The orientation of the fast axis, α_R, is equal to the apparent angle, θ_{app} (Fig. 2). It represents the angle between the projection of the director on the (O_x, O_y) plane and the smectic layer normal (O_y axis). The retardance, R, is linked to the angle, χ, by the following relationships:

$$R = \frac{2\pi\Delta n(\chi)d}{\lambda_0} \qquad\qquad \Delta n(\chi) = \frac{n_e n_o}{\sqrt{n_e^2 Cos^2[\chi] + n_o^2 Sin^2[\chi]}} - n_o \qquad (1)$$

where n_o is the ordinary index of the FLC, and n_e is the extraordinary one. As R gives insight into the director position in the (O_y, O_z) plane, the knowledge of θ_{app} and χ is sufficient to describe $\vec{n}$ in a 3-D system on condition to assume a homogeneous distribution of the latter within the light beam. The last two parameters characterize the homogeneity of the director distribution. The ellipticity of the FLC cell-associated retarder appears when the distribution of the director orientation is non-homogeneous in the O_z direction (twist). Indeed, if one considers a succession, in the O_z direction, of thin layers of thickness, ξ, composed of molecules with an orientation, ψ_i, the resulting MM is $M_Z = \prod M_R(\xi, \psi_i)$, where M_R is the MM of a linear retarder of thickness, ξ, and fast axis orientation, ψ_i. If ψ_i is varying across the total thickness, d, M_z will be an elliptic retarder. Ellipticity gives thus insight into the homogeneity of the director orientation along the cell thickness. The depolarization index, P_D, is equal to 1 for an elliptic retarder. If one now

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considers, within the light beam (*~0.5mm*), multiple areas with different orientation, ψ_i, the resulting MM is $M_{xy} = \sum M_R(d,\psi_i)$. If ψ_i is varying within the (O_x,O_y) plane, P_D associated to M_{xy} will decrease. One should note that this statement is valid because the FLC cell was imaged on a depolarizing medium, in turn imaged on the entrance of the detection system. The depolarization index is thus expected to evidence the presence of domains with different apparent angles at the beam scale. A key-asset of this method is the simultaneous extraction of all of the above parameters. It is the warrant of a good correlation between them, conversely to the case where several experiments are needed to quantify the described effects since they generate additional noise [4].

A square voltage between ± 15 V at 30 Hz was applied to the FLC cell to investigate the dynamics of switching between the up and the down states. But the great difference between the CCD refreshment time and the duration of switching (≈ 1 ms vs $\approx 100\,\mu$s) hindered its measurement over a single period. This drove us to synchronize the detection with the applied voltage; then, the acquisition was progressively delayed of the same time-increment to reconstruct the transition. Each MM was measured with a gate aperture of $5\,\mu$s and 25 accumulations. A total of 50 points were acquired with an increment of $6\,\mu$s. Figure 3 illustrates the time evolution of the four parameters under study.

The rotational viscosity, η, of the liquid crystal is determined through extraction of the switching duration from $\alpha_R(t)$. Indeed, in the simplest theoretical model, the director behavior over the transition is described by $\eta.\partial\phi/\partial t = P_S.E.Sin[\phi]$, where ϕ is the angle between the applied electric field, $\vec{E}$, and the spontaneous polarization, $\vec{P_S}$ [9]. The transition rise-time t_{ud} is given by the interval between, for example, 10 and 90%. In this study, the spontaneous polarization value was given by $P_S = 33$ nC/cm^2, the applied voltage was set at $E \approx 10$ V/µm and

the measured rise-time was $t_{ud} \approx 70 \,\mu s$. From the solution of the above equation expressed in [9],

the relation $t_{ud} = \eta/(1,32.P_S.E)$ led to $\eta \approx 0.175$ Pa.s for the rotational viscosity. However, this

simple relation gives a poor account of the true dynamic behavior since it fails to give a good fit

of the experimental curve when only the ferroelectric torque is taken into account. A more

elaborated model of the FLC switching dynamics will be presented and tested against

experimental data elsewhere.

The minimum observed on the plot of $R(t)$ (Fig. 3) corresponds to the lowest value of the

angle, χ (see Eq.(1)). It indicates that, over the transition, the molecules get out of the cell plane

for a while. Indeed, according to the theory, in FLC cells the molecules switch around a cone.

One should also note that P_D is equal to 1 (uniform state) in the up and down states. The

short minimum (≈ 0.900) means that, within the light beam, the various molecules switch from

the up to down states at different times (dispersion of switching starts). However, a domain

propagation-induced switching would have strongly lowered depolarization value (≈ 0.650).

According to Fig. 3, ε_R is close to $0°$ in the up and down states, which means that the

director distribution is almost homogeneous in the O_z direction despite a small twist, likely

located near the surfaces due to the anchoring strengths. The change in ε_R sign indicates a change

in the direction of twist. Analysis of ε_R behavior at the transition is more complex because of the

non homogeneity of the director distribution in the (O_x, O_y) plane. On-going researches are

focused on the development of a more detailed model of the director distribution within the cell

to further quantify ellipticity and depolarization.

To gain more insight into the molecule behavior over the up/down switching, the

trajectory of the director extremity in the (O_x, O_z) plane was plotted by using the experimental

angles, χ and θ_{app}, linked to the director coordinates (x,y,z) by the relations $x = r.Sin[\chi].Sin[\theta_{app}]$,

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$y = r.Sin[\chi].Cos[\theta_{app}]$, $z = r.Cos[\chi]$, where r is the size of the rigid stick that symbolizes the molecule. Figure 4 illustrates the trajectory followed by the molecules when they switch from the up to down states for two different positions of the light beam within the cell. One should note changes (from a cone to a deformed ellipse) in the shape of the different trajectories. This finding suggests the existence of different layer structures within the cell. The authors are currently trying to give more thorough interpretations of these results, which will be published elsewhere.

This study demonstrated that snapshot Mueller matrix polarimetry can give account of the behavior of FLC molecules over a fast transition. All of the parameters relative to the collective motions of the molecule director were simultaneously extracted from a single acquisition shot through use of a Lu and Chipman decomposition. For liquid crystal characterizations, the simplicity and power of time-resolved Mueller polarimetry together with the resulting enhancement of the signal-to-noise ratio make it more attractive than classical optical techniques. Further investigations are in progress to better describe the theoretical and experimental dynamics in FLCs, where the chevron structures of the smectic layers will be taken into account.

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Fig. 1. Snapshot Mueller Matrix Polarimeter in the configuration (*e,e,5e,5e*).

Fig. 2. SSFLC cell and representation of the director ($\vec{n}$) in an (*x,y,z*) coordinate system; FLC molecules are symbolized by rigid sticks. The spontaneous polarization $\vec{P_S}$ is perpendicular to the director, and tangential to the circle of intersection of the cone with the boundary plane of the layer.

Fig. 3. Evolution of the parameters, ($\alpha_R(t)$, $R(t)$, $P_D(t)$, $\varepsilon_R(t)$), throughout the up/down transition. The voltage switches from +15V to -15V at $t = 100\,\mu$s.

Fig. 4. Representation of the director end in the (O_x, O_z) plane during the up/down transition for two positions of the beam within the cell. The trajectory showed in the left is calculated from the parameters depicted in Fig.3. This representation assumes a collective movement of the directors (no depolarization). The associated error bars are for an uncertainty of 0.5°on the retardance value; r is normalized to 1.

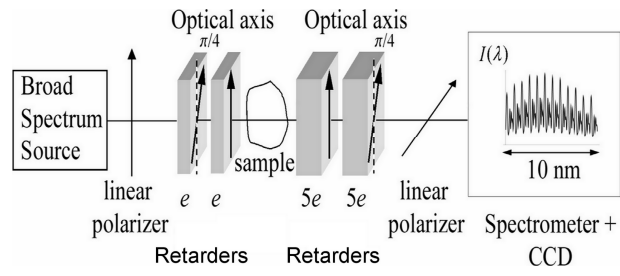Fig. 1. Snapshot Mueller Matrix Polarimeter in the configuration (*e,e,5e,5e*).

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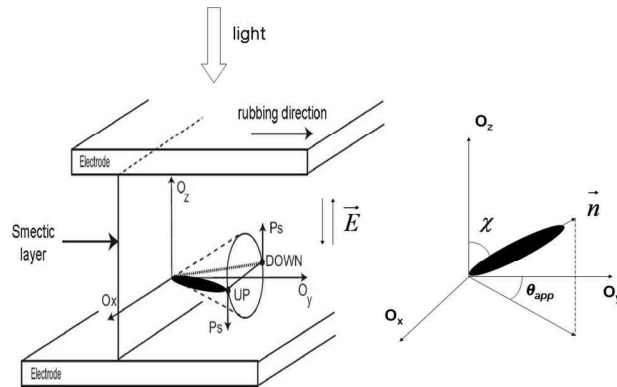
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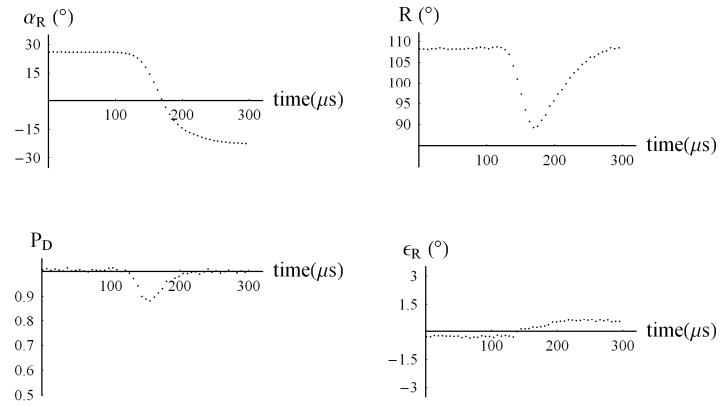
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