Electrical Modulation of Coherent Scattering in Dual Frequency Liquid Crystal Tissue Models

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Molecular orientation clusters are created in a cell of non aligned liquid crystal to study the light propagation in electrically controllable anisotropic non uniform media. Coherent scattering phenomenon and predominant role of large orientation clusters in such scattering are demonstrated. Fast relaxation of molecular orientation and modulation of light scattering in those clusters is obtained by using "dual frequency" liquid crystal mixtures and frequency modulation of the drive signal. The obtained results show the good potential for the creation of electrically tunable tissue models by using liquid crystalline materials.

Keywords Dual frequency; electro-optics; liquid crystals; light scattering; tissue optics

Introduction

Light propagation in complex (disordered and anisotropic) media is of paramount interest in medical diagnostics for tissue study [1,2]. An example of such media is the human skin, the non uniform and anisotropy character of which is shown in the Figure 1. Scientific investigation of real samples of such tissue is however rather limited given the important moral and legal considerations. That is why there is a necessity of creation of artificial tissue models (sometimes called "phantoms"). Preferably, such models should be dynamically varied to conduct a comparative analyzes of different kinds of tissue. The presently used method of such variation is the mechanical stretching of a given "elastic" scattering model. This stretching however provides a rather limited variation of parameters of interest and, in addition, "couples" (changes simultaneously) some key parameters, such as the matrix anisotropy and the distance of scattering non uniformities. To the best of our knowledge, there are no methods (including electrically variable) to obtain better tissue models so far.

We believe that liquid crystals (LC) could be used to create such models since they are naturally optically non uniform and anisotropic. The application of the electric field here may change the average anisotropy and refractive index modulation contrast while preserving the distance of scattering centers and their

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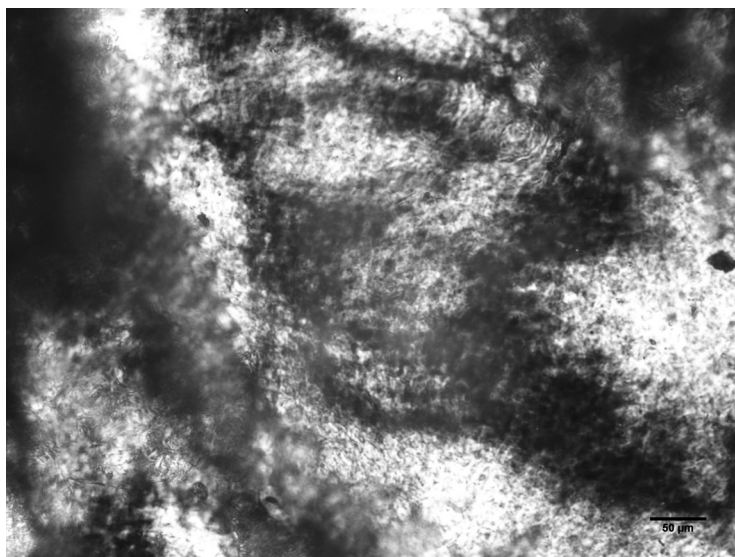
Figure 1. Demonstration of the non uniform anisotropy character of the living skin. The thin layer of human skin (extremity of the finger of one of the authors, TG) is placed between two polarizers and is illuminated by white light. The horizontal bar (50 μm) on the right bottom corner is introduced to show the spatial scale of orientation defects.

form. In addition they can be easily doped by reactive, absorbing or chiral molecules and be electrically controlled. In the case of their doping by reactive molecules, the cell morphology may be "programmed" (and then reversibly tuned by an electric field) by the photo polymerizing light intensity distribution in space (e.g., by using an amplitude mask) to create different defect structures, such as gradient-polymer stabilized liquid crystals (PSLCs), see Figure 2. Those are the reasons why, in the present work, we propose, what we believe is for the first time, to use LC materials as *tissue models* and demonstrate the feasibility of such electrically variable models.

The propagation of light through LC containing scattering media was already well analyzed in polymer dispersed liquid crystals (PDLCs), [3]. Apart of their potential use in privacy windows and scattering (polarizer free) displays, very interesting scattering behavior can be anticipated in such media under certain conditions. Thus, a significant increase of "ballistic" transmission was recently demonstrated in a scattering media by the appropriate choice of the input light's phase front modulation [4]. On the other hand, electrically tunable interferential quenching was demonstrated in large-droplet PDLCs [5] and non-aligned LC clusters [6]. Both approaches were using the phenomenon of electro optic molecular reorientation and corresponding refractive index modulation to control the quenching degree [7]. The phenomenon was interpreted in the framework of "single" scattering model. The large sizes of orientation clusters were generated (in Ref. [6].) by introducing relatively big micro spheres into the LC matrix and by drop filling this microsphere – LC dispersion (MSLCD) between two substrates with non-rubbed surfaces. Thus, significant number of clusters, disclination walls and other orientation defects of the director (average orientation of axes of LC molecules [8]) were formed in the cell. It is well known however that such defects slow down significantly the orientational relaxation of the LC [9], which may be a drawback for potential applications.
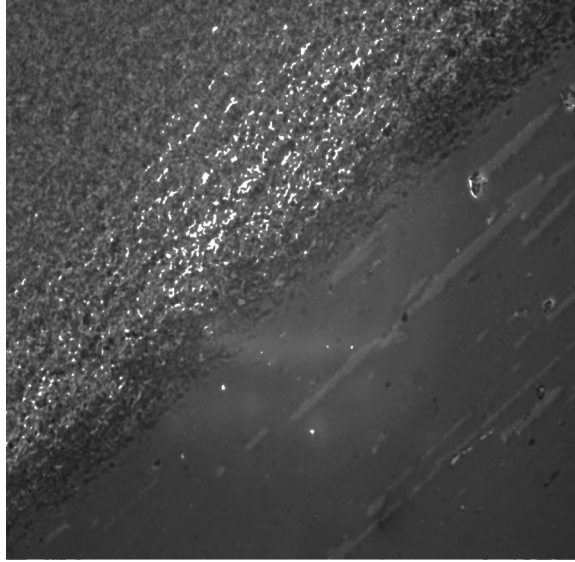
Figure 2. Example of a gradient-polymer stabilized liquid crystal structure, obtained by protecting the right bottom corner of the LC cell (that was doped by reactive monomers) and by UV polymerizing the remaining (upper left corner) part to create a gradient of polymer network. The application of voltage (6.8 Volts) results in the non uniform reorientation of LC in correlation with the polymer network's spatial distribution (courtesy of *L. Zohrabyan*).

In the present work we build non uniform MSLCD cells, demonstrate interferential quenching phenomenon, show the predominant role of large orientation clusters (versus the role of micro spheres) and use a dual frequency LC to build scattering models with relatively fast (faster by an order of magnitude compared to "normal" LC based models) electro-optic response time.

Material System and Experimental Set-Up

We use the nematic LC mixture MLC-2048 (from Merck) as host material. In contrast to "traditional" LCs, the MLC-2048 changes the sign of its dielectric anisotropy $\Delta\varepsilon = \varepsilon_{II} - \varepsilon_{\perp}$ for different driving frequencies allowing thus the potential fabrication of fast modulators by "forced" relaxation of the director [10] (see the *Inset* of Fig. 3 for the dependence of the parallel ε_{II} and perpendicular $\varepsilon_{\perp}$, with respect to the director, dielectric constants of the LC versus driving frequency). Several typical values of $\Delta\varepsilon$ at various driving frequencies are given for example (at room temperature): $\Delta\varepsilon = 3.22@1\,\text{kHz}$; $\Delta\varepsilon = 0.72@10\,\text{kHz}$ and $\Delta\varepsilon = -3.08@50\,\text{kHz}$.

The microspheres (Soda lime glass) were purchased from Polysciences inc. in the form of powder and were mixed with the MLC-2048 without further modification. The diameters of those spheres were in the range from $10\,\mu\text{m}$ to $30\,\mu\text{m}$. The obtained dispersion was homogenized using magnetic mixer and ultrasound bath. The choice of the LC and glass microsphere was made taking into account the fact that the refractive index of the glass n_g (1.51 @ 589 nm) is between the ordinary n_o (1.4978 @ 589 nm) and extraordinary n_e (1.7192 @ 589 nm) refractive indexes of the LC (at room temperature). Thus, the "traditional" multiple scattering here should
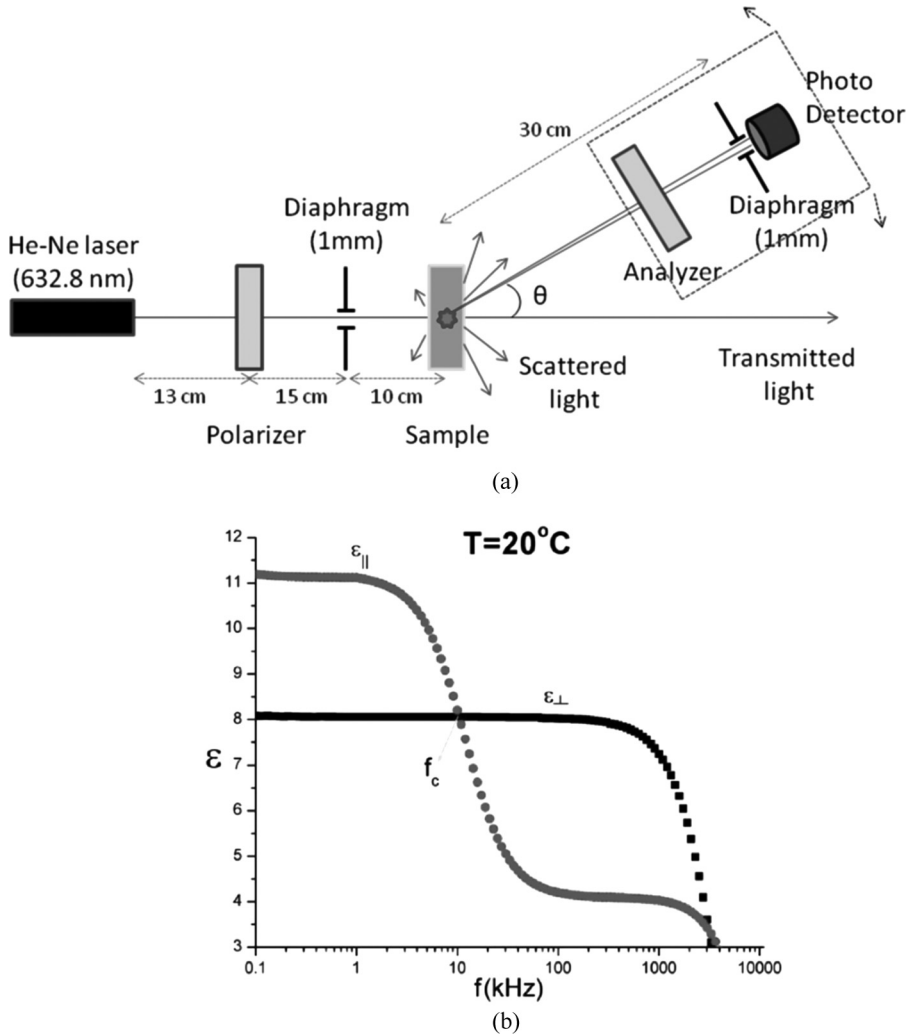
(a)

(b)

Figure 3. Schematic representation of the experimental set-up used for the electro optic scattering measurements. The *inset* represents the frequency dependence of dielectric constants of the host material used (MLC-2048). The f_c denotes the critical frequency when the change of sign happens for the dielectric anisotropy.

demonstrate a rather simple dependence (with one possible "clear" state when $n_g = n_{LCeff}$) upon the reorienting voltage (see later).

Two commercially available ITO coated glass substrates were used to build sandwich-like cells. Those substrates were first cleaned and then covered by a thin film (approximately $1\,\mu m$) of PMMA by spin coating, the PMMA being dissolved in advance in a solvent (Toluene). The PMMA coated ITO-glass substrates were then heated in an oven (to remove the rest of the solvent) prior of their use for building the LC cell sandwiches. The PMMA surface was not rubbed to avoid the formation of an easy axis and to minimize the polarization dependence of the cell. The substrates were slightly mutually shifted in opposed directions to partially
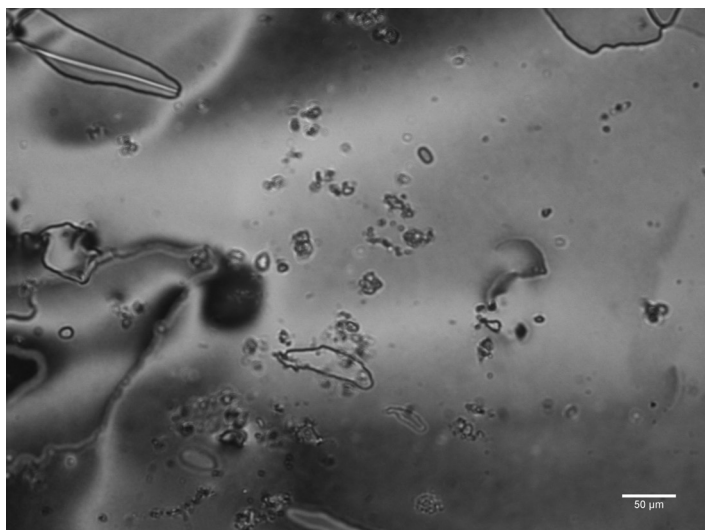
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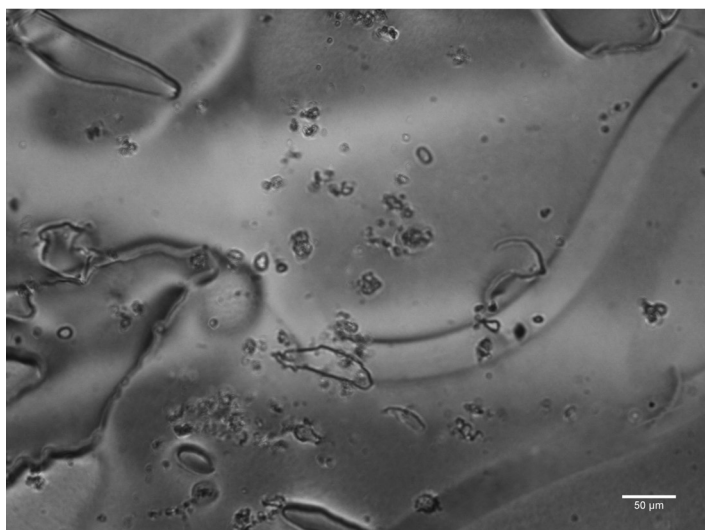
(b)

Figure 4. Polarizing microscope micro photography of MSLCD (cell based on MLC-2048 with 5 Wt% of glass spheres with diameters ranging from 10 μm to 30 μm) for (a) V = 0 Volts and (b) V = 9.9 Volts. The horizontal bar (50 μm) on the right bottom corner is introduced to show the spatial scale of orientation defects.

expose the ITO areas, which were then used to wire the cell for electro optic experiments.

The injection of the MSLCD mixture was made in the drop-fill mode (see below) in the isotropic phase of the mixture using a heat sink (the substrates also were maintained at a temperature where the mixture is in the isotropic phase). To perform the drop-fill, the bottom substrate was positioned in the horizontal plane; U-form polymer tape was used as spacer providing cell gaps ranging from 40 μm to 85 μm.
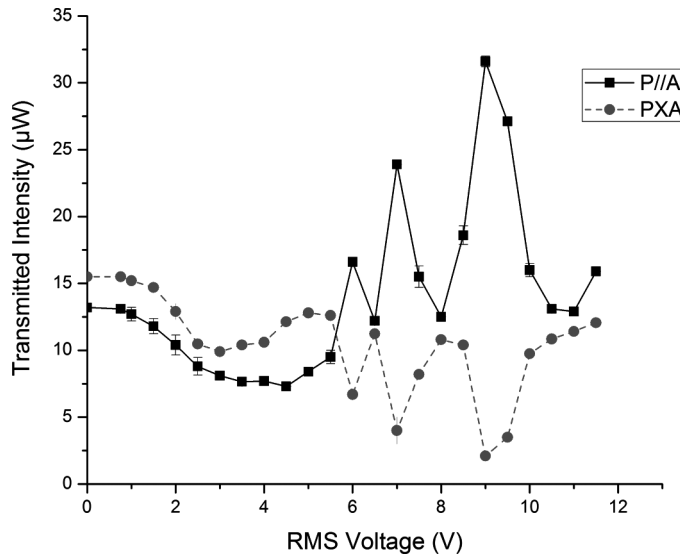
Figure 5. Transmission signal's (in microwatts) dependence upon the applied RMS voltage for the MSLCD dispersion of MLC-2048 with glass spheres (with diameter of 10–30 μm). The solid line (squares) represents the behavior of the transmitted light having the same polarization as the incident probe beam. The dashed line (spheres) represents the signal with perpendicular polarization.

The drop of the MSLCD was then dispensed in the center of that substrate. The second substrate was then positioned on the top of the first one forcing the drop of MSLCD to adopt a planar form. UV curable adhesive was then used to seal the periphery of the cell.

The experimental set-up, used for the electro optic scattering experiments, is presented in the Figure 3. CW He-Ne laser (operating at 632.8 nm) beam was used as probe. A polarizer was used to insure the linear polarization of the incident probe beam. The incident beam was "cleaned" by a first diaphragm with diameter of 1 mm. The incidence angle of the probe on the MSLCD cell was 0° (normal incidence). The directly transmitted and scattered light were detected by means of a group of elements (analyzer, second diaphragm and photo detector), which were all mounted on the same rotation stage, the rotation being performed in the horizontal plane and the center of rotation coinciding with the incidence point of the probe beam on the vertically positioned cell of MSLCD (its substrates were perpendicular to the horizontal plane). The second polarizer (analyzer) was oriented in parallel and (switched to) perpendicular directions with respect to the incident probe's polarization. The working surface of the photo detector was larger than the diameter of the second diaphragm. Thus, the diameter (1 mm) of that diaphragm and its position with respect to the MSLCD cell (at 30 cm) define the angular reception of the detection system being $\approx 0.3 \times 10^{-2}$ rad.

Experimental Results

The obtained material morphologies and experimental results have some dispersion, while their behavior is rather reproducible taking into account the way of
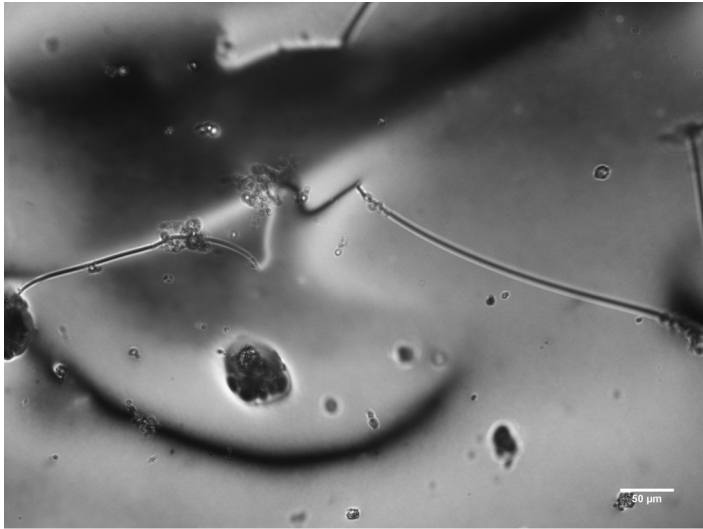
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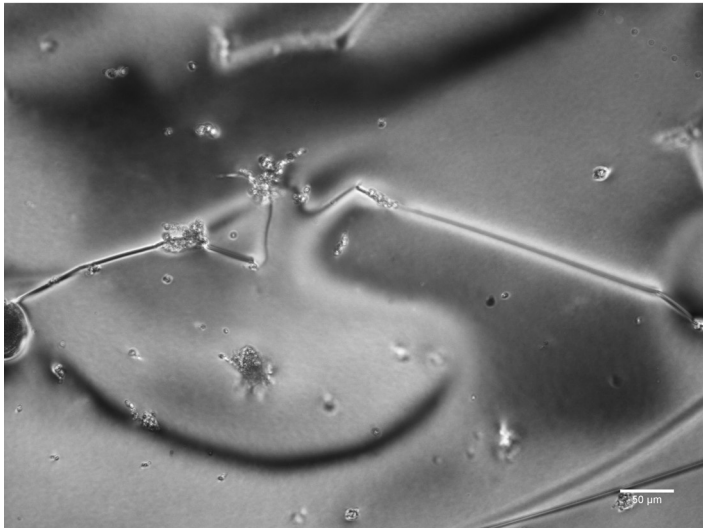
(b)

Figure 6. Polarizing microscope micro photography of MSLCD (cells based on MLC-2048 with 0.5 Wt% of Polystyrene spheres with diameter of 0.5 μm) for (a) $V = 0$ Volts and (b) $V = 12.18$ Volts. The horizontal bar (50 μm) on the right bottom corner is introduced to show the spatial scale of orientation defects.

fabrication of the MSLCD cells (no rubbing and random dispersion of micro-spheres). They all show the expected disordered and cluster-rich character. The Figure 4 is a typical micro-photography of such a cell at the ground state and at the electrically reoriented state ($U = 9.9$ V, AC sin-form signal at 1 kHz). As it can be seen, there are orientation defects around the microspheres, large orientation clusters as well as disclination lines. Importantly, the characteristic sizes of majority
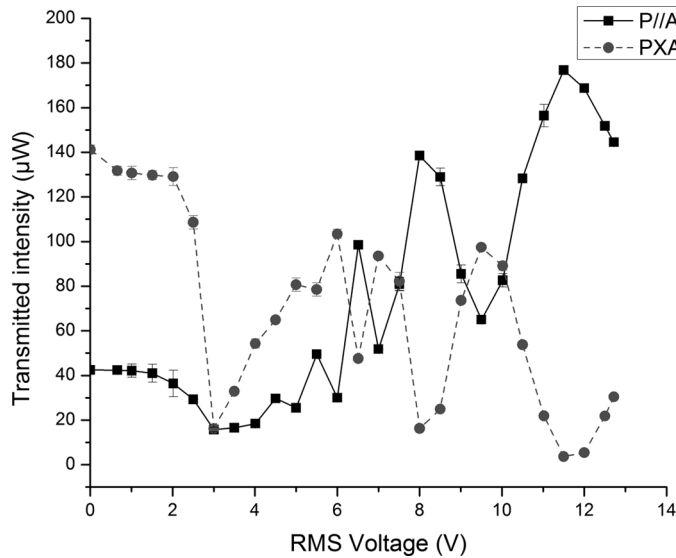
Figure 7. Transmission signal's (in microwatts) dependence upon the applied RMS voltage for the MSLCD dispersion of MLC-2048 with Polystyrene spheres (with diameter of 0.5 μm). The solid line (squares) represents the behavior of the transmitted light having the same polarization as the incident probe beam. The dashed line (spheres) represents the signal with perpendicular polarization.

of optical non uniformities are larger than the thickness of the cells. The optical properties of the obtained structures may be easily electrically tuned (see Fig. 4a versus Fig. 4b) since the relative contrast of corresponding refractive index modulation depends upon the applied voltage. Consequently, light transmission also depends upon the applied voltage. It is important to notice that, in contrast to the mechanical or "stretch" tunable models; the properties of the matrix and scattering centers in our case are controlled by the electrical field, but the relative positions and forms of light scattering defects are not changed making the comparative studies much easier.

The study of the angular distribution of the scattered light has shown a standard monotonically decreasing (with the increase of scattering angle θ) behavior. For the sake of shortness those data are not shown here. In contrast, the evolution of the "directly" transmitted signal (for $\theta = 0°$ in Fig. 3) is shown in the Figure 5 for two polarization components of the transmitted light (parallel and perpendicular to the polarization of the incident probe beam). The solid line (squares) represents the case when the analyzer allows the transmission of light, which is polarized parallel with the polarization of the incident probe beam, while the dashed line (circles) represents the perpendicular orientation of the analyzer. Both curves initially demonstrate a similar (decreasing) behavior for growing voltages. This is probably related to the increase of scattering at large angle (out of acceptance angle) for both polarizations. Then interferential quenching is observed in the form of strong anti phase oscillations (for cross polarizations) with voltage variations, achieving (e.g., for the parallel polarization component) modulation depth as high as 72% (between 4.6 V and 9 V) and approximate polarization discrimination ratio of 15 (at 9 V). This transmission curve is
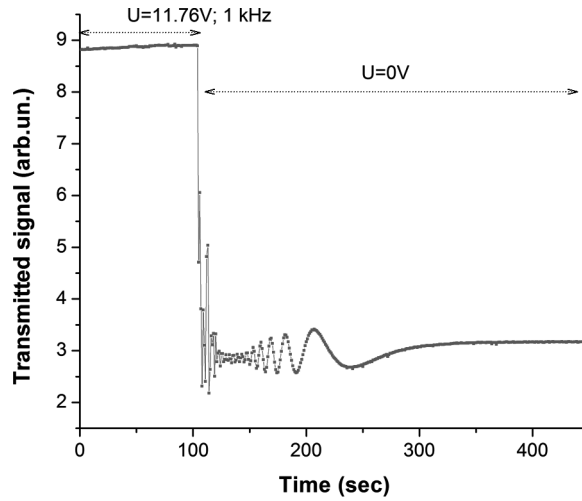
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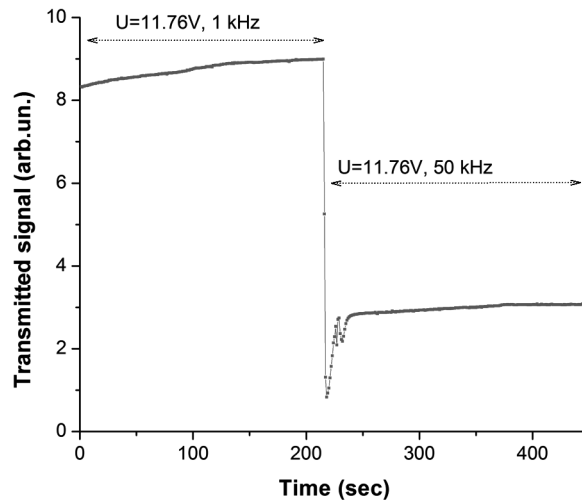
(b)

Figure 8. Typical dependence of the transition signal upon time for (a) ''free relaxation,'' when the voltage is simply switched off, and (b) ''forced'' relaxation, when the voltage is maintained, but the frequency is switched from 1 kHz (positive dielectric anisotropy) to 50 kHz (negative dielectric anisotropy). Approximately an order of magnitude of acceleration is observed.

reproducible for increasing and decreasing voltages and there is no noticeable memory effect.

It is worthy to note that the presence of large micro spheres (with diameters up to 30 μm comparable to the thickness of the cell 40 μm) dispersed in the LC matrix could already justify the observation of this interferential quenching effect. This is not the case when the thickness of the cell is significantly higher than the sizes of defects and the characteristic distance between scattering centers providing thus strong multiple scattering. In some cases (e.g., in stretched PDLCs or PSLCs), this

multiple scattering may also demonstrate strong anisotropic character too (with polarization discrimination factors up to 600), but its electrical tuning still shows a rather monotone variation of scattering without interferential oscillations [5,11,12].

To establish the relative roles of large orientation clusters and micro sphere generated non uniformities, we have replaced the large (10 μm–30 μm) micro particles by much smaller ones (polymer spheres with diameters of ≈0.5 μm). Similar results were obtained both for the morphology (Fig. 6) and for the electro optic response (Fig. 7). First of all, here also, in contrast to the "stretch" tuning models; the properties of the matrix and light scattering centers are controlled by the field, but relative positions and the forms of those defects are not changed. Also, while the initial distribution of scattered energy here is different for two cross polarized components, both of them are decreasing with the voltage increase as in the previous material dispersion. Then, again interferential quenching of scattering is observed showing similar number of anti-phase oscillations (as in the case of large micro particle dispersions). This shows the dominant role of the non-rubbing of the cell substrates (which is at the origin of orientation clusters) compared to the micro sphere dispersion. It is worthy to note that, e.g., for the parallel polarization component, rather strong modulation depth (>90%, between 3 V and 11.5 V) is achieved. Also, a very strong polarization discrimination ratio (almost 50) may be observed at 11.5 V.

The response time of obtained structures (e.g., with polymer spheres of 0.5 μm diameter) was tested by two methods. In the first approach, the cell was placed between parallel polarizer and analyzer and a reoriented state of clusters was created by applying low frequency (1 kHz) excitation of RMS voltage U = 11.76 V. Then the voltage was simply switched off. The Figure 8a shows the detected relaxation of the directly transmitted signal. As it can be seen, the total relaxation time is very long, at the order of 300 sec! (let us note that the typical relaxation time for a well aligned LC in a 40 μm thick cell is at the order of few seconds). This makes such samples rather unpractical.

In the second approach, the initial reorientation was achieved in the same way, while the voltage was not switched off, but was kept the same. However, the excitation frequency was switched from 1 kHz to 50 kHz, at which, the dielectric anisotropy of the LC is negative. In this case, the LC molecules are "repulsed" from the electric field instead of "free" relaxation. The obtained acceleration (see Fig. 8b) is remarkable, at least by an order of magnitude.

Resume and Conclusion

We have developed a simple method for the fabrication of randomly distributed orientation clusters for the study of the properties of tissue models or other scattering media. Their properties, such as effective anisotropy and scattering, may be easily controlled by the application of a weak electric field. Electrically tunable interferential quenching is demonstrated in these materials and it is shown that this quenching is primarily related to the orientation clusters rather than the dispersions of micro particles. The use of dual frequency LC is shown to allow the fast electro-optic manipulation of the parameters of light scattering. We believe thus that LC materials are promising candidates for artificial tissue model fabrication.

Acknowledgments

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