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SELECTIVE AMPLITUDE-FREQUENCY ELECTRO-OPTICAL MODULATION BY POLYMER-DISPERSED LIQUID CRYSTAL FILMS ALIGNED BY TEFLON NANOLAYERS

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Abstract

We report the amplitude-frequency electro-optical (EO) modulation by a specific type of polymer-dispersed liquid crystal (PDLC) composites – planar single layers of large nematic microdroplets aligned by teflon nanolayers. Such a surface modifying PDLC system exhibits a selective modulated 2nd harmonic EO response by the dielectric oscillations of the nematic director. The band-like behavioural characteristic of the amplitude-frequency modulation of light, achieved by the single-layered PDLC films, can be tuned by AC voltage applied on the PDLC cell. This property can be applied to tunable EO modulators operating in the infrasound frequency range.

Key words: polymer dispersed liquid crystal (PDLC), PDLC films, single layers, electro-optical properties, amplitude-frequency modulation

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1. Introduction. Due to their attractive electro-optical (EO) properties, the polymer-dispersed liquid crystal (PDLC) composite films [1] are currently applied as smart EO materials in switchable glasses [2,3], for displays [4] and devices for active control of light [5,6]. Single-layered PDLC films consisting of large microdroplets of liquid crystal (LC) disposed in an optically-transparent polymer matrix have also found advanced EO applications [7–10].

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In this work, we studied PDLC composites of large droplets of the nematic LC commercially known as E7 and the photocurable polymer NOA65, produced by photopolymerization-induced phase separation. By this process, single droplets of LC with a size of 20 µm were confined in thin films (10 µm thickness) between transparent conducting substrates coated with teflon nanolayers. The teflon nanolayers determine the surface anchoring and the orientation of the LC in droplets. The use of rubbed teflon nanolayers, as orienting surfaces for LC orientation, is well known [11, 12]. In particular, we have previously reported alignment effects of teflon nanolayers on the nematic 5CB [13, 14]. Here, we report the amplitude-frequency EO modulation characteristics of E7/NOA65 PDLC composites – single layered films of relatively large nematic droplets aligned in this manner.

2. Experimental. The PDLC single layered films were prepared as composites of nematic LC E7 (from Merck) and the UV-curable polymer NOA65. The E7/NOA65 mixture (50:50 wt.%) was heated well above the nematic-isotropic phase transition temperature of E7 (59 °C), and then filled in the empty cells. The cells were assembled with 1 mm-thick glass plates coated inside by thin transparent conductive layers of indium tin oxide (ITO). Mylar spacers with a thickness of 10 µm were used. The PDLC composites were obtained by photo-polymerization-induced phase separation (illumination with UV light was employed). It should be pointed out that after the teflon nanolayer deposition on the ITO glass, the UV transmittance of the latter (at the wavelength of 365 nm) is slightly reduced (from 81.5% to 78.3% in our case, as we have precisely measured by photometry).

The surface modification (both a morphology change and LC alignment) of the PDLC system was performed by rubbed teflon-deposited ITO glasses before the PDLC preparation. The ITO-coated glasses were covered by teflon nanolayers using a lower temperature modification [13] of the Wittmann and Smith technique [11]. Thus, configurations of unidirectional grooves of the rubbed teflon with an adjacent spacing in the submicrometer range, an average thickness of a few nm, and a width from around 100 nm are expected, in correspondence to the experimental procedure used in [13, 14]. The teflon rubbing of both glasses of the PDLC cells was orthogonal.

The experiment setup for measurement of the frequency dependence of the light modulation by PDLC single layers was described in details elsewhere [8]. Briefly, the transmittance of He-Ne laser beam (wavelength λ = 632.8 nm) through the electrically driven PDLC was investigated by a photodiode and computer-controlled lock-in amplifier (SR830, Stanford Research Systems). The temperature of the PDLCs was kept at 35 ± 0.1 °C by a Mettler FP82 hot stage.

3. Results and discussion. The PDLC morphology (Fig. 1) is characterized by non-ordered LC droplets, as well as a predominantly twisted alignment

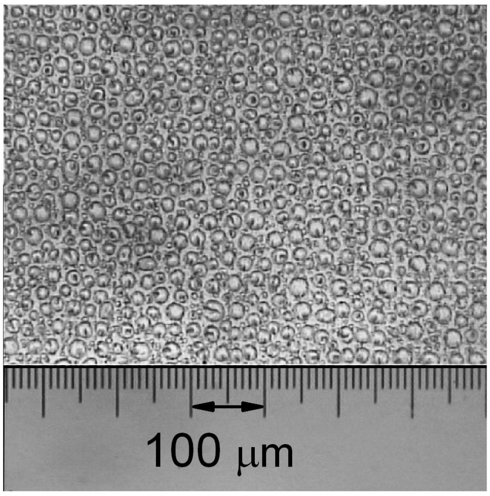

Fig. 1. Optical microscopy image (magnification ×10) of PDLC single layer with a thickness of 10 μm is studied here. The average size of the droplets is 19.5 μm (with a standard deviation of 2.8 μm)

of the LC in the droplets due to the orthogonal anchoring. Reasonably, owing to the orthogonal rubbing of both plates of the PDLC cell, the LC orientations induced by both plates are orthogonal, thereby no unidirectional LC orientation could be achieved. The layered PDLC composite contains relatively large LC droplets with oval but flattened shapes. The droplets are arranged in a planar single layer and are quite uniformly distributed.

Figure 2 presents the 2nd harmonic EO modulation of transmitted laser light (the amplitude-frequency modulation – the amplitude of the dielectric oscillations versus the frequency of the applied electric field) measured for the single-layered PDLC with orthogonally rubbed teflon nanolayers. In a certain frequency region (in our case – in the low-frequency region), this frequency dependency is a band-like. For comparison, in Fig. 2, it is also given the corresponding frequency-dependent curve measured for PDLC prepared in much the same manner in an identical cell but with no alignment teflon nanolayers. As it is seen, in this particular case no selective EO modulation is registered.

The specific band-pass behaviour can be explained as follows. At the high-frequency side of the band, the PDLC EO response is reduced by the increasing frequency due to the well known effect of viscosity damping of the nematic director oscillations. This effect originates from the balance of driving torques acting on the nematic director. External AC electric field creates a dielectric torque

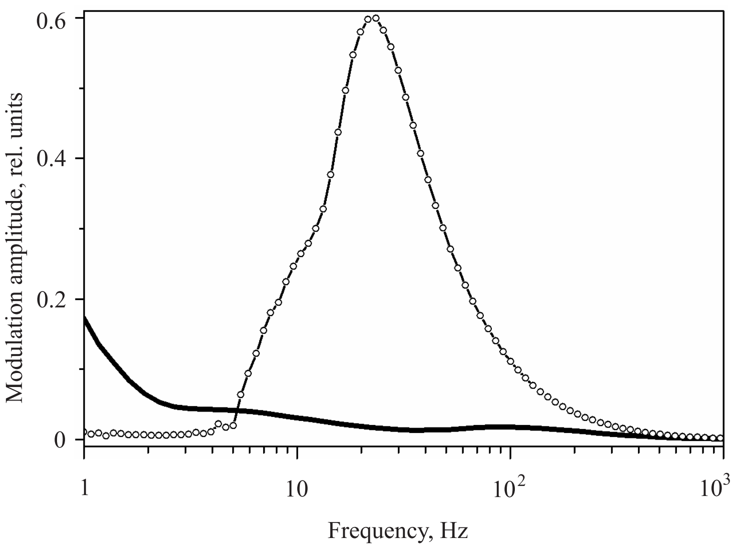

Fig. 2. EO modulation of laser light (wavelength $\lambda = 632.8$ nm) transmitted through single-layered PDLC cells with a thickness of 10 µm: with orthogonally rubbed teflon nanolayers on the ITO glasses (line with open circles); and without alignment layers on the ITO glasses (bold line). In both cases, the temperature of the PDLC cell was 35 °C

that is balanced by a friction term [15]. This leads to an exponential decreasing of the amplitude of the deviation of the LC molecule orientation from the average director equilibrium state. In the low-frequency region, the decrease of the modulation amplitude with the decrease of the frequency is ascribed to another known effect – the ion screening of the applied electric field due to domination of free ion conductivity.

The operation of PDLC devices is achieved by switching between their translucent and transparent states under the application of an external AC electric field [1]. The electric field re-orients the director in the droplets in the field direction (due to the positive dielectric anisotropy of E7), changing the optical properties of the PDLC composite film. As a result, a refractive index mismatch between the droplets and the polymer matrix (translucent state) is removed (transparent state). The critical voltage required to switch the device is related to the Schadt–Helfrich effect.

The problem of the LC reorientation by the internal electric field inside the LC droplets is rather complicated. Besides, of great significance for EO PDLC properties is the electrical conductivity of LC material, for the physical properties of droplets, polymer matrix and droplet surface anchoring. Each of these factors may affect both the effective dielectric permittivity and the internal field, thus indirectly affecting the light scattering properties of the PDLC film. In the case of conductive material, the dielectric constant is a complex quantity

$$\varepsilon = \varepsilon_{\mathrm{LC}} + i\frac{\sigma_{\mathrm{LC}}}{\omega}, \tag{1}$$

where σ_{LC} is the LC conductivity and ω is the angular frequency of the external AC electric field. Due to many circumstances of experimental relevance, the conductivity often prevails over the dielectric response, giving rise to a significant magnitude variation of the internal electric field. The conductivity effects are large at lower frequencies, at which they can dominate. In this case, due to many circumstances of experimental relevance, the conductivity prevails over the dielectric response, giving rise to the magnitude of the internal electric field

$$E_{\mathrm{int}}|_{\omega\to 0} = \frac{3E_{\mathrm{ext}}}{2 + \dfrac{\sigma_{\mathrm{LC}}}{\sigma_p}}, \tag{2}$$

where E_{ext} denotes the magnitude of the external electric field applied on the PDLC medium, and σ_p is the conductivity of the polymer component of the PDLC composite.

Practically all LCs contain free ions. The amplitude of the EO PDLC oscillations is closely related to the presence of these mobile charges in the LC droplets. In particular, the amplitude of EO modulation by PDLC diminishes due to the presence of mobile charges in the LC droplets. The free ions can reduce the AC electric-field driven reorientation of LC, and even severely terminate it. This effect known as a screening of the external electric fields does weaken the strength of the electric field that orients the LC molecules within the PDLC composite films by separation of charges at the opposite interfaces of the LC droplets. The phenomenon can be attributed to the Maxwell–Wagner–Sillars effect relevant to the presence of charges located at the polymer/LC interfaces. Below the charge relaxation frequency $f = \sigma_{\mathrm{LC}}/\varepsilon_{\mathrm{LC}}$ (conductive mode of the PDLC operation), ionic currents arise that produce a depolarization field [16, 17] across the PDLC film and along the direction of the external electric field. This depolarization tends to screen the applied field and therefore switch the PDLC film from a transparent to a scattering state by lowering the frequency f, e.g. towards 1 Hz and less. As a consequence of the separation of free charges, the applied voltage drop is compensated on the electric double layers formed at the opposite sides of the droplets [18]. Finally, owing to the electric double layers, the dielectric molecular reorientation becomes less effective and the LC molecule oscillations can be cancelled.

Our observations for the impact of the screening on the frequency dependence of the dielectric oscillations by the aligned PDLC under study are consistent with

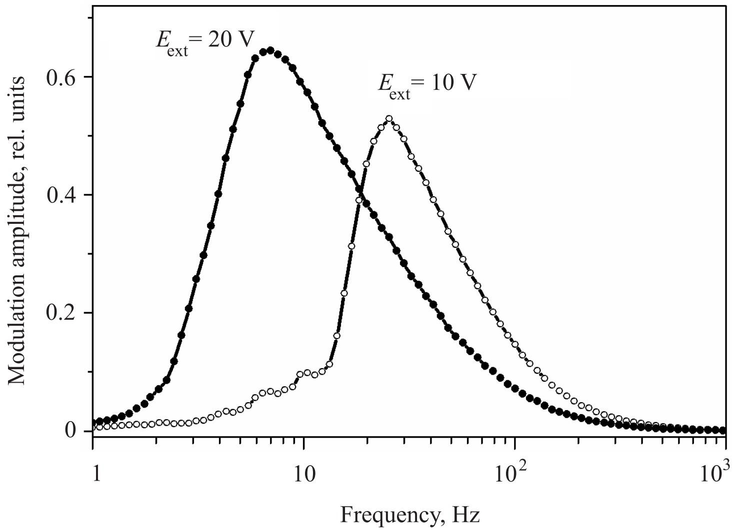

Fig. 3. The amplitude-frequency EO response (by light transmittance, $\lambda = 632.8$ nm) of PDLC cell (10 µm thick). Spectra measured for two values of the applied voltage, under identical other experimental conditions. The temperature was kept at 35 °C

results reported in [19] by investigation of the relaxation of the surface polarization at the interface PDLC/conductive substrate. It is worth to mention that, whereas the dielectric constants of the typical LC materials are almost of the same order of magnitude, their conductivities exhibit significant variations. Generally, rigorous explanation of the dielectric response in such heterogeneous media is still under discussion. In particular, a complicating factor is the tensor nature of the dielectric permittivity of the real PDLC systems.

The EO filtering that can be achieved by PDLC exhibits a narrow band (e.g., 30 – 50 Hz full width at half maximum). This is of interest for potential applications. The band of selective EO amplitude-frequency modulation can be tuned by varying the voltage applied on the cell. Indeed, by increase of the voltage, the band is shifted towards the lower frequencies (Fig. 3). Furthermore, the amplitude-frequency modulation of transmitted light by the examined PDLC films exhibits a very good frequency and amplitude stability.

4. Conclusions. In conclusion, we have studied PDLC films with a thickness of 10 µm where single droplets of nematic LC with diameters as large as 20 µm were confined. The nematic director field in this composite material was efficiently modified by nanostructuring teflon rubbing of the glass plates of the PDLC cell. When these plates have rubbing directions orthogonal to each other, single layers of PDLCs arranged and oriented in this way exhibit a selective amplitude-frequency EO modulation well the controllable by the voltage applied.

The band-like EO modulation within a certain frequency range is attributed to the effect of screening of the external electric field driving the PDLC. The selective band-pass filtering may be useful for applications based on various schemes by exploiting an efficient EO modulation by PDLC in the infrasound frequency range which is of interest for military, geo-acoustic and bio-medical monitoring.

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