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Dielectric and flexoelectric oscillations in PDLC studied by flexoelectric spectroscopy and laser light diffraction

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Nematic droplets of variable size of E7 were dispersed in a photopolymer NOA65 matrix by the method of UV photopolymerization-induced phase separation. Dielectric and flexoelectric oscillation of the director orientation in the droplets were excited by an ac driving voltage in the range 1Hz to 3 kHz. Both thelinear and quadratic electro-optical response of the PDLC films were studied by the flexoelectric spectroscopy method and by laser light diffraction. The temperature and voltage dependence of the 1st and 2nd harmonic electro-optic spectra (amplitude and phase of the transmitted light vs frequency) were obtained, and strikingly deep minima in all spectra were found. These minima were interpreted as resulted from a spatial filtering (i.e. selective diffraction) of the time-modulated components of the transmitted light.

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1. Introduction

Polymer dispersed liquid crystal (PDLC) films, consisting of micron sized droplets of a nematic liquid crystal dispersed in a polymer binder, are of particular current interest for projection television, direct view flexible displays and switchable window applications [1]. Understanding the optical response of these smart optical materials at higher frequencies is important from both a fundamental and an applied point of view.

2. Materials and methods

Polymer: NOA65 (Norland Optical Adhesive 65, Norland Products Inc., New Brunswick, N.J.) is a clear, colorless, liquid photopolymer. It is cured by UV light, with a maximum absorption within the range 350-380 nm. The refractive index of the cured polymer is n = 1.524.

Liquid crystal: The eutectic cyanobiphenyl mixture E7 (BDH Limited Pool, England) was used, showing a nematic-to-isotropic (N-I) transition at 59-60°C, and a smectic-to-nematic transition below –20°C. The refractive indices of the LC material at a 633 nm for extraordinary and ordinary light are 1.737 and 1.5185, respectively [2]; the dielectric anisotropy is positive, $\varepsilon_{II} = 19$ and $\varepsilon_{\perp} = 5.2$ at 1 kHz frequency and room temperature [3]. Therefore, the random-like LC orientation in the droplets in the absence of an applied field creates a refractive index contrast, giving rise to intense light scattering. However, this scattering is field-reducible, and when the a.c. field is of sufficiently low frequency, it is also time-modulated (see below).

Samples: The PDLC samples were prepared by a photopolymerization-induced phase separation (PIPS) technique. Predetermined mixtures of NOA65 and E7 in the ratio 50:50 wt % were heated to 60°C in an isotropic phase to achieve homogeneous mixing. The blended compounds were sandwiched between two In-Sn oxide (ITO) coated glass plates and set to a thickness of 25 μm or 5 μm using polymer spacers. The UV curing was performed by either homogeneous irradiation of the whole sample area, or by a lateral gradient of the UV intensity, thereby producing a gradient in droplet size.

Experimental set-up: The technique for exciting and recording the dielectro-optic and flexoelectro-optic response of the PDLC films has been described previously for continuous nematic layers [4, 5]. The frequency dependence of the 1st and 2nd harmonics of time-modulated He-Ne laser light of normal incidence transmitted through the film (forward scattered) was registered by a photodiode and fed to a lock-in amplifier (SR530, Stanford Research Systems). The lock-in in-built function generator was interfaced to a computer, thus providing a frequency sweep of variable range and rate. The amplified generator output was applied to the sample by means of ITO electrodes.

Diffraction patterns were obtained by a 628 nm He-Ne laser (Melles-Griot) on a black screen placed 50 cm away from the sample, and photo images at different voltages were taken by a HP 735 digital camera. The spatial distributions of the 1st and 2nd harmonics were obtained during a scan along the equatorial section of a diffraction pattern, by the photodiode being fixed to a translator.

3. Results

MThe microscopic appearance of a PDLC film in non-polarized light is shown in Fig. 1. The superimposed objective scale permits one to determine a typical droplet size between 10 and 20 μm.

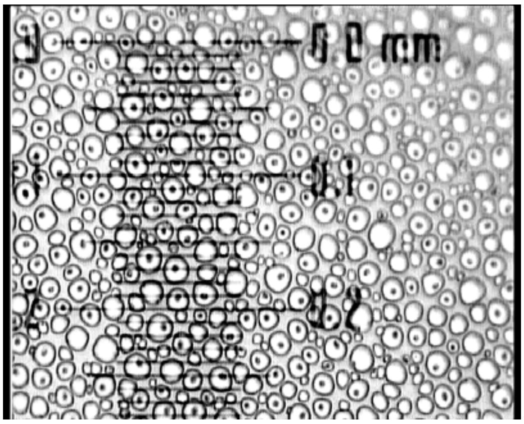

Fig. 1. Microscopic picture of a PDLC with a typical droplet size between 10 and 20 μm.

The frequency spectrum of the amplitude and phase of the 1st and 2nd harmonics of the time modulated transmitted light is shown in Figs. 2 and 3. The dielectric oscillations (2nd harmonic) display a single deep minimum, which depends on the amplitude of driving voltage and shifts towards lower frequencies from roughly 40 Hz to roughly 10 Hz upon increasing the voltage from 15 to 25 V_{rms}. It also shifts with temperature (not shown in this paper). At the minima points, the phase jumps by almost 180°.

In contrast, the flexoelectric oscillations (1st harmonic) display 2 minima in the vicinity of 100 and 1000 Hz. These minima are shifted towards higher frequencies with increasing driving voltage. The jumps in phase were less than 180° in this case, and were less steep when the minimum is shallower.

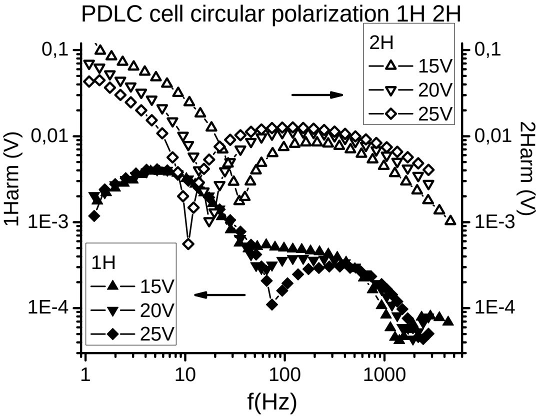

Fig. 2. Frequency spectrum of the amplitude of the 1st(full symbols) and 2nd(hollow symbols) harmonics of circular polarized light transmitted through the PDLC sample at various applied voltages.

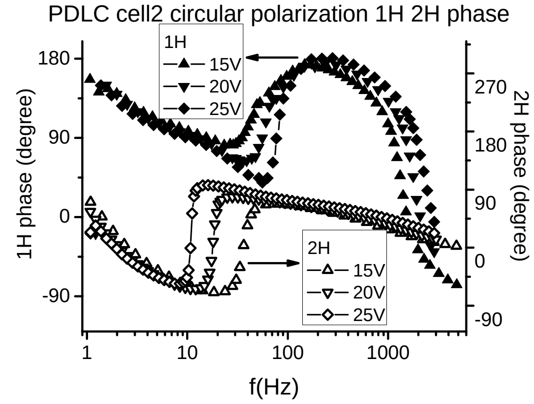

Fig. 3. Frequency spectrum of the phase of the 1st (full symbols) and 2nd(hollow symbols) harmonics of circular polarized light transmitted through the PDLC sample at various applied voltages.

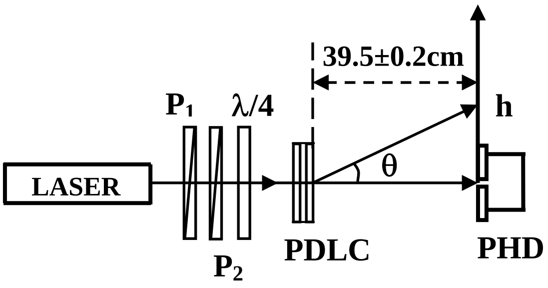

Fig. 4. Set-up for laser light diffraction measurements.

The polarization of the incident beam was fixed to a circular one by a polarizer (P_1) and a quarter wave plate (λ/4). The aperture of the photodiode was reduced by a pinhole of 2 mm diameter. The diode was fixed to a translator of 100 μm resolution.

The set up for diffraction measurements is shown in Fig. 4 and some representative diffraction pattern are compared in Fig. 5. At zero voltage, the pattern displayed just one diffraction maximum, while with increasing voltage the pattern was spread out to higher angles, and in many cases a second diffraction ring was clearly defined (cf. Fig. 7). At still higher voltages, the pattern again collapsed to a single ring.

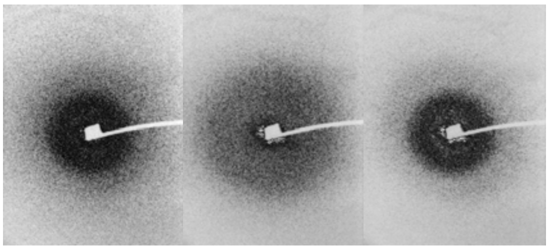

Fig. 5. Diffraction patterns (negatives) at different applied voltages, from left to right: 0 V_{rms}, 3.4V_{rms} and 50 V_{rms}. The driving frequency is 75 Hz, giving a minimum of the 1st harmonic at 15 V_{rms}.

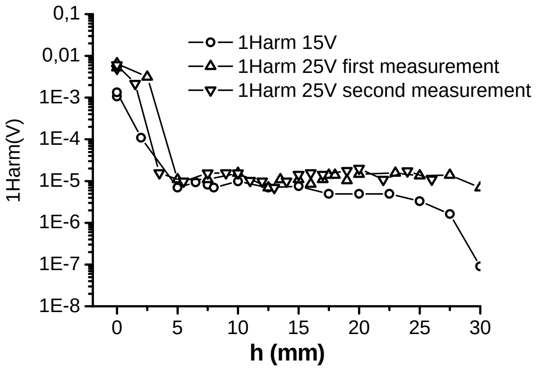

Fig. 6. Spatial distribution over the diffraction pattern of the 1^{st} harmonic excited by 15Vrms at $f_{min(2Harm)}$ = 29.4 Hz and by 25 Vrms at $f_{min(2Harm)}$ = 8.8 Hz, vs. the translation of the photodiode, h(mm).

This time-averaged picture of scattering was complemented by a detailed study of the spatial distribution of the 1^{st} and 2^{nd} harmonics across the diffraction picture using a photodiode fixed to a translator (Figs. 6 and 7). Two successive measurements were performed with a 25 V_{rms} driving voltage after a time interval of a few days. The frequencies at which the lock-in was set were chosen to equal the corresponding minima of the amplitude spectrum of the 2^{nd} harmonic, i.e. 29.4 Hz at $15V_{rms}$ and 8.8 Hz at $25V_{rms}$ for that particular sample. The formation of a double ring structure is clearly visible in the spatial distribution of the 2^{nd} harmonic (Fig. 7), and not visible for the 1^{st} harmonic (Fig. 6). Thus, this type of measurement provided us with additional dynamic information for our system, important for the understanding of the whole picture of the scattering.

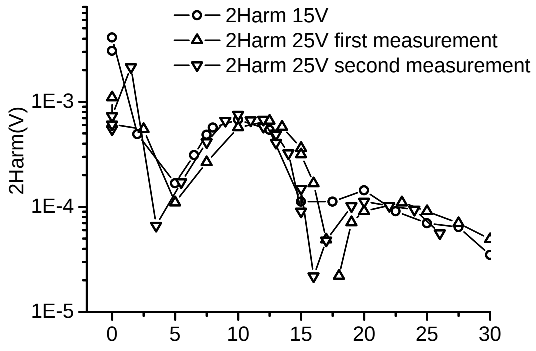

Fig. 7. Spatial distribution over the diffraction pattern of the 2^{nd} harmonic excited by 15Vrms at $f_{min(2Harm)}$ = 29.4 Hz and by 25 Vrms at $f_{min(2Harm)}$ = 8.8 Hz, vs. the translation of photodiode, h(mm).

4. Discussion and conclusions

For the first time, minima in the frequency spectrum of the 1^{st} (flexoelectrically conditioned) harmonic of the transmitted light were reported by us previously [6]. Their temperature dependence and the dependence on the droplet size were elucidated [7]. Depending on the polydispersity of the PDLC, the minimum could be deeper or shallower (smeared).

Here, we report for the first time deep (more than 10 dB) minima in the 2^{nd} (dielectrically conditioned) harmonics, along with the novel observation that the minimum frequency of both harmonics is dependent on the amplitude of the driving field.

Rigorous theoretical treatment of the problem is hindered for several reasons [8]. The first is the randomness of orientation: the initial director field within a droplet is nonhomogeneous, but rather radial or bipolar; therefore some regions of the same droplet will experience a Freedericksz transition, while others will be dielectrically stable. Our droplets are predominantly radial (judging from the presence of a central defect in many of them, see Fig. 1). Then, a published numerical solution for the director configuration in radial droplets (Figs. 1 and 2 in [9]) demonstrates that under intermediate fields, due to the dielectric torque, a central spherical part of the droplet takes an orientation that is roughly orthogonal to the remaining spherical shell. Therefore, a new refractive index contrast will be formed and the (roughly) two times smaller central region will give rise to a second, twice as broad, diffraction maximum. When the whole droplet becomes uniaxially oriented, this second maximum will disappear, in accordance with our findings.

Furthermore, the time-dependent solution of the Freedericksz problem in planar layers (Eq. 6.42 in [10]) shows that above the Freedericksz threshold small perturbations of planar orientation are exponentially growing, but at the same time periodically vibrating with a doubled field frequency around a certain (finite) value. Thus, the light scattered by the vibrating sphere (or its central part) will be time-modulated (2^{nd} harmonic) and so will the light scattered to the first and to the second diffraction maxima. This is fully in accordance with our findings (see Fig. 7).

However, when the time-modulated component of the transmitted light is maximally deflected, its intensity in the forward scattering will naturally be diminished; thus the appearance of a minimum becomes understandable. A remote analogy can be drawn with the acousto-optic tunable notch filter based on a vibrating optical fiber [11]. Moreover, since the size of the central reoriented spherical portion of a droplet is field-dependent, so will be the minimum frequency: the larger such a part (at higher fields), the smaller the frequency (due to the larger viscosity damping of partial reorientations).

The exact frequency of this minimum will be a complicated function of viscoelastic nematic properties, droplet size and temperature.

Final remarks about the 1^{st} harmonic: flexoelectric oscillations of homogeneous layers are surface-driven rather than bulk-driven [4, 5]. However in droplets, due to both electric and director field inhomogeneity, a bulk contribution of the gradient flexoeffect is rather probable. Thus, the spatial distribution of the 1^{st} harmonic director oscillations within a droplet would be fairly homogeneous. As a result, the spatial distribution of the 1st harmonic in the scattered light would be fairly homogeneous, although frequency dependent, again in accordance with our observations (Fig. 6).

The electrically tunable notch filter performance of our PDLC system could find some interesting applications in modern optoelectronic devices operating in the infrasound frequency range, where the realization of such filters is notoriously difficult using standard electronic components.

Acknowledgements

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