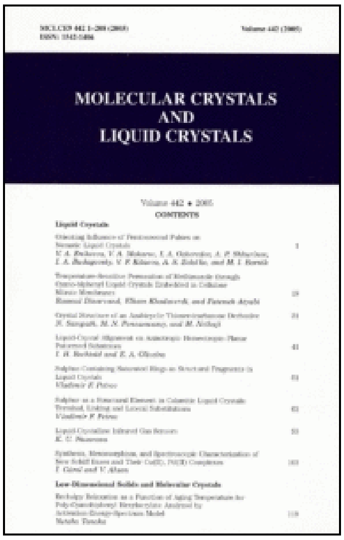

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Single-Layered PDLC for Diffractive Optics

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Single-Layered PDLC for Diffractive Optics

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The electrically and spatially controllable coherent light diffraction by single-layered polymer-dispersed liquid crystal (PDLC) films is studied. Particularly, a wedge-formed thin film of E7/NOA65 microscale PDLC is examined. The PDLC single layer of thickness varying from a few micrometers to 25 μm contains liquid crystal (LC) droplets with double-truncated spherical shapes and a linear-gradient size distribution along the film length. The single-layer arrangement and compact packing of the micrometer-sized PDLC structure render the ellectrically-commanded coherent light diffraction controlled by LC/polymer interface.

Keywords Coherent light diffraction; gradient layers; polymer-dispersed liquid crystals (PDLCs); single layers; thin films

PACS 42.25.Fx; 42.70.Df; 61.30.Gd; 61.30.Pq; 78.66.Qn

1. Introduction

Thin films of polymer-dispersed liquid crystals (PDLCs) are very attractive for variety of electro-optical (EO) applications [1–6]. Structures based on PDLCs are currently of growing interest due to light-control applications, other than the simple light-scattering devices. Particularly, the PDLCs have been applied in optical engineering of diffractive structures and devices, where the electrically-controllabe diffraction of the incident light is exploited [7–10].

As known, the PDLC EO devices operate through the change in the refractive index of the dispersed liquid crystal (LC) upon applying an alternating current (AC) electric field. By that, the gradient distribution of LC droplet size in PDLC systems provides the opportunity their structural and EO properties to be spatially controlled and tuned. This is certainly of importance for diffractive-optics applications, e.g., Fresnel lens, phase gratings, grating beam splitters, etc. [11,12]. Here we present a study on the electrically-commanded and spatially-controlled coherent

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light diffraction of a single-layered micrometer-sized PDLC film of interest for device applications. The PDLC films under study consist of nematic E7 droplets whose diameter reaches several tens of micrometers. The LC was dispersed in an optically transparent polymer matrix of the photo-cured polymer NOA65.

2. Experimental

The PDLC films used in our experiments were prepared by technology commonly used for making PDLC devices [1–4,13,14]. Briefly, a wedge-shape cell was comprised of a 25-μm-thick Mylar spacer and two 1-mm-thick 17.5 mm × 17.5 mm plane-parallel glass plates ($n = 1.5170$ at 633 nm) with inner surfaces coated with a thin (~50 nm) conductive layer of indium-tin-oxide (ITO). A 50:50 wt.% mixture of UV-curable NOA65 (Norland Optical Adhesive) monomer and the LC E7 (Merck) was filed in the cell. At room temperature, the LC E7 exhibits a nematic phase and is characterized by ordinary and extraordinary refractive indices $n_0 = 1.5185$ and $n_e = 1.737$, respectively ($\lambda = 633$ nm and at 20°C) [15]. To prepare a droplet-gradient PDLC wedge-formed single layer, the LC/prepolymer mixture was subjected to a very slow UV-curable polymerization-induced phase separation (PIPS) upon irradiation with a high-power UV laser. The photopolymerization was performed at room temperature with nanosecond pulses [16,17].

A morphology of well separated LC droplets was evidenced for the examined PDLC film by means of polarizing optical microscope (Zeiss NU-2 Universal Research Microscope) with PDLC cell placed between crossed polarizers. The images were obtained by a Hitachi VK-C150ED video camera and captured by a computer. For EO and diffraction measurements, the PDLC cell was mounted on a micro-manipulating translation stage. A circularly polarized TEM_{00} laser light from a NG HN-40 He-Ne laser (wavelength $\lambda = 632.8$ nm) was used, having a beam diameter of ~0.6 mm (at intensity level $1/e^2$) with Gaussian distribution in intensity and a divergence less than 0.8 mrad. The laser beam was directed normally to the PDLC cell. The laser power incident on the PDLC film was 3.8 mW.

The light diffracted from the PDLC single layer was measured by a photodiode and computer-controlled lock-in amplifier (SR830, Stanford Research Systems). A sinusoidal voltage from the lock-in instrument was applied to the ITO electrodes of the PDLC cell. For the voltage-dependent light intensity measurements, AC voltage at a frequency of 350 Hz was used, and the laser beam was chopped at 90 Hz. The photodiode was combined with an aperture (pinhole diameter 1 mm). The intensity distribution (profile) of the light diffracted from the PDLC film was recorded at a distance of 76 cm from the PDLC cell by translating photodiode perpendicularly to the direction of the incident laser beam. The spatial resolution of the scan was 100 μm.

For integral measurements of the intensity of entire diffraction ring, additional apertures (diaphragm and disk with adjustable diameters) were used to spatially separate the diffracted light, which was subsequently focused on the photodiode by a lens (f = 8 cm) within the pinhole aperture. Diffraction pictures were taken on a distant screen. The temporal measurements were performed by Tektronix TDS1002B digital storage oscilloscope. In this case, the lock-in build-in function generator supplies to the PDLC cell 1 kHz sinusoidal-frequency packets at repetition frequency 6 Hz. All experiments were carried out at room temperature.

3. Results and Discussion

A detailed study on the morphology of the micrometer-sized single-layered E7/NOA65 PDLC material and how the morphology factors repercute on characteristics, such as diffraction profile, background scattering and diffraction efficiency, was presented in [17]. It should be emphasized that highly regular and well-dispersed LC droplets with a very defined and constant shape take place on the PDLC film. Having a gradient along the film length, the droplet size varies from a few to several tens of micrometers. The LC droplets exhibit double-truncated spherical shapes whose average diameter varies as twice the cell gap [17]. Thus, by simple translation of the PDLC film across the light beam, one can use a layer region of desirable droplet size. The droplet size gradient can be neglected within a range comparable to the probe laser beam diameter. At a low microscope resolution (e.g., by a magnification of 4×), a compact packing of neighboring droplets within the polymer matrix is clearly visualized (Fig. 1). The distribution of the LC droplets on such a scale resembles a regular structure of well-ordered two-dimensional array, e.g., the highly monodisperse and long-range ordered LC/polymer systems [18,19].

Polarizing microscope images of the PDLC composite display a texture that corresponds to oriented LC, separated by isotropic polymer matrix. The texture exhibits a parallel alignment for nematic E7 that is consistent with the tangential boundary condition for E7 molecules imposed from the cured NOA65 polymer in the E7/NOA65 system [12]. Depending on the orientation of the droplet symmetry axis, droplets with a baseball or cross pattern take place in the PDLC film between crossed polarizers (Fig. 2b). According to the observed textures, the microscale PDLC studied here can be regarded as predominantly bipolar droplets, with the nematic director aligned between two disclination points positioned at opposite poles of the droplets [2]. Furthermore, the polarization microscopy of the examined film reveals an orientation of the droplet polar axes (piercing both disclinations) in the

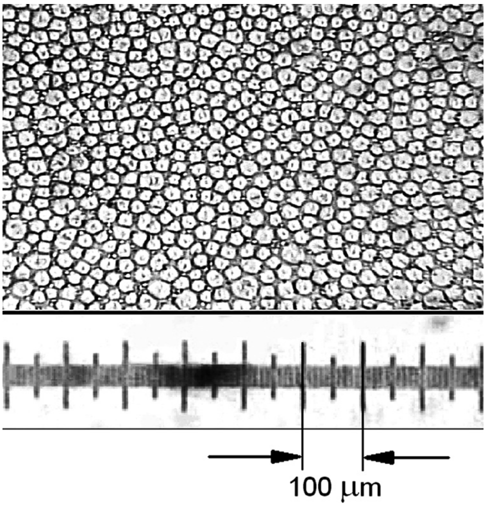
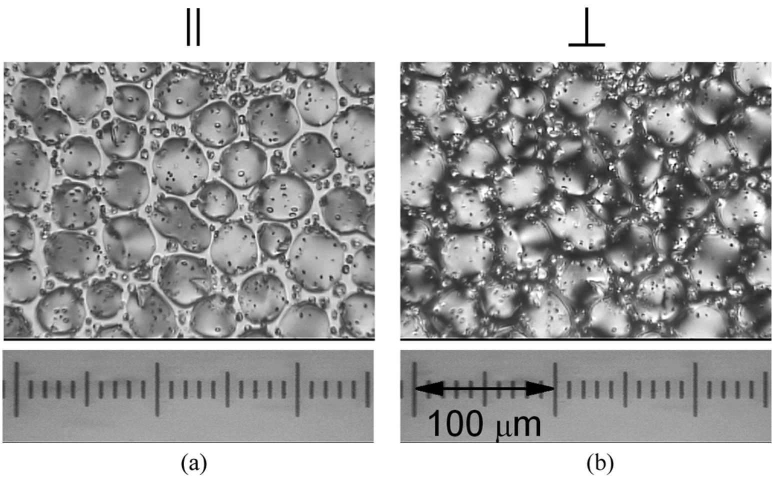


Figure 1. Long-range ordering of nearly identical and close-packed LC droplets in the microscale PDLC single layer studied here. The optical microscope image corresponds to a film thickness $\delta = 18.4\,\mu\text{m}$.



(a) (b)

Figure 2. Optical microscope images of the single-layered PDLC film at thickness of 18 μm. The micrographs are taken under parallel (a) and crossed (b) polarizers.

PDLC layer plane. Since the droplets are flattened out (their equatorial size (diameter) is about twice than the transversal one), this droplet shape anisotropy favors the droplet polar axis orientation parallel to the film layer plane. Because the droplet shape anisotropy results in an optical anisotropy, one can expect an efficient EO control by both birefringence and optical phase shift, induced in such a PDLC structure through the electric field dependent effective refraction index, $n_{eff}(E)$, of the dispersed nematic LC.

Figure 3 illustrates the optical diffraction obtained from the examined single-layered PDLC film upon illumination with a coherent light beam from a He-Ne laser. Well-resolved bright hexagonal spots were observed in the diffraction picture (Fig. 3a). As found in [17], these diffraction peaks are defined by film morphology and optical interference. The latter is enhanced by the hexagonal order of the PDLC structure of closely-packed LC droplets dispersed in the transparent polymer matrix. Being depending on the droplet organization in the film region probed, the intensity of the diffraction peaks can be spatially controlled by means of LC droplet size gradient [17]. On the other hand, at a fixed location on the PDLC film, the light intensity distribution within the complex diffraction pattern can be commanded by the applied AC voltage. Such a spatial light energy redistribution between the diffraction pattern is of a practical interest.

To analyze the electrically-commanded diffractive properties of the examined single-layered PDLC film, the intensity change of the diffraction peaks with the applied AC voltage was measured under identical conditions. As an example, Figure 4 plots the voltage-dependent light intensities of diffraction peaks (A), (B) and (D) as designated in Figure 3(b). The position of the probe laser beam on the PDLC film was kept constant ($\delta = 18.4\,\mu m$). Note that the photo-detector aperture of 1 mm allows the spots of the diffraction peaks to be well separated from surrounding speckle light pattern, and adequately measured. As seen in Figure 4, the intensity dependencies of transmitted and diffracted laser light are reciprocal as expected due to the competition and intensity redistribution between them.

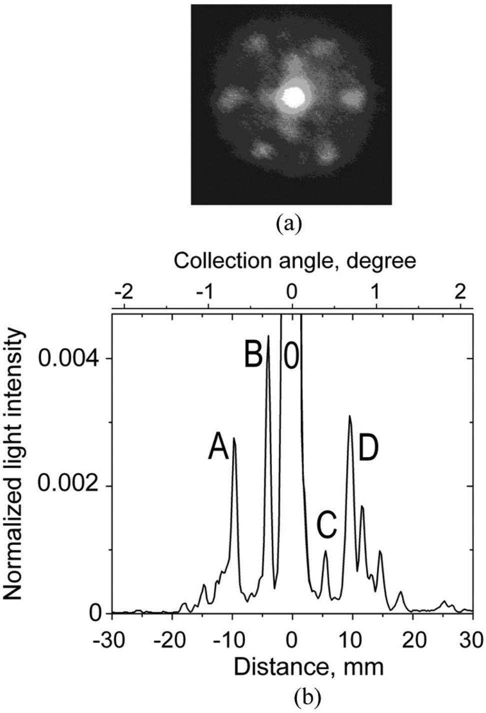


Figure 3. Diffraction picture (a) and the corresponding profile (b) taken in an equatorial plane of the diffraction pattern (a). The main diffraction features are denoted as 0, A, B, C, D. The applied voltage is 20 V_{rms}, the temperature is 20°C. The diffraction profile is normalized to the intensity of the transmitted He-Ne laser beam (0, zero-order diffraction).

Particularly, the peaks (A) and (D) are almost inversely related to the transmitted light, i.e., the zeroth order diffraction (0). Also, the temporal traces measured for the diffraction beam splitting show the same trend (Fig. 5). Further, the frequency spectra of the amplitude of the modulated light measured for the diffraction peaks provide an additional information about the intensity interplay in the EO response, both flexoelectric (the 1st harmonic) and the dielectric (the 2nd harmonic). Figure 6 represents frequency spectra taken in saturation regime (ON-state) upon 20 V_{rms} applied. The difference in the frequency-dependent dielectric response measured for the diffraction beam splitting is indicative of significant intensity redistribution between the diffraction pattern. This difference is much enhanced especially in the pre-switching regime (not shown here). More detailed analysis of interference effect on the dielectric and flexoelectric response of E7/NOA65 single-layered PDLC system will be presented separately. It should be noted that all these behaviours strongly depend on the film thickness, amplitude of applied voltage and temperature, in accordance with data previously reported for E7/NOA65 (50:50 wt.%) PDLC films [20,21].

The strong and sharp oscillations which take place at low voltage in the pre-switching region (recall Fig. 4) also exhibited an intensive energy redistribution between the transmittance and diffraction peaks, as well as between the diffraction peaks themselves. In contrast to the transmittance (0), the intensity of the diffraction

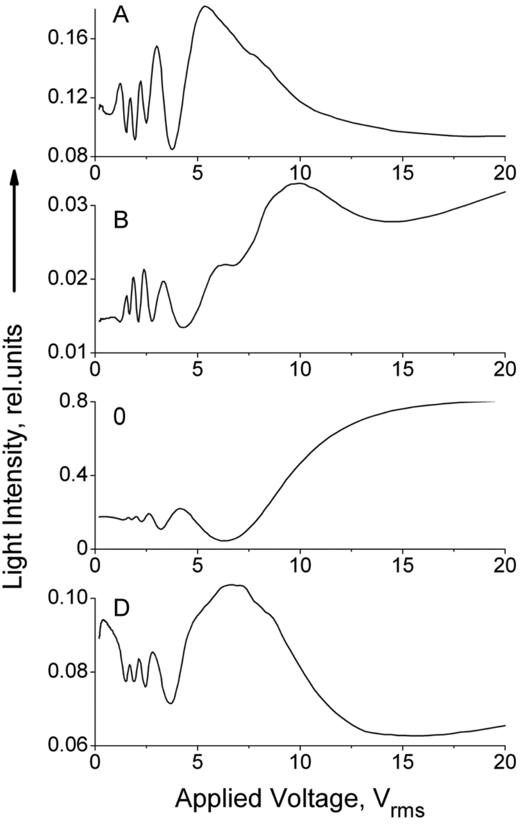


Figure 4. The voltage-dependent intensity of light ($\lambda = 632.8$ nm from He-Ne laser) diffracted from microscale PDLC single layer. The probed thickness $\delta = 18.4\,\mu$m, 20°C. The probed diffraction peaks (A), (B) and (D) are indicated in Fig. 3(b).

peaks in this region (up to 6 V_{rms} for the example shown in Fig. 4) can be significant. These observations imply that the electrically-commanded optical diffraction from single-layered PDLC film in pre-switching mode can be also practically used for light control and provide additional flexibility in the switching behaviour.

We have to point out that the intensity distribution in the diffraction pattern is very sensitive to the PDLC film thickness (δ) [17]. However, the correlation between the EO response of both coherently transmitted and diffracted light remains as described above. As established for the examined single-layered PDLC film, δ is linearly related to the average droplet diameter ($D \approx 2\delta$) [17]. The LC droplet size is very important factor since the coherent light diffraction from PDLCs is governed by the LC/polymer interface and the refractive-index difference at that interface. Actually, all structural characteristics such as the LC droplet size, shape, droplet number density, ordering and alignment, play a role in governing the elastic, dielectric, optical, etc., properties of PDLC system [2,22,23], and thereby, do influence also the optical diffraction (and scattering) from the PDLC films. In this context, the diffraction greatly depends on the orientation ordering. The LC droplets in the examined PDLC single layer possess a preferential planar orientation resulted from the flattened droplet shape. The identical droplet

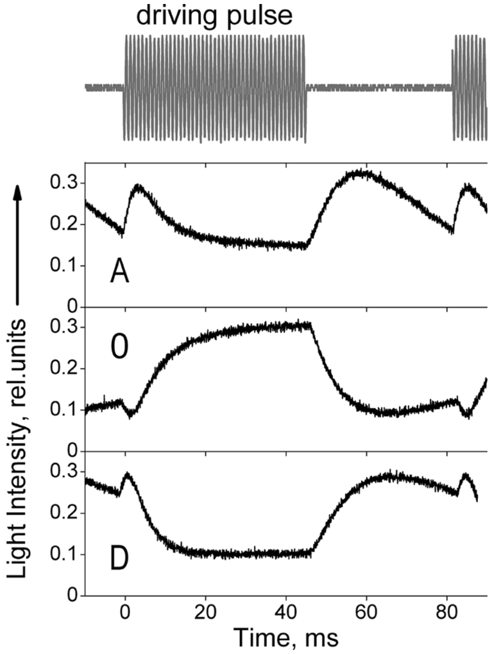


Figure 5. Oscilloscope traces measured for diffraction peaks (A) and (D) from single-layered PDLC film illuminated with He-Ne laser. For comparison, the temporal trace measured for the zeroth order diffraction (0) is also given. The film thickness $\delta = 18.4\,\mu m$ was probed, the temperature was 20°C. The pulse of the driving AC voltage is shown on the top.

characteristics, the two-dimensional arrangement and the alignment of the local optical axes of LC droplets in an ensemble probed by laser light beam whose diameter envelops the ensemble dimensions, should be of advantage by coherent interaction of single-layered PDLC film with the electro-magnetic field of the laser light wave [24–26]. Especially, the optical coherent effects are well pronounced in highly-ordered two-dimensional hexagonal-close-packed monodisperse PDLC monolayers [27,28] where the optical interference has been reported to significantly affect the electrically switched diffraction of the incident coherent light. Being not existing in the conventional PDLCs, the interference-enhanced mechanism of diffraction by closely packed PDLC two-dimensional single layers is of a practical interest.

Various factors play a role for the efficiency of the interference-enhanced diffraction. Among these are the droplet size polydispersity and the degree of order, as well as the orientational anisotropy and alignment of the PDLC film [26–30]. Further, the electrically-commanded optical phase shift induced by LC/polymer interface, is a factor that controls the brightness of the diffraction peaks. The optical phase difference is responsible for the efficient optical interference at the ensemble level. In this case, especially important is the droplet monodispersity, closely related to the hexagonal order of the LC droplets within the polymer matrix. Generally, the efficient interference-based optical diffraction by PDLCs on the microscale requires highly-ordered arrays of LC droplets with a narrow-size distribution, like monodispersed monolayers of highly-ordered two-dimensional hexagonal-close-packed arrays [27,28].

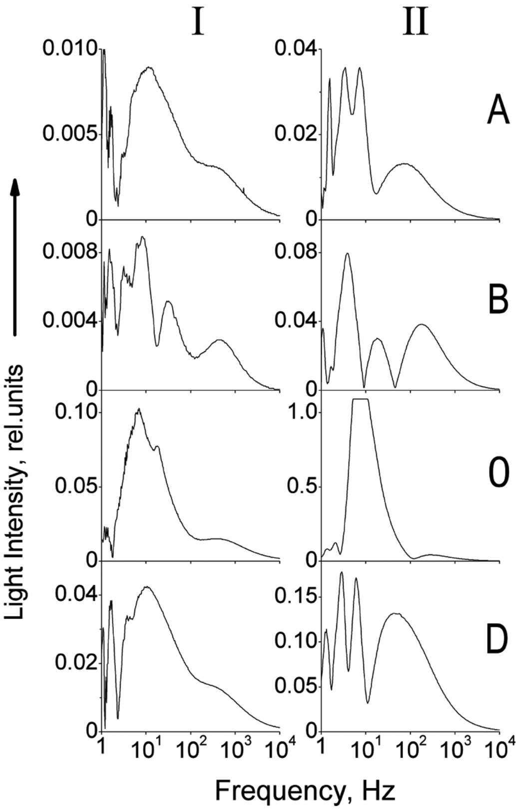


Figure 6. Frequency spectra for the 1st (I) and the 2nd (II) harmonics measured by the lock-in instrument for the diffraction peaks (A), (B) and (D), as compared to the zero-order diffracted light (0). The peaks are the same as denoted in Fig. 3(b). The voltage of the applied electric field was fixed at 20 V_{rms}, the film thickness was $\delta = 18.4\,\mu m$, the temperature was 20°C.

The interference-based spatial diffraction of coherent light propagating through PDLC film is controlled by the optical phase difference that is governed itself by the LC/polymer interface. When a coherent light beam passes through the PDLC film, the index difference between the adjacent LC-rich and polymer-rich regions induces a relative phase difference (Δ) that can be electrically commanded through the dielectric orientation of nematic director, i.e., by $n_{eff}(E)$. The electrically-produced phase retardation depends on the strength of the electric field applied, as well as on all material characteristics which govern the electrically-driven LC reorientation process, such as the microgeometry and morphology of the PDLC, its dielectric properties and the interfacial interaction between the polymer and the LC at the droplet walls [18,31–36]). At normal incidence, Δ is given by:

$$\Delta = \frac{2\pi}{\lambda}[n_{eff}(E) - n_p]\delta \tag{1}$$

where δ denotes the film thickness, $\lambda = 633\,\text{nm}$, n_{eff} is the effective refractive index of the dispersed LC, and $n_p = 1.524$ is the refractive index of the cured NOA65 polymer at 633 nm and 20°C. As known for PDLCs using a nematic phase as the dispersing LCs with positive dielectric anisotropy, the application of an electric field greater than a critical threshold (Freedericksz transition [37]) reorients the average director along the electric field, changing n_{eff}. According to Eq. (1), the phase retardation can be electrically commanded by the field-dependent index $n_{eff}(E)$, as well as spatially controlled by the film thickness δ. Thus, exploiting the double modulation effect from δ and E, one can control the diffraction output of the PDLC film.

The voltage-dependent light intensity oscillations in the pre-switching region take place also for other diffraction patterns clearly observed simultaneously with the bright hexagonal peaks. They are two concentric rings (or more correctly, disks) resulting from coherent light diffraction from LC droplet shape. As such, the well-defined second ring with a larger diameter occurs due the optical phase contrast following the electrically-induced refraction index difference between the central spherical part of the LC droplet and remaining shell layer. These diffraction patterns were previously studied in the composite E7/NO65 by optical diffraction and frequency spectra [21] and were explained in terms of the dynamics of the electric field driven nematic reorientation and by the specifics of the nematic director distribution in the confined volume within a vibrating LC droplet [2,38–41]. In the present study, the mutual intensity correlation of both diffraction rings was also evidenced. Figure 7 reports the spatial distribution of the light intensity over these diffraction patterns when the applied voltage was fixed at 20 V_{rms}. The spatial profile recorded is consistent with data in [21]. Figure 8 represents the change in the light intensity measured for the first diffraction ring (Ring I) and the second one (Ring II), as compared to the

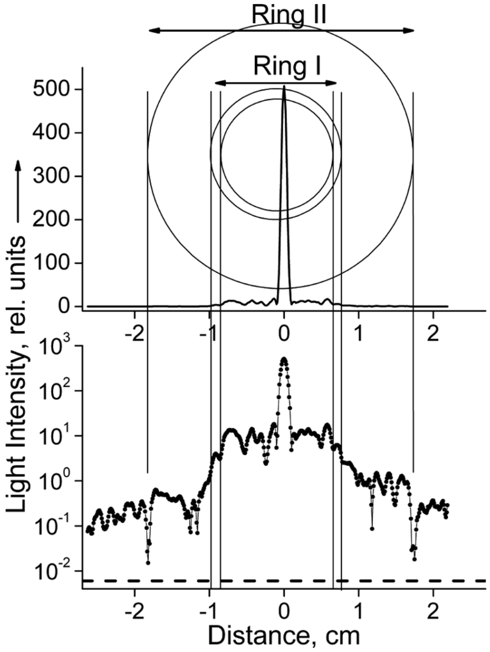


Figure 7. The spatial profile (in equatorial plane) measured for He-Ne laser light passed through the single-layered PDLC film at 20 V_{rms} ($\delta = 18.4\ \mu m$, 20°C). The dashed line indicates the sensitivity limit of the apparatus.

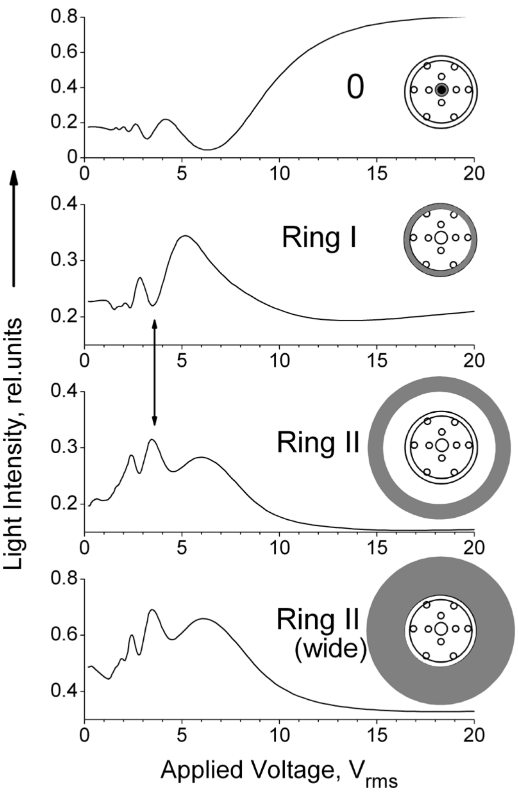


Figure 8. The voltage-dependent change in the light intensity of the diffraction rings integrally measured, as compared to the intensity of the zeroth order diffraction (0) ($\delta = 18.4\,\mu$m, 20°C, He-Ne laser). The probed diffraction patterns are drawn in grey in the schematics given in the right.

intensity of the zero-order diffraction (0). The obtained behaviors suggest an intensity redistribution between diffraction patterns. Particularly, at a certain voltage value in the pre-switching region (shown with a double arrow in Fig. 8), the intensity of Ring II is at its local maximum, whereas the intensity of Ring I is at minimum. The voltage-dependent intensity of the light output within a wide space angle corresponding to the second ring (Ring II 'wide' in Fig. 8), exhibits a behavior which is almost the same as that of the Ring II. The result indicates an optically homogeneous bulk material in the inner spherical part of the LC droplet having also an uniform director orientation, in accordance with the theory and experiment for LCs spatially confined to curved geometries [2,21,39,40,42]. Being sensitive to phase contrast, the coherent light diffraction is a delicate tool to carry out experiments to mapping the core layer inside a LC droplet when an electric field is applied to the PDLC cell (work in progress).

Conclusion

The diffraction beam splitting, namely diffraction peaks of six-fold symmetry, observed in our experiments by single-layered PDLC film illuminated with laser light, is determined by PDLC film morphology and LC droplet arrangement, and is related to the electrically produced optical phase retardation. Upon applying an electric field, a spatial redistribution of the intensity of the diffracted light can be achieved. Complemented with the spatial control by simple translation of the linear-gradient PDLC wedge film across the incident laser beam which utilizes the well-controllable and continuously tunable LC droplet size, this can further facilitate the diffractive-optic operation of single-layered organization of nearly identical and preferentially aligned LC droplets in two-dimensional microscale PDLC films.

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