Optical interference effects in microscale PDLC two-dimensional layers*

G. B. HADJICHRISTOV^{*}, Y. G. MARINOV, A. G. PETROV

Georgi Nadjakov Institute of Solid State Physics, Bulgarian Academy of Sciences, 72 Tzarigradsko Chaussee Blvd., 1784 Sofia, Bulgaria.

Optical interference effects are studied in electrically controlled microscale two-dimensional layers of polymerdispersed liquid-crystal (PDLC) bipolar droplets with a size gradient. The peculiarities observed in the transmission and diffraction of coherent light, illuminating such a PDLC thin film, are attributed to the phase retardation induced in the layer and are related to both material and structural properties of the PDLC system. Controlling the induced optical phase shift, and, thereby, the optical interference, the droplet gradient makes the microscale PDLC two-dimensional layer feasible to operate as a tunable phase-grating device.

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1. Introduction

Thin films of polymer-dispersed liquid crystals (PDLCs) are mixed soft-solid composites, very attractive for electro-optical (EO) applications [1–6]. Consisting of liquid crystal (LC) droplets dispersed within an optically transparent and isotropic polymer matrix, these smart materials have highly controllable optical and EO properties. Among the PDLC systems of special interest are the gradient PDLCs. Based on a gradient and periodic refractive index determined by the applied electric field, PDLC devices can be used as electrically switchable and tunable phase gratings [7,8] and prism gratings [9], for control of both the intensity and spatial distribution of the passed or reflected light.

Generally, efficient optical phase switching by PDLCs on the microscale requires ordered arrays of LC droplets with a narrow-size distribution, like monodispersed monolayers of highly-ordered two-dimensional hexagonalclose-packed arrays of an LC emulsion [10,11]. However, the production of the latter is very complicated. If properly formed and organized, single layers of LC micro-droplets dispersed in a transparent polymer matrix should also provide the opportunity to achieve optical phase control. Well defined sizes and shapes, as well as constant shapes and narrow size distributions of the LC droplets are necessary in this case. A PDLC material which approaches these requirements has been recently produced in our laboratory [12]. The well-controllable and continuously tunable droplet size, having a linear gradient can further facilitate the phase-switching operation of such PDLC

two-dimensional layers.

The focus of the present work is on the optical interference effects in gradient microscale PDLC twodimensional layers. They are inspected by optical transmission and diffraction measurements, **and related** to the PDLC structure. Unlike conventional PDLC films, where similar effects cannot be observed due to the random droplet distribution and broad droplet size distribution, the optical interference in single PDLC layers is well pronounced [10,11,13].

2. Experimental

A PDLC cell with a variable gap was used to study the phase retardation effects in a droplet-gradient microscale PDLC single layer. The latter was prepared from NOA-65 monomers (Norland) and E7 nematic LC (BDH). They were mixed at a 50:50 wt. % ratio and injected into a wedge-shaped cell constructed from a 25- μ m-thick Mylar spacer and two 1mm thick 17.5mm long glass plates (n = 1.5170 at 633 nm), each coated inside by a thin (~ 50 nm) conductive layer of indium tin oxide (ITO) (Fig. 1). The material was cured using a high-power

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nanosecond UV laser [12]. The morphology of the prepared PDLC thin film was characterized by optical microscopy (Zeiss NU-2 Universal Research Microscope), and the images were recorded by a Hitachi VK-C150ED video camera and computer.

The PDLC cell was mounted on a micro-manipulating translating stage. The optical interference effects in the PDLC film were studied by linearly polarized TEM₀₀ laser light from a NG HN-40 He-Ne laser, having a beam diameter of ~ 0.6 mm (at a level $1/e^2$) and a divergence less than 0.8 mrad. The laser beam was directed normally to the PDLC cell. The laser power in the PDLC film was about 3 mW. The laser beam was chopped at 90 Hz. The intensity of the light passed through the examined PDLC was detected at a distance of 76 cm from the PDLC cell by a photodiode (~ 6 mm open aperture), combined also with a pinhole aperture with a diameter of 1 mm. The signal was measured by a lock-in amplifier (SR830, Stanford Research Systems) controlled by a computer. A sinusoidal voltage with an amplitude of 0 - 20 V_{rms} at 350 Hz was applied to the ITO electrodes of the cell. The experiments were carried out at room temperature.



Fig. 1. Schematic cross-section (a) of a wedge-shape PDLC single layer, as well as the corresponding in-plane view (b). Single droplets within the PDLC film are illustrated, as well as the molecular directors.

3. Results and discussion

As observed by optical microscopy, the nematic LC droplets dispersed in the examined PDLC wedge film are organized in a single layer of thickness ranging from 4 to 25 μ m. Formatted in the PDLC wedge, the mean diameter of the LC droplets varied as approximately twice the cell gap [12]. Thereby, relatively large LC drops with dimensions up to 25 μ m (in height) × 50 μ m (in diameter) were formed. In a region comparable to the

area of the laser beam spot, the size distribution of the droplets was quite regular.

Polarizing microscope images of the PDLC texture display colored droplets separated with dark regions corresponding to the oriented liquid crystal and isotropic matrix, respectively. As known, the cured NOA65 polymer provides a planar anchoring for E7 molecules, i.e. the nematic LC adopts an alignment which is parallel to or tilted towards the polymer surface [4,14,15]. This was confirmed for the PDLC structure under study, by a polarizing microscope. In fact, the textures observed over the PDLC film exhibited droplets with a baseball pattern or an extinction cross pattern between crossed polarizers (Fig. 2), depending on the orientation of the symmetry axis of the droplet with respect to the polarizer. Generally, these textures could be modelled as bipolar droplets, with the nematic director (n) aligned, on average, between two disclination points positioned at opposite poles of the droplets [4,14,15].



Fig. 2. Polarizing optical microscope images of the examined PDLC material. The micrographs are taken: (a) without polarizers; (b) with the PDLC film between crossed polarizers.

The identical asymmetry of the droplet shapes (equal shape for all droplets in the examined PDLC film) fixes the polar axis of the droplet in the layer plane, and, thereby, the droplet shape anisotropy results in an optical anisotropy. Thus, the local optical axes of the bipolar droplets confined in a planar single layer are rather uniformly aligned in the PDLC film under consideration. Noteworthy, regions of fully aligned nematic directors were observed in some zones of these PDLCs. Since the dimensions of these zones exceed the spot size of the probing laser beam, one can expect an efficient EO control by both the birefringence and optical phase shift induced in such a PDLC structure through the electric field (E), dependent on the refraction index (n) of the dispersed nematic LC. In this way, the electrically induced optical phase can be efficiently utilized.

At normal incidence, the optical phase shift (Δ) induced by light propagation through PDLC material of thickness δ is given by:

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

where $\lambda = 633$ nm, and $n_p = 1.524$ is the refractive index of the cured polymer at 633 nm and 20° C. As known, *n* is a function of the LC droplet size [4]. Furthermore, for the PDLC two-dimensional layer examined here, the film thickness defines the droplet size and both quantities are linearly related [12]. As a result, according to Eq. (1), the phase retardation can be electrically controlled by the field-induced change n(E), as well as spatially controlled by the droplet size (the larger LC drops result in a larger phase shift). Thus, exploiting the double modulation effect from δ and *E*, one can generate a variable phase difference. In actuality, the periodical structure of alternating regions of a fixed refractive index n_p and the field-dependent index n(E) form an electric-field induced variable phase grating, based on optical interference.

The induced optical phase difference Δ results in an optical interference of the laser light propagating through the microscale PDLC single layers. In such a PDLC film, the interference effects lead to a modulation of the transmitted coherent light [13] when the voltage (*V*) of the applied electric field is below the switching value (V_{sw}). Well resolved oscillations, as well as a remarkable decline, are present in the low-voltage region of the voltage-dependent coherent transmittance of the PDLC film studied here. Fig. 3 shows a series of voltage-dependent switching curves, obtained for various δ .

As known, the mechanism responsible for the EO response of PDLC is the change of the effective birefringence with E. Macroscopically, the switching process in the nematic droplet/polymer films can be described in terms of the reorientation dynamics of the nematic droplets by an electric field. To a good approximation, the switching value of the applied voltage $V_{\rm sw}$ is given by an expression, well established for microscale PDLCs [16]. For the PDLC film under study, V_{sw} varies between 2 V and 5 V, depending on the droplet size. On the other hand, the director pattern becomes distorted above a threshold voltage value closely related to the Fréedericksz transition threshold (V_{th}) [17] defined by the elastic properties of the LC, in our case E7. At $V < V_{th}$, the uniform LC remains undistorted, and at $V > V_{th}$ the nematic is optically activated, thus reducing the effective birefringence of the PDLC film.

The oscillations in the voltage-dependent coherent transmission in the range $V_{th} < V < V_{sw}$ can be described in terms of the phase retardation change (Eq. (1)).



Fig. 3. Voltage-dependent intensity of laser light transmitted through single-layer PDLC film of various thicknesses (the ascending values follow the arrows): from 7 μ m to 11.5 μ m (a - d) and from 15.5 μ m to 23 μ m (e - j) in an interval ~ 1.5 μ m.

These oscillations, and the change in the switching curve with δ , can be more clearly seen when the intensity of the coherent light passed through the PDLC film is recorded at a narrow spatial angle. The voltage-dependent transmission, (i.e. the zero-order diffraction) measured when the detector aperture is reduced, is shown in Fig. 4 and can be compared to that of the diffracted light measured in the same manner.

As reported for highly-ordered monodispersed PDLC monolayers [11], the optical interference significantly affects the electrically switched diffraction. Being not present in conventional PDLCs, such an operation mode is of practical interest. When illuminated with coherent light, well-defined diffraction peaks were observed for the examined microscale PDLC film, in addition to the ordinary diffraction ring. This happens in the case of compact packing of the LC droplets, which resembles the regular structure of an ordered two-dimensional layer.

As seen from Fig. 4, the appearance of voltagedependent oscillations in the intensity of both coherently transmitted and diffracted light are very sensitive to δ . Especially, a strong change by varying δ takes place in the voltage-dependent interference-based diffraction in the ring pattern, as demonstrated in Fig. 4 (a, c) for $\delta = 18.4$ μ m and $\delta = 16.4 \mu$ m. Also, the behaviours for the intensity dependencies of both transmitted and diffracted laser light are reciprocal and inversely related, as expected due to the competition and intensity redistribution between them.

0.018 (a) δ = 18.4 μm 0.016 0.014 0.012 0.15 (b) 0.1 δ = 18.4 μm Light intensity, rel. units 0.05 0.02 0.012 (C) δ = 16.4 μm 0.010 0.008 0.006 0.5 0.4 (d) 0.3 δ = 16.4 μm 0.2 0.1 ż Ó 6 8 10 4 12 14 Applied voltage, V

Fig. 4. Voltage-dependent intensity of light diffracted from a gradient microscale PDLC single layer with thickness δ . The inserts illustrate the main diffraction pattern observed, and the arrows indicate the measured ones.

Considering the PDLC structure as a resultant phase grating, the number *N* of the oscillation maxima can be estimated from the condition for constructive interference, $\Delta = 2\pi N$. For example, using the values of the refractive indices $n_e = 1.737$ (extraordinary) and $n_o = 1.5185$ (ordinary) of E7 at $\lambda = 633$ nm and 20° C [18], one can calculate for $\delta = 18.4 \mu$ m that N = 6.3 interference maxima should appear in the light intensity, in good agreement with the oscillations observed.

4. Conclusions

We considered some aspects of the optical interference in the interaction of coherent light with a gradient PDLC single layer of droplets whose size reaches up to several tens of micrometers. Resulting from the optical phase retardation induced in the PDLC film, the optical interference makes such layers of dispersed LC droplets with controllable size, suitable for an efficient control of the coherent transmittance. Such thin PDLC films are feasible to operate as electrically switchable and continuously tunable interference-based phase gratings.

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*Corresponding author: georgibh@issp.bas.bg