Periodic and aperiodic liquid crystal-polymer composite structures realized via spatial light modulator direct holography

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Abstract: In this work we present the first realization and characterization of two-dimensional periodic and aperiodic POLICRYPS (Polymer Liquid Crystal Polymer Slices) structures, obtained by means of a single-beam holographic technique exploiting a high resolution spatial light modulator (SLM). A first investigation shows that the gratings, operating in the Raman Nath regime, exhibit a morphology and a electro-optical behavior that are typical of the POLICRYPS gratings realized by two-beam interference holography.

OCIS codes: (050.1970) Diffractive optics; (070.6120) Spatial light modulator; (160.5470) Polymers; (160.3710) Liquid crystals.

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1. Introduction

POLICRYPS gratings are switchable composite diffractive structures made of polymer slices alternated to uniformly aligned nematic liquid crystal (NLC) films [1, 2]. The distinctive technique exploited for their fabrication allows an almost complete phase separation between the two components. This represents a fundamental property that distinguishes these gratings from the HPDLC (Holographic Polymer Dispersed Liquid Crystals) ones, in which the phase separation process is characterized by the formation of liquid crystal droplets dispersed in the polymeric matrix [3, 4]. The droplet size in HPDLCs can be controlled by acting on different fabrication parameters [3–6], but it represents a critical feature because it affects optical and electro-optical properties of the gratings: when the droplet size is comparable with visible light wavelengths, the grating diffraction efficiency is reduced by scattering losses; when that size is smaller than the light wavelength, scattering losses are reduced but there is a noticeable increase of the amplitude of the external electric field that is requested to re-orient liquid crystal molecules inside the droplets, due to high surface tension of small droplets. POLICRYPS gratings represent an optimal solution to the illustrated issues: scattering losses are very limited (almost absent) and the switching voltages are relatively low [7]. On the other hand, while in literature several examples are reported of two and three dimensional HPDLC structures, obtained by multi-beam interference holography [8, 9] and, more recently, by alternative holographic techniques [10–12], the possibility of realizing a 2D POLICRYPS structure has been only recently explored by De Sio et al. [13]. In that case, a multi-step procedure was used to

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cure the sample with consecutive applications of the same two-beam interference pattern; this enabled the fabrication of a 2D switchable photonic crystal. Although the idea behind this result is quite smart, the procedure is not particularly simple; furthermore the range of achievable morphologies remains quite limited. It is therefore challenging to master a one-step procedure which can enable the fabrication of more exotic, two dimensional, periodic and aperiodic structures showing the typical POLICRYPS features. To achieve this result, we exploited the use of a reflective Spatial Light Modulator (SLM); the main feature of this device is that it can modify the wavefront of an impinging laser beam in such a way that any desired two dimensional light pattern can be obtained. Some of these patterns are impossible to be realized by using standard interference holography; on the contrary, they can be obtained by just reconfiguring, in real time, the SLM display. In the following, we report on the attempts done to fabricate some 2D POLICRYPS structures by utilizing the new procedure, along with related electro-optical characterization experiments. A valid example of this new scenario of possibilities is the realization of a fork grating that, at our knowledge, is impossible to be realized by using standard holographic techniques. The electro-optical characterization of all fabricated structures shows the typical behavior of the POLICRYPS ones. This promising result put the basis for a further development and optimization of the technique.

2. Method and set-up

SLMs have been extensively exploited for holographic writing purposes [12, 14, 15]. Methods that allow imaging of the desired light pattern in the object plane are usually referred to as “direct”, in contrast with “indirect” ones which allow the image reconstruction in the Fourier plane. In literature, some examples of direct imaging techniques exploitable by using SLMs are reported [12, 16]. The direct imaging method used in the present work is based on the solution proposed by Davis in 1989 [17] to solve the problem of edge-enhanced response from a binary SLM. The technique allows the realization of any generic binary pattern, characterized only by two intensity levels. The images, loaded on the SLM display, are represented by areas with pixels of the same color (white for example) alternated to areas made of a random distribution of gray levels. Monochromatic and randomized areas in the input plane (SLM) produce bright and dark areas in the output plane respectively. In our case, in order to magnify the obtained pattern to the desired size a 4f Fourier lenses system has been set-up (Fig. 1). The beam, coming from a Nd:YAG laser doubled in frequency ($\lambda = 532$ nm), is spatially filtered, enlarged and collimated by the lens L1; then, it is used to illuminate the SLM, which modifies the wavefront. Finally, the light pattern coming from the SLM is magnified and relayed to the sample plane by the two lenses L2 and L3. The magnification ratio $M = f_3/f_2$ depends on the focal lengths f2 and f3 of the lenses L2 and L3 respectively. A photo-sensitive pre-polymer syrup, made of the pre-polymer system NOA61 (70-72% wt, by Norland), the Nematic Liquid Crystal E7 (28-30% wt by Merck) and the photo-initiator Irgacure 784 (1-2% wt by BASF Resins) is used to fill in,

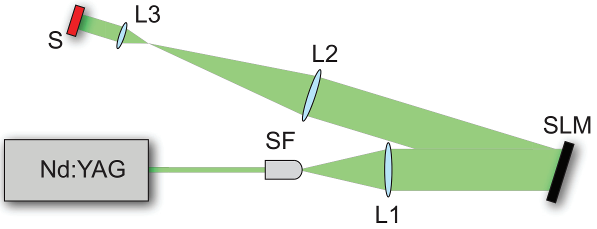

Fig. 1. 4f Fourier set-up for image reconstruction

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by capillarity, the sample cell, obtained by putting two glass substrates at a controlled distance. The choice of this mixture, formulated by Natarajan et al. [18] and very similar to the one previously used in reference [19], is the result of a dedicated investigation performed to extend the use of POLICRYPS technology also to systems exploiting visible-light curing sources; the detailed study of the attempts that brought to the determination of this chemical composition and the obtained results are reported elsewhere [20]. In our experiments, the exploitation of the POLICRYPS technology consisted in exposing the sample to the light pattern produced by the SLM at a temperature higher than the Nematic-Isotropic transition point. It is worth noting that the NLC concentration in the pre-polymer mixture has a solubility threshold of about 30%wt; when this value is overcome, NLC droplets appear during the polymerization process, thus affecting the typical POLICRYPS morphology. Polymeric branches being formed in correspondence of the bright areas of the curing pattern [21], it is convenient to adjust, a priori, the ratio between bright and dark areas as the one corresponding to the maximum achievable phase separation (70:30); the possibility of doing this kind of choice represents an innovative advantage of using a SLM for fabricating POLICRYPS structures.

3. Result and discussion

The first example exploitable both for checking the SLM functionalities and fabricating a 2D POLICRYPS structure in a one-step process is represented by the classic rectangular grid. In Figs. 2(a), 2(b) the polarized optical microscope (POM) pictures of the obtained structures and the related diffraction pattern are reported. The structure operating in the Raman-Nath regime is characterized by a periodicity of 38 μm. The sample is clearly formed by a 2D grid of NLC where the polymeric material fills the squares inside the grid; it represents the "negative" of the only 2D structure obtainable using the troublesome two-step interferometric method [13], in which the grid was made of polymeric material, while the NLC occupies the holes in the grid. It is interesting to notice that the NLC molecular director within the stripes (the NLC

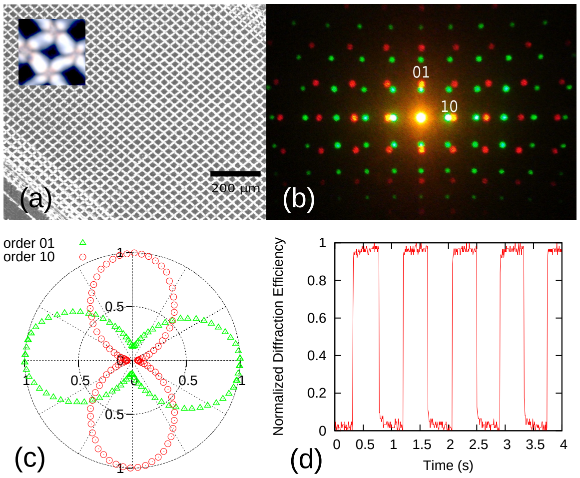

Fig. 2. (a) Optical microscopy image of a 2D POLICRYPS grid with a pitch of 38 μm. The inset shows the presence of disclinations in the region of intersection between vertical and horizontal stripes; (b) Far-field diffraction pattern for red and green light probes. (c) Polar graphs of the (1,0) and (0,1) orders; (d) Optical response of the sample to an applied (ON-OFF) electric stimulus.

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is contained in the bright grid) has a well-defined orientation, as indicated by the diffraction efficiency behavior of the (1,0) and (0,1) orders plotted as a function of the incident polarization (polar graph of Fig. 2(c)). The maximum reached diffraction efficiency, evaluated like the intensity ratio between the first order of diffraction and the sum of all the orders, is 7% for both orders. The main axes of the two polar curves of the two different polarizations are indeed perpendicular to each other, thus indicating that the NLC director in the horizontal stripes is oriented perpendicularly to the NLC director in the vertical ones. Moreover, by observing the sample at the POM, it is possible to notice disclinations located at the crossings of the grid (inset of Fig. 2(a)). In these regions, two different orientations of the NLC director intersect each other, thus creating a singularity in the local nematic director orientation. This is a clear evidence of the presence of a pure nematic phase within the NLC films of the grid. Finally, the time dependence of the diffraction efficiency on the ON-OFF application of an external electric field is shown in Fig. 2(d): a square wave, with a frequency of 700 Hz and an amplitude of 5 V/μm, is applied in successive intervals of 400 ms. The OFF state is reached in about 800 μs, while the ON state is restored in about 2 ms. It is worth noting that the OFF state corresponds to zero diffraction, that is a further evidence of the presence of a pure nematic phase in LC stripes. The demonstration that we have, in fact, realized a 2D POLICRYPS is given not only by the circumstance that the sample has been realized in the curing regime theoretically and experimentally proved for POLICRYPS structures, but also by the evidence that it shows a good morphology, characterized by a complete phase separation, low switching amplitudes and short switching times as well.

The main benefit of the new exploited technique is represented by the possibility to realize asymmetric geometries, hardly achievable by using even a multi-beam interference technique. In this framework, the fork grating is one of the most representative examples. This is a diffractive optical element whose diffraction pattern is composed by several orders, each of them being an optical vortex (Fig. 3(c)). A vortex is a light beam characterized by an helical wavefront described by the phase function $\psi_1 = \exp(iq\theta)$, where θ is the azimuthal angle of a cylindrical coordinate system (r, θ, z) around the z axis, which indicates the beam propagation direction. For points belonging to the helical axis ($r = 0$), the phase is not defined and the corresponding field amplitude is zero: an optical vortex projected on a screen appears therefore like a ring with a zero intensity region in the center (Fig. 3(d)). The integer value q that appears in the ψ_1 expression indicates the number of phase winding around the dark spot.

Optical vortexes are widely used for optical trapping of microscopic dielectric particles. Indeed, in conventional optical trapping, a Gaussian laser beam can trap those particles whose refractive index is greater than that of the surrounding medium. On the contrary, due to the gradient of intensity with the dark center, optical vortexes can trap particles whose refractive index value is both higher or lower than the one of the surrounding medium [22,23]. We reckon, therefore, that the realization of cheap devices endowed with POLICRYPS electro-optical characteristics and able to produce switchable optical vortexes represents a quite interesting application [24]. The pattern designing the fork grating has been calculated via computer generated holography [25,26]. More precisely, it has been obtained by plotting the intensity distribution I_{fork} calculated as the interference of two waves: the object beam containing the vortex phase ψ_1 and a plane-wave reference beam with phase ψ_2.

$$I_{fork} = |\psi_1 + \psi_2|^2 = |\exp(iq\theta) + \exp(ikz)|^2 = 2[1 - \cos(kz - q\theta)] \tag{1}$$

The produced image (calculated for $q = 1$) has been implemented on the SLM via the direct imaging method described in the previous section. In Fig. 3(a) a picture of the produced POLYCRIPS structure is shown. The resulting structure is, in fact, a POLICRYPS fork grating able to produce optical vortexes that can be easily switched on and off by applying an external electric field of 5 V/μm. The grating periodicity in this case is 13 μm.

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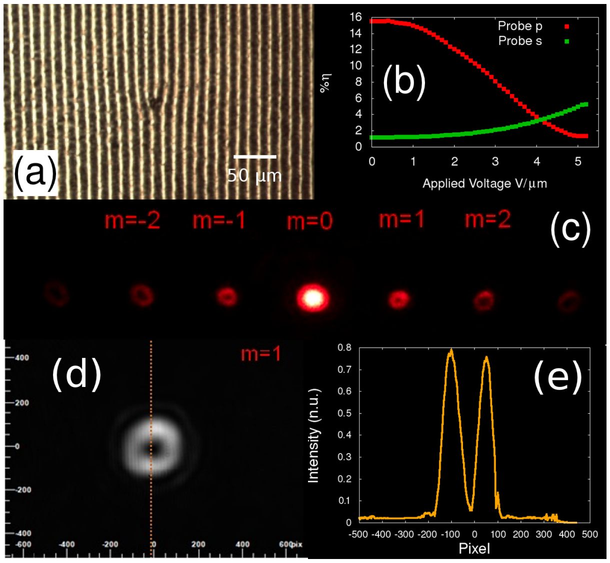

Fig. 3. (a) Optical microscopy image of a fork grating with a pitch of 13μm. (b) Switching curves for s- (red squares) and p-polarized (green squares) incoming light. (c) Far-field diffraction pattern for a red probe. (d) Beam-profiler acquisition of the first diffracted order and (e) the related cut along a radius ($\theta = \pi/2$)

A complete characterization of the first diffracted order beam is reported in the following: In Fig. 3(d) a beam profiler acquisition of the first diffracted order is presented, its intensity profile is similar to the Gaussian profile but with a zero intensity region in the center (Fig. 3(e)). The switching behavior of the first diffracted order is presented in Fig. 3(b). The green and the red squares are related respectively to the diffraction efficiency for s and p waves and show a strongly selective response in polarization. A measurement of the characteristic response times of the system to an applied electric field has been performed. Switching times necessary to turn off and on the structure are $t_{\rm off} = 800$ μs and $t_{\rm on} = 4$ ms respectively. The diffraction efficiency of the fork grating is 16%. By comparing these results with the one presented in reference [24] it is evident that the fork grating performances have been improved by the exploited POLICRYPS technique, resulting in higher diffraction efficiency and smaller switching voltages.

4. Conclusions

Standard holographic techniques can reveal quite complicate and troublesome or, eventually, not suitable when the aim is the fabrication of 2D periodic or aperiodic structures. In order to face this issue, we combined the excellent morphology and features achievable by exploiting the POLICRYPS structure with the freedom in design offered by the utilization of a SLM. In this way, we obtained a quick and low-cost one-step procedure that allows the fabrication of a large variety of high quality structures. Moreover, the technique, used can be further developed in order to realize diffractive structures operating in the Bragg regime. In fact, despite SLM limited resolution, smaller periodicities up to few μm can be achieved.

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