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On-Demand Photopatterned Twisted Nematics for Generation of Polychromatic Vector Fields

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Abstract

A simple and efficient approach to spatially addressed polychromatic modulation of light polarization using a photopatterned nematic liquid crystal film is proposed and investigated. In particular, we demonstrate linear polarization structuring of the broadband probe beam, including the formation of polarization singularities under the adiabatic propagation of linearly polarized light, which is achieved through in situ, rewritable photoalignment of nematic liquid crystal by a pump beam. This opto-optical control of polarization does not involve dynamic phase modulation and enables spatially resolved polarization patterning of broadband linearly polarized light in real time.

Keywords: structured light; vector beams; polarization singularity; polychromatic optical element; photoalignment; nematic liquid crystals



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1. Introduction

The rapid development of liquid crystalline materials and devices over the last several decades has transformed our view of optical elements, from conventional displays [1] to advanced optical modulators [2–5]. Liquid crystals (LCs) combine seemingly contradictory properties, such as fluidity and strong anisotropy of physical properties, which make them highly responsive to external fields. LC anisotropy originates from partial long-range molecular ordering, defined by a unit vector called the director, which usually coincides with the optical axis. An external electric field causes deformation of the LC director field and consequent modulation of the effective refractive index and, hence, the dynamic phase of light. Thus, the electro-optical response of LCs is the key effect underlying most of their applications [6]. In particular, this approach has proven to be effective in the development of LC-based spatial light modulators (SLMs) used for the phase control of light [4].

Typical SLMs can control not only the light phase, but also the polarization, enabling the formation of linear polarization patterns [7–9] or even full control of polarization [10]. At present, SLMs are widely employed for the generation of structured, or topological, light beams [11–13]. However, the drawback of these spatial polarization control approaches is that they rely on dynamic phase modulation, which inherently depends on the light wavelength. Consequently, all such methods are limited to a narrow spectral range of modulated light.

In contrast to the dynamic phase, which is typically utilized and modulated by SLMs, the geometric phase is associated with light polarization modulation and is independent of

the light wavelength [3,14,15]. This polychromatic property is especially important in the case of spectrally broadband light, such as femtosecond light pulses [16]. One of the most effective approaches to creating and controlling the geometric phase is the fabrication of LC films with patterned substrates.

Photoalignment [17,18], i.e., light-induced modification of the surface alignment of LC films, is considered one of the most effective approaches to adjust the LC orientation pattern and thus adapt the geometric phase of the propagating or reflecting light beams [8,9]. The LC alignment direction on a substrate depends on the polarization of the incident writing light beam: typically, for azobenzene photoalignment materials, the easy axis is aligned perpendicularly to the light polarization [19]. Alignment patterns formed by azobenzene layers can be rewritten multiple times, providing a means of constructing tunable optical elements [20–25] and electronic papers [26–28], as well as enabling optical control of LC colloids [29] through the manipulation of topological defects [30]. Broadband geometric phase modulation can also be used for polarization control when light beams modulated in photopatterned LCs interfere with each other [31].

Here, we propose a simple and robust approach for spatial–temporal polychromatic structuring of light polarization including the generation of vector fields. This method is based on the effect of adiabatic light propagation through twisted anisotropic media [32]. If the twist angle of the optical axis is much smaller than the dynamic phase shift (Mauguin limit), the polarization plane rotates in the same manner as the optical axis [33]. Therefore, when the Mauguin condition is satisfied, the polarization rotation angle is determined exclusively by the rotation angle of the LC cell director, regardless of the wavelength of light. The extraordinary and ordinary light waves follow the rotating optical axes of the LC. If the input substrate of the LC cell is uniformly aligned and the output substrate is patterned, a light beam with uniform linear polarization along the input substrate alignment will acquire the same polarization pattern as the director alignment on the output substrate [34]. Our goal is to demonstrate the feasibility of this approach for generating linear polarization patterns and for real-time polychromatic control of the probe beam polarization by combining the Mauguin regime in a nematic LC with rewritable photoalignment on one of the LC substrates.

2. Materials and Methods

We used a commercially available nematic liquid crystal E7 (Qingdao Grant Winton Int, Qingdao, China). The LC cell was composed of two glass substrates with one passive and one photoresponsive alignment layer, separated by Teflon stripes with a thickness of $L = 30 \mu\text{m}$. The phase shift between the extraordinary and ordinary light waves, $\Gamma = 2\pi L\Delta n/\lambda \approx 24\pi \gg 1$, where the light wavelength is $\lambda \sim 0.5 \mu\text{m}$ and $\Delta n \sim 0.2$, satisfies the criterion for adiabatic light propagation in the LC cell [32]. To prepare the passive alignment layer, a 1 wt % solution of polyimide (PI2525) in N-methyl-2-pyrrolidone (CAS 874-50-4, Merck Sigma-Aldrich, Darmstadt, Germany) was spin-coated onto the substrate, followed by curing in an oven at 250 °C for one hour and subsequent unidirectional rubbing. As the photoalignment material, poly[1-[4-(3-carboxy-4-hydroxy-phenylazo)benzenesulfonamido]-1,2-ethanediyl, sodium salt] (PAzo, CAS 219957-04-7, Merck Sigma-Aldrich) was used [35]. A 0.5 wt % solution of PAzo in N,N-dimethylformamide was spin-coated, followed by curing in an oven at 100 °C. Before filling the LC cell, it was illuminated from the side of the PAzo-coated substrate for 10 min with an ultraviolet light-emitting diode, with a luminescence peak at $\lambda \approx 370 \text{ nm}$ close to the absorption maximum of the photoalignment material, and an optical power of $P \approx 30 \text{ mW}$, through a linear polarizer with an axis oriented perpendicular to the polyimide rubbing direction. The liquid crystal was then filled into the LC cell by capillary

forces at room temperature. The LC film exhibited a uniform planar alignment because the PAzo photoalignment direction is perpendicular to the polarization direction.

The optical setup consisted of a continuous-wave semiconductor laser (OBIS LX, Coherent, Saxonburg, PA, USA) operating at a wavelength of $\lambda_{pump} = 473$ nm, used as a pump beam for manipulation of the LC photoalignment (Laser 1, Figure 1). The wavelength λ_{pump} corresponds to the edge of the spectral absorption peak of the PAzo material [35], while still being efficient for photoalignment and lying within the spectral range considered safe for the spatial light modulator (SLM). The light beam passed through a $5\times$ beam expander, and the central part of the beam was used for photoalignment. The light intensity was 64 mW/cm². The angle α_{pump} of the pump beam polarization plane rotation was controlled by a combination of a commercial SLM (Meadowlark Optics, Longmont, CO, USA) placed between a pair of crossed quarter-wave plates (QWP1,2) oriented at $\pm 45^\circ$ with respect to the slow axis of the SLM.

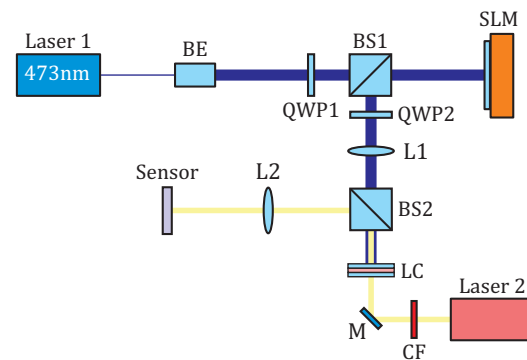


Figure 1. Experimental setup for dynamic polarization control. The pump beam ($\lambda_{pump} = 473$ nm) is used for LC photoalignment, while the broadband light from a supercontinuum laser (2) with a set of light filters serves as the probe beam. BE, BS, M, and CF are beam expander, non-polarized beam splitter, mirror, and long-pass color filter, respectively. Interference filters can be used instead of long-pass color filters.

Note that, if an SLM pixel with coordinates (l, m) , where l and m are integers corresponding to the row and column indices of the pixel, respectively, can be considered as a phase retarder with a phase shift $\Delta\Phi_{l,m}$ between extraordinary ($\Phi_e \in [0, 2\pi]$) and ordinary (Φ_o) light waves, the Jones matrix J_{total} for the pixel placed between crossed QWPs is calculated by multiplying the Jones matrix for the phase retarder J_{SLM} and the waveplates $J_{QWP}(\pm 45^\circ)$ as

$$\begin{aligned} J_{total} &= J_{QWP}(-45^\circ) \cdot J_{SLM} \cdot J_{QWP}(45^\circ) \\ &= e^{i\Delta\Phi/2} \begin{bmatrix} \cos(\Delta\Phi/2) & \sin(\Delta\Phi/2) \\ -\sin(\Delta\Phi/2) & \cos(\Delta\Phi/2) \end{bmatrix}, \end{aligned} \quad (1)$$

where the matrix J_{total} is a rotation matrix at the angle $\alpha_{pump} = \Delta\Phi_{l,m}/2$, multiplied by the dynamic phase term $\exp(i\Delta\Phi_{l,m}/2)$ [7]. To avoid redistribution of the light intensity due to diffraction caused by $\Delta\Phi_{l,m}/2$ phase modulation, the sample was placed in the image plane of the SLM screen using lens L1 in a $2f - 2f$ configuration.

Thus, the linearly polarized light beam passed through the beam expander (BE), the first quarter-wave plate (QWP1), forming circular polarization, a non-polarizing beam splitter (BS1), and the second quarter-wave plate (QWP2), forming a polarization tilt angle of $\alpha_{pump} = \Delta\Phi_{l,m}/2$. Recalling that the photoalignment direction is perpendicular to the polarization direction, the presented optical approach enables the creation of a patterned alignment with the azimuthal tilt of the easy axis [19], given by $\alpha_{dir} = \Delta\Phi_{l,m}/2 + \pi/2$, on the photoresponsive upper substrate of the LC cell.

The probe beam (Laser 2, Figure 1), with a diameter of $2w_0 = 3.4$ mm, from a super-continuum laser (SuperK EVO HP, NKT Photonics, Birkerød, Denmark), linearly polarized parallel to the polyimide rubbing direction on the bottom substrate, passed through a long-pass color filter ($\lambda_f > 585$ nm), the LC sample, and a non-polarizing beam splitter (BS2). The spectrum of the probe light beam is characterized by a spectral window from 590 nm to 740 nm. Additional interference color filters were used instead of the long-pass filter if necessary. The light power of the probe beam was 5 mW without interference filters and 30–120 μ W with any of the interference filters. The polarization camera (Alkeria Celera C5S-MP, Cascina, Italy) was placed in the image plane of the sample using lens L2 in a $2f - 2f$ configuration.

The presented optical setup allowed us to investigate the formation of polarization patterns of the broadband probe light beam (or of relatively spectrally narrow beams using interference filters) under structured pump beam irradiation, which controlled the LC photoalignment.

The following procedure was used to investigate the properties of probe light polarization. The tilt angle of the long axes of the polarization ellipses (Angle of Linear Polarization, AoLP) and the degree of linear polarization (DoLP), which corresponds to the ratio of the axes of a polarization ellipse, are calculated from the intensities I_0 , I_{45} , I_{90} , and I_{135} , corresponding to the polarization axes tilted at 0° , 45° , 90° , and 135° as

$$\begin{aligned} \text{DoLP} &= \frac{\sqrt{A^2 + B^2}}{I_0 + I_{90}} \\ \text{AoLP} &= \frac{1}{2} \arctan(B, A), \end{aligned} \quad (2)$$

where

$$A \equiv I_0 - I_{90}, \quad B \equiv I_{45} - I_{135}.$$

To evaluate the similarity of AoLP distributions for theoretically calculated $\text{AoLP}_{th} = \alpha_{dir}$ and experimentally obtained AoLP_{exp} polarization patterns, the Pearson correlation coefficient was calculated as

$$C_{\text{AoLP}} = \frac{\text{cov}(\text{AoLP}_{th}, \text{AoLP}_{exp})}{\sigma(\text{AoLP}_{exp})\sigma(\text{AoLP}_{th})}, \quad (3)$$

where $\text{cov}(\text{AoLP}_{th}, \text{AoLP}_{exp})$ is the covariance between AoLP_{th} and AoLP_{exp} distributions, and $\sigma(\text{AoLP}_{th})$ and $\sigma(\text{AoLP}_{exp})$ are the standard deviations of AoLP_{th} and AoLP_{exp} , respectively, calculated over the whole image [36].

3. Results and Discussions

By rotating the polarization plane of the pump beam by a variable angle $\alpha_{pump} = \Delta\Phi/2 = Q\varphi + \pi/2$ (Figure 2a,e), where φ is the polar angle in a polar coordinate system, the resulting polarization patterns (Figure 2b,f) exhibit singularities characterized by topological defects with charge Q . For demonstration, we chose the corresponding phase distributions $\Delta\Phi$ to achieve $Q = -1/2$ and $Q = -1$.

The pump beam induces photoalignment of the upper LC substrate (Figure 2c,g). The distributions of the LC director fields on the photopatterned substrates are characterized by singularities with topological charges $Q = -1/2$ and $Q = -1$. This leads to corresponding modulation of the polarization distributions of the probe beam (Figure 2d,h). The polarization rotation of the extraordinary light wave follows the twisted director configuration when propagating in the LC bulk, which arises from the angular difference between the alignment directions on the top and bottom substrates (Figure 2c,g). The corresponding polarization distributions of the probe beams can be described as vector fields with topological charges $Q = -1/2$ and $Q = -1$. Dark lines in the intensity distributions arise from

LC defect lines in the bulk of the LC film, which appear due to the opposite signs of the LC twist on both sides of the disclination line [30]. These lines correspond to the $\pm 90^\circ$ director rotation between the alignment directions on the two LC cell substrates (Figure 2c,g).

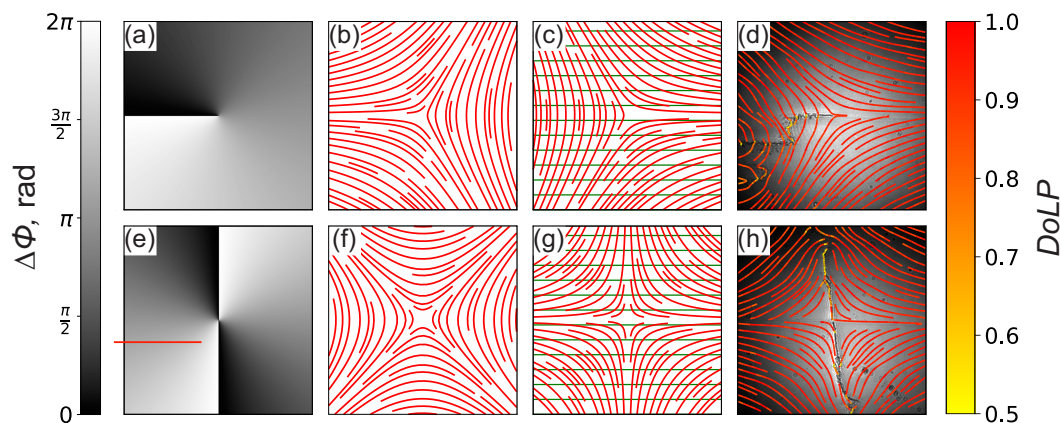


Figure 2. Light polarization modulation of a probe beam using spatially addressable photoalignment in a nematic liquid crystal: (a,e) calculated distributions of phase shift on the SLM, (b,f) corresponding polarization of the pump light, and (c,g) easy-axis alignment on the photosensitive (red lines) and passive substrates (green lines). (d,h) Experimental images of the intensity distributions of the probe light beam (with an interference filter at 530 nm) overlapping with lines visualizing the long axes of the polarization ellipses. The image size is 2.6 mm. Exposure time of the pump beam is 300 s. Color corresponds to the degree of linear polarization (DoLP).

Figure 3a–f illustrates the time-dependent formation of a polarization pattern with topological charge $Q = -1/2$ in the probe beam under sample irradiation by a pump beam having a polarization pattern with topological charge $Q = -1/2$. The dark line, which forms due to light scattering in the LC defect, appears when the polarization pattern that coincides with the easy-axis alignment on the upper substrate is nearly fully developed (Figure 3d–f). The polarization pattern is considered to be in a steady state if C_{A0LP} changes by less than 0.01 during 1 min.

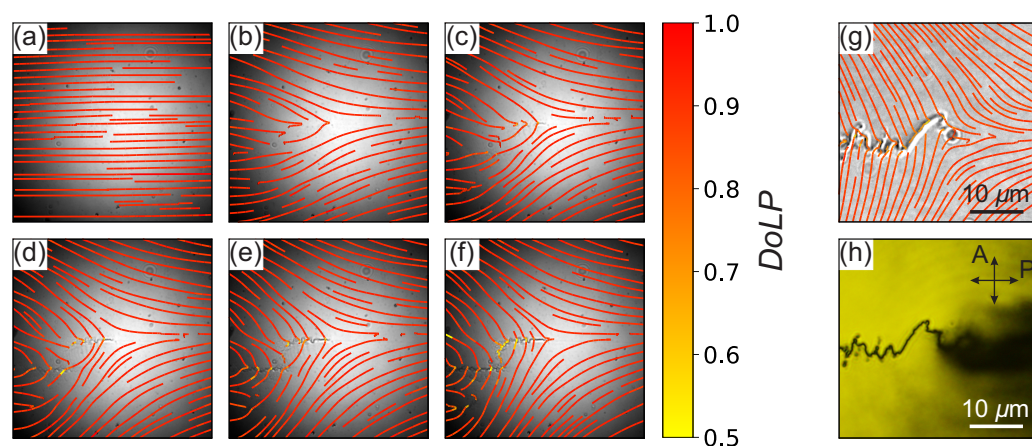


Figure 3. (a–f) Formation of a polarization singularity with topological charge $Q = -1/2$. Experimental images of the probe light intensity distributions of the broadband probe beam overlapping with lines visualizing the long axes of the polarization ellipses (the color of the lines corresponds to the degree of linear polarization, DoLP) at different times of pump light exposure: $t =$ (a) 0 s, (b) 60 s, (c) 120 s, (d) 180 s, (e) 240 s, and (f) 300 s. The image size of (a–f) is 2.6 mm. (g,h) Experimental images of the probe light intensity distributions in the central part of the LC structure after pump beam irradiation, obtained with an optical microscope equipped with (g) a polarized camera and (h) a color camera. The lines in (g) represent the long axes of the polarization ellipses.

After the photoalignment procedure described above, we investigated the optical textures and the polarization of the output light in the central part of the LC structure using a polarized optical microscope (Leica DMi8, Leica, Leica Camera AG, Wetzlar, Germany) with a $\times 20$ objective. The microscope was equipped with either a polarized camera or a color camera (Amscope MU1803, Amscope, Irvine, CA, USA). In the first case, we demonstrate the intensity pattern overlapped with the polarization distribution (Figure 3g). In the second case, the microscope texture of the sample between the crossed polarizers is shown (Figure 3h). The input light polarization of the microscope backlight, after passing through the long-pass filter, was aligned with the easy axis on the input substrate coated with a polyimide layer. The size of the inhomogeneity of the LC director and, consequently, the polarization singularity core is even smaller than the pixel size of the SLM ($\sim 10 \mu\text{m}$) due to continuous director deformations.

The alignment patterns on the photoresponsive layer of the LC cell can be changed multiple times by altering the polarization pattern of the pump beam (Figure 4). Starting from a uniform alignment on the photoresponsive layer, coinciding with the alignment on the polyimide-coated substrate, the pattern with $Q = -1/2$ was obtained using the aforementioned procedure (Figure 4a,b). Then, a uniform polarization pattern of the pump beam was used to return the sample to its initial state, although with some artifacts of the residual LC defect line (Figure 4c) due to the slight non-uniformity of the polarization. The pattern with $Q = -1$ was then obtained using the corresponding polarization pattern of the pump beam (Figure 4d). This sequence of different polarization patterns demonstrates the possibility of using the sample as a re-writable linear polarization modulator with switchable states.

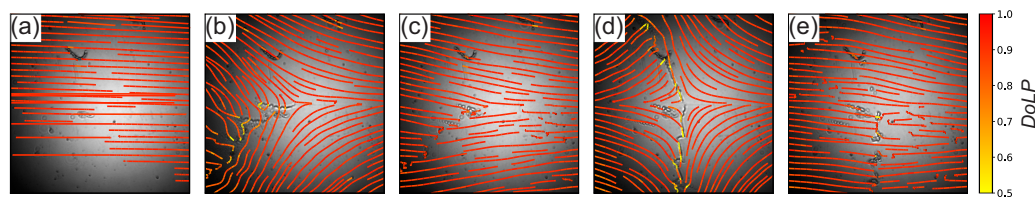


Figure 4. Experimental images of the probe light intensity distributions of the broadband probe beam overlapping with lines visualizing the long axes of the polarization ellipses (the color of the lines corresponds to the degree of linear polarization, DoLP). The image size is 2.6 mm. The pump light, with consequently changed (a–d) polarization patterns with singularities $Q =$ (b) $-1/2$, (d) -1 and without singularities (a,c,e), was used. The exposure time for each pattern is $t =$ (a) 0 min, (b) 7 min, (c) 7 min, (d) 12 min, and (e) 20 min.

To ensure additionally that the polarization patterns shown in Figure 4b,d are the same for any light wavelength, we used the interference filters at 495 nm, 530 nm, 550 nm, 574 nm, 600 nm, and 641 nm, where the full width at half maximum is ≈ 10 nm, and calculated the correlation coefficients C_{AoLP} and DoLP averaged over the region of interest. The correlation coefficient for the tilt angle distribution of the long axes of the polarization ellipses was found to be $C_{AoLP} = 0.92 \pm 0.02$ for both $Q = -1/2$ and $Q = -1$. At the same time, the degree of linear polarization averaged over the region of interest is $DoLP = 0.93 \pm 0.02$ for both $Q = -1/2$ and $Q = -1$, for all wavelengths. The weak depolarization effect can be explained by light scattering in the region of the LC defect.

4. Conclusions

An opto-optical approach to generating linear polarization patterns, including polarization singularities of various topological charges, in a polychromatic light fields is demonstrated. Spatially inhomogeneous rotation of the polarization plane is provided by adiabatic light propagation in a twisted nematic LC, oriented by patterns written in

a photosensitive layer. The effect of controlling the polarization distribution is observed across a wide spectral range and is, in principle, not limited at the long-wavelength edge of the spectrum. This property is particularly important for structuring broadband light beams. Adiabatic propagation of the extraordinary wave does not involve phase modulation, thereby avoiding additional distortions of the wavefront caused by dynamic phase modulation. Qualitative analysis of the vector fields shows good agreement with the predicted patterns. However, the LC defect lines, formed in photopatterned LCs, locally affect the intensity and polarization distributions.

The results demonstrate the efficiency of in situ rewritable photoalignment of nematic liquid crystals for polychromatic, spatially addressed polarization modulation.

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