Self-induced liquid crystal q-plate by photoelectric interface activation

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ABSTRACT
Here, we report on the experimental demonstration that highly efficient self-induced spin-orbit optical vortex generation can be achieved by using standard liquid crystal materials and surface treatment agents. This is done by revisiting the recent attempt by Zolot’ko and coworkers to produce self-induced liquid crystal vortex plates using the dc electric field [I. A. Budagovsky, S. A. Shvetsov, and A. S. Zolot’ko, Mol. Cryst. Liq. Cryst. 637, 47 (2016)] that remains, so far, limited to moderate efficiencies. The phenomenon is the result of the self-back-action of light arising from the spontaneous creation of a liquid crystal topological defect. These results demonstrate photo-electric interface activation as a candidate towards the development of a self-adapted spin-orbit photonic toolbox, thus enabling agile management of the orbital angular momentum of light.

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Since the early days of a research field dedicated to the study and use of the orbital angular momentum of light, numerous strategies have been developed to manipulate the spatial degrees of freedom of a light field. In the paraxial regime, the description of the optical orbital angular momentum is commonly associated with wavefront shaping according to a pure phase factor $\exp(i\ell\phi)$, where the $\ell$ integer is the topological charge of the phase singularity and $\phi$ is the polar angle in the plane transverse to the light’s propagation direction. The development of optical elements imparting helical phase profiles to light beams is therefore a key technological requirement.

Two kinds of optical phase shapers have been designed for such a task: scalar and vectorial ones, which rely on the space-variant dynamic phase (case 1) and the geometric phase (case 2), respectively. A prototypical example of case 1 refers to refractive spiral phase plates—three-dimensional optically isotropic objects whose thickness varies linearly with the azimuthal coordinate. Since the first demonstration in 1992, fabrication technologies have emerged which now allow creating optically perfect spiral phase plates and also endowed with additional structural information related to the radial degree of freedom of light. Case 2 is based on the spin-orbit interaction of light and exploits the opposite phase shifts imparted on the two orthogonal circular polarization states under rotations of the spatial coordinates of optically anisotropic media. The first demonstration dates back 2002 (Ref. 5) and since then, robust technologies have been developed based on various solid-state materials that are either naturally or artificially birefringent (e.g., dielectrics, metals, glasses, and liquid crystal polymers). Remarkably, both dynamic and geometric phase properties can be merged into “geometrodynamic” phase optical elements.

Still, despite an indisputable level of maturity which has been reached, an agile toolbox to manage the orbital angular momentum of light temporally, spatially, and spectrally remains a significant challenge.

Tunable and reconfigurable features obviously require materials whose optical properties can be modified under external stimuli. Liquid crystals are promising spin-orbit candidates owing to their sensitivity to electrical and magnetic fields. They have been successfully applied in spectrally tunable optical vortex generators with or without surface patterning of the liquid crystal orientation. Adaptive multiple vortex masks have also been achieved. Moreover, combining spectral and spatial control capabilities into a single device provides multispectral vortex modulation, as reported in Ref. 11. Based on such prospects, the study of optical elements, endowed with self-controlled spin-orbit photonic functionalities, is an important topic to study for its scientific and technological potential.
the past few years, various situations showed a self-induced liquid crystal topological reorientation taking place under all-optical, photovoltaic, photo-assisted ac-electrical, photo-assisted dc-electrical, or photo-thermal processes. Finally, a recent work reporting on liquid crystal defects generated by the photo-activated interface between iron doped lithium niobate and liquid crystals suggests an alternative approach worth being explored. All liquid crystal orientational patterns obtained correspond to the self-written version of a so-called q-plate with a topological charge of \( q = \pm 1 \), enabling a change in orbital angular momentum associated with \( \delta \ell = \pm 2 \) depending on the incident photon helicity \( \sigma = \pm 1 \).

Importantly, two key figures of merit of such vortex generators are the purity (i.e., the fraction of the output power that is associated with an orbital angular momentum change) and the efficiency (i.e., the transmittance of the generated vortex state). To date, only the photo-assisted ac-electrical option has demonstrated self-induced vortex generation with high purity, while the efficiency remains inherently limited to the use of absorbing photoconductive crystals. In contrast, the photo-assisted dc-electrical option is based on the photoelectric interfacial effect between a thin polymer layer, coated on a glass substrate with a transparent electrode, and a liquid crystal thin film. Therefore, this option based on the surface photoresponsive effect has the potential for being simultaneously pure and efficient, despite reported record efficiencies of the order of \( \sim 30\% \).

Here, we revisit previous attempts and we experimentally demonstrate that highly efficient (\( \sim 90\% \)) self-induced spin-orbit optical vortex generation can be achieved.

In our experiments, we use a 7 µm-thick layer of a nematic liquid crystal having negative dielectric anisotropy at low frequencies, namely, the liquid crystal mixture MLC-6608 (Merck Japan) that is characterized by a dielectric anisotropy \( \Delta \varepsilon = \varepsilon_1 - \varepsilon_\perp = -4.2 \) at 1kHz frequency, a birefringence \( \delta n = n_\parallel - n_\perp = 0.08 \) at a wavelength of 589 nm, and a bend elastic constant \( K_0 = 181 \text{pN} \), all given at 20 °C temperature. The subscripts \((\parallel, \perp)\) refer to the direction parallel and perpendicular to the director, i.e., the headless unit vector that defines the average molecular orientation. The nematic film is sandwiched between two transparent glass substrates provided with indium tin oxide (ITO) electrodes on which a thin layer of a standard polyimide (PI) alignment material is spin-coated (SE-1211, from Nissan) which provides perpendicular boundary conditions for the director field. A sketch of the sample and of the setup is shown in Fig. 1, where the laser beam is a continuous-wave fundamental Gaussian beam at a wavelength of 332 nm.

Highly efficient vortex generation implies that the sample behaves everywhere as a half-wave plate. Namely, the birefringent phase retardation, \( \Delta \), is expected to satisfy the condition \( \Delta = \pi \). It corresponds to full polarization conversion of an incident circular polarization state with helicity \( \sigma \) into the orthogonal polarization state with helicity \( -\sigma \). The first task is therefore to identify the required dc voltage. This can be done by preparing a circularly polarized incident beam with helicity \( \sigma \). In our case, it had a waist radius of 1.3 mm and a power of 0.25 mW. Then, we evaluate the ratio

\[
\eta = \frac{P_{\sigma}}{P_{\sigma} + P_{-\sigma}},
\]

where \( P_{\sigma} \) is the power of the circularly polarized component of the output beam with helicity \( \pm \sigma \). The results are reported in Fig. 2 where the dependence of \( \eta \) as a function of the applied voltage \( V \) is shown for both dc and ac situations for comparison. In the ac case, the liquid crystal reorientation is detected above 2.2 V in agreement with the electrical reorientation threshold voltage given by \( V_{ac} = \pi \sqrt{K_0/\varepsilon_0 (\delta n)} = 2.19 \text{V} \), and that after half-wave retardation condition corresponds to \( V_{ac}^r = 2.63 \text{V} \). In the dc case, the whole set of measurements is performed over the duration of three minutes, hence minimizing the back-action of light at the PI/nematic interface, connected to the positive pole of the electrical power supply, as confirmed by the results in Fig. 3(a). The fact that the reorientation takes place above an applied voltage that is slightly larger than for the ac case, and that \( V_{ac}^r > V_{dc}^r \) in the framework of our transient measurement protocol, can be understood by both partial charge migration and also by the dispersion of the dielectric anisotropy.
The second step consists of applying the constant voltage $V = V_n$ to the sample. In the dc case, charged impurities in the bulk of the nematic migrate to the PI/nematic interfaces in such a way that the external electric field is eventually screened sufficiently to ensure that the effective applied voltage is smaller than $V_{Dc}$. This is illustrated in Fig. 3(a) where the long-term dynamics of the polarization conversion is displayed. Indeed, after a few minutes of irradiation $\eta$—hence the liquid crystal reorientation amplitude—has no longer a monotonous increase and eventually decays to zero, in stark contrast to the ac case where $\eta$ asymptotically converges to unity. As ionic transport takes place on different timescales, it is an important factor for the liquid crystal reorientation, interface effects, and the liquid crystal response to the electric field, as investigated in Ref. 24. Unfortunately, we do not have available specification of the conductivity of MLC-6608 to ensure a quantitative discussion on the screening time that is influenced by the concentration and the nature of ionic impurities. Note that, when reoriented, the liquid crystal film exhibits in both cases a random collection of nonsingular topological defects called umbilics, with a topological charge of $\pm 1$, as illustrated in Fig. 3(b) where the sample is imaged between crossed linear polarizers.

A self-induced, on-axis, umbilic is then obtained in the dc case by the local photo-electric effect, which releases the screening. In turn, this activates the local reorientation of the liquid crystal, notating that, despite its photoelectric origin, the applied torque acting on the director is of electrical nature. Since $V = V_n$, one can expect full photo-descreening to have the potential to create highly efficient q-plates. This implies an appropriate combination of incident power, beam waist, wavelength, and materials. In the present case, we used a moderately focused incident laser beam with a waist radius of $w = 260 \mu m$ in the liquid crystal layer. Photo-descreening dynamics as a function of the incident beam power, $P$, is shown in Fig. 4(a) where the dynamics of $\eta$ is presented for $P = 1$, 5, and $10 \ mW$. Steady-state purity as high as $\eta > 90\%$ is obtained for $P > 10 \ mW$, and this corresponds to the purity of the spin-orbit vortex generation process. Indeed, the reoriented structure is a localized umbilical defect with a topological charge of $\pm 1$, whose retardance increases with time to reach $\Lambda \simeq \pi$ at the steady-state, as qualitatively illustrated when imaging the sample between crossed linear polarizers at various stages of the dynamics, as shown in Figs. 4(b) and 4(c).

Quantitatively, the effective director structure at the steady-state is assessed by spatially resolved polarimetry [see Fig. 4(d)] where the map of the in-plane director orientation angle, $\psi$, is displayed. As discussed in previous works, the swirled structure of the umbilic (whose handedness is random) is inherent to the elastic anisotropy of nematic liquid crystals.

Self-induced optical vortex generation itself is illustrated in Figs. 5(a) and 5(b) where the total output far-field intensity
patterns are shown for both incident helicities \( \sigma = \pm 1 \). The characteristic doughnut profile associated with a vortex beam is observed, and the helicity-dependent topological charge \( \ell = 2\sigma \) of the vortex is inferred from the interference pattern resulting from the coherent superposition of a Gaussian shaped reference beam [see Figs. 5(c) and 5(d)]. Indeed, two-arm spiraling patterns, with opposite handedness, are observed for \( \sigma = \pm 1 \). Note that the distinct radii of the annular intensity profiles are the signature of the above-mentioned swirled nature of the localized photo-induced q-plates that act as aspheric asymmetric vortex lenses. They are best represented by a complex transmittance phase mask of the form \( \Gamma = \exp(2i\sigma(\phi + \chi(r))) \), where the real function \( \chi(r) \), with \( r \) the distance from the defect core, refers to the deviation of the director orientation with respect to a purely radial structuring given by \( \psi = \phi \).

The observed high efficiency implies a spatial extent of the self-induced q-plate, associated with birefringent phase retardation \( \Delta \approx \pi \), larger than the incident beam waist diameter. It can be experimentally verified by retrieving the radial dependence of the polarization conversion purity. This is done by evaluating, in the sample plane, the ratio

\[
\xi(r) = \frac{\langle |I_{\omega}(r)\rangle}{\langle |I_0(\sigma_\omega)\rangle},
\]

where \( \langle |I_{\omega}(r)\rangle = \frac{1}{2\pi} \int |I_{\omega}(r, \phi)\rangle d\phi \) is the azimuth average radial intensity profile of the output circularly polarized component with helicity \( \pm \sigma \). Experimentally, it is realized by modifying transiently the focusing condition of the laser beam, namely, by increasing the beam waist more than five times. This allows probing the sample without affecting the steady-state associated with the focused writing beam. The results that correspond to the steady-state situations, presented in Fig. 4(a), are shown in Fig. 6. We find \( \xi(r) > 90\% \) up to \( r \approx 1.3w \) for \( P = 10 \text{ mW} \), which highlights the uniformity of the self-induced q-plate retardance. Finally, regarding the relaxation times of the generated space-variant anisotropic optical element, we note that our system has an elastic relaxation time of the order of 1 s, whereas the characteristic time associated with the spatial distribution of the electric charges is \( >10 \) min.

Summarizing, the obtained record value for the purity of the optical vortex generation process has demonstrated the potential of photoelectric activation of liquid crystal/polymer interfaces as a promising route to develop a self-adapted spin-orbit photonic toolbox for agile management of the orbital angular momentum of light, based on the combined action of light and a dc electrical stimulus. These results, therefore, extend the previous use of liquid crystal light valves based on photoconductive crystals to usual polymer-coated transparent glass substrates. Achieved performances can be improved further if air/glass interfaces are properly treated by anti-reflection coatings since Fresnel reflections obviously limit the efficacy.

REFERENCES


Indeed, the optical contribution to the net torque exerted on the director is, at least, four order of magnitude smaller than the electrical contribution, ensured by having \( P/P_{th} \approx 10^{-4} \propto (V/V_{th})^{2} \approx 1 \), according to the optical Fréedericksz threshold power estimated from Ref. 28.
