

# Photoalignment

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## ABSTRACTS

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# **On the history of reactive mesogens and the importance of dye-enhanced chirality**

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Since their development in the 80's of last century, reactive mesogens (RM's) form a versatile class of soft materials. Advancing on the liquid crystal elastomer technology as developed by Heino Finkelmann et al., the frozen-in molecular order of the low molar mass liquid crystal monomers that is preserved during their photo-crosslinking led to a wealth of new applications. Initially, at the Philips Research Laboratories, the RMs were developed for their low linear thermal expansion coefficient thus reducing thermal stresses in their use as protective optical fiber coatings. But soon it was found that the polymer networks as produced by the RM's demonstrated fascinating optical properties, such as high birefringence and polarization selective reflectivity, which drew the attention of developers in the then emerging flat panel display industry and their suppliers. Typical examples of new applications were polarizing beam splitters, birefringent films improving liquid crystal displays on their viewing angle, non-absorbing polarizers improving the displays on brightness and efficiency, and polarizing gratings as produced by polarization holography.

A crucial step in the decision of companies as Philips and Merck to scale up the production of the RM's was the ability to control chiral pitch and network morphology by dye-enforced reaction diffusion processes. It provided the possibility to form wide band reflective polarizers. But also later, by adding dichroism to either dye or photoinitiator, products could be developed with new functions in optics and actuated mechanics. The lecture will place the development of RMs in a historical perspective and discuss several of the early-day applications, some of them even been forgotten but still with the potential to inspire those new in the field as well as some recent development within the framework of photoalignment and photopatterning.

# Sunlight-triggered reversible soft-bonded azo dye materials for optical patterning and controlled photo-dis-assembly

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Photo-reversible azobenzene (azo) materials have been studied for many years, for applications as optical devices, photo-actuators, and most recently as light-triggered disassembly /reassembly systems. Reversible dis-assembly is based on soft-bonding a material together, *via* non-covalent interactions such as hydrogen bonds, halogen bonds, or electrostatic or ionic interactions, with azobenzene as the photo-reversible cross-linker. Upon irradiation, visible light photons are sufficient in energy to disrupt the self-assembled structures mechanically or electrostatically, gently dis-assembling the material back into its starting components, unchanged and unharmed. If one or both of the starting materials is volatile, or soluble in the environment of the irradiation, then an abrupt and distinct phase change is triggered by the light.

The systems most amenable to detailed quantitative study are co-crystals, of either *trans* or *cis* azos, with a volatile co-former such as dioxane or pyrazine, where the photo-disassembly process can be monitored carefully with X-ray crystallography and Raman spectroscopy in real-time. This leads to stable crystalline solids that can be cut, shaped, and ‘carved’ precisely at room temperature gently with low powered visible light, at a resolution limited only by the laser focusing. A more recent variation on this photo-dis-assembly technique is to combine water-soluble azo food dye cross-linkers with natural aqueous biobased polymers such as cellulose or chitosan, where sunlight triggers the *trans-cis* isomerization of azo dyes, leading to dis-assembly while being gently washed, returning the material back to its non-toxic water-soluble components.

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# Real-time probing of orientation and deformation of azo-polymer films under pulsed irradiation

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Photo-sensitive polymer films containing azo-benzene chromophores show deformation and optical diffraction even after brief irradiation with holographic polarisation patterns on a scale of a few seconds. Using atomic force microscopy and the diffraction of a weak probe beam, we monitor the topography and the molecular alignment *in situ* and in real time. [1] The data show temporal profiles with a complex response combining saturation and sustained growth. The experimental data are compared to theoretical models of the orientation induced by repeated *trans*  $\rightarrow$  *cis* cycles and of the corresponding photo-stationary state. We derive closure relations for 2nd and 4th order moments of the angular distribution functions and construct an efficient way to solve the kinetics of the relevant order parameters. [2, 3] The model improves upon the Ansatz of Bublitz & al (2001) who were assuming that molecules can only orient along three orthogonal directions. [4]

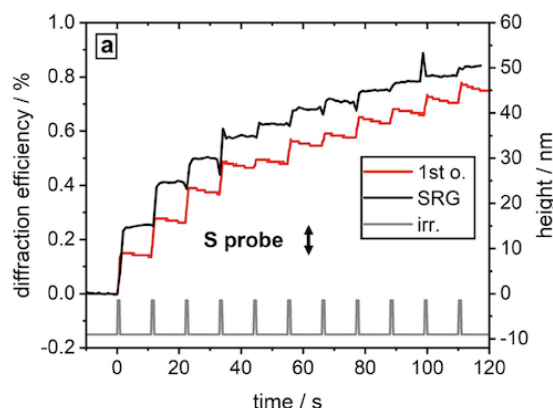


Figure 1. Diffraction efficiency (red) and surface topography amplitude (black, top-to-valley) of a 1 $\mu$ m PAZO film under pulsed irradiation (gray, on/off line) with an S:S interference pattern.

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# Light-induced structuring of azobenzene-containing materials: from single beam to holographic photopatterning

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When exposed to UV/visible light, the free surface of thin films of amorphous azobenzene-containing materials (azomaterials) deforms producing a topographic relief pattern. The morphology of such pattern replicates the geometry of the of the illuminating optical field, depending simultaneously on the spatial gradients in both the intensity and the polarization distributions of the light. The surface reliefs remain stable when the illumination stops, but they can be deformed or erased when a new configuration of the optical field is used. Reconfigurable structured surfaces at micro and nanoscale can be directly produced by light irradiation, without any post-exposure processing [1].

Despite the enormous implications that this simple phenomenology has for applications, the development of new optical configurations to manipulate the geometry of the surface reliefs has not, surprisingly, kept pace with the enormous development on new azomaterials over the years.

Here, I will present the recent achievements of my group in the direct photopatterning of azomaterials with vectorial light fields in different experimental configurations, starting from spontaneous surface self-structing under single beam irradiation and interference-based photopatterning, to state-of-the-art optical configurations based on digital holographic projections for arbitrary patterning [2]. This will help the discussion in the community about underlying physical processes and will trigger the attention on the disruptive application potential of azomaterials in photonics and surface engineering [3-5].

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# Azobenzene-powered optical Fourier elements

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In this talk, I will briefly introduce how our friend azobenzene can play a pivotal role in the development of optical Fourier elements, which have been viewed as a central part of emerging augmented reality (AR) and virtual reality (VR) displays. The inherent optical potential of the interference light pattern is well compatible with Fourier optics, as intensity or polarization is sinusoidally modulated across the grating vector of the interference light pattern. However, unfortunately, conventional photolithography using interference light patterns typically results in binary digitated grating, inevitably leading to undesired frequency-mixing at the far-field regime, such as diffraction. This limitation arises from the fact that the photochemistry of conventional photoresists is generally designed to react in an all-or-nothing fashion based on the threshold intensity. In contrast, as well recognized, particularly in this conference community, azobenzene-incorporated material platforms, such as supramolecules and polymers, allow the Fourier potential of interference light pattern to be readily translated into grating structuring. Surface relief gratings (SRGs), accessible by illumination of interference light pattern onto azobenzene materials, have been a representative example in this regard.

Although azo-SRGs have a relatively long research history of about 28 years (as of Sep 2023), provisioning of them as optical Fourier elements (referred to as optical Fourier surfaces, henceforth OFSs) has only recently been suggested [1,2]. Herein, I will revisit this aspect in terms of the azobenzene-powered development of OFSs and its limitations. Also, I would like to emphasize that the role of azobenzene in the development of optical Fourier elements is not restricted to surface gratings. Volumetric gratings can also benefit from the exotic photophysics of azobenzene (i.e., optical Fourier volumes, OFVs) [3]; I'll introduce the recent progress made by our group in reshaping OFVs together with azobenzene. Of course, it is important to note that there are technological drawbacks to using azobenzene-based methods for fabricating OFSs and OFVs at this point. In the last part of this talk, I will share our group's results and strategies for overcoming these technological challenges.

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# Polarization-driven reversible deformation in an azopolymer-elastomer composite

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Within the field of light-responsive polymers, sidechain azopolymers present the particularly intriguing feature of anisotropic deformation response upon exposure to linearly polarized light. This feature has been widely used for the creation of sinusoidal surface relief gratings (SRGs) on polymer thin films as well as the reshaping of microstructures along arbitrary, polarization-dependent directions. However, the deformation of such polymers is usually plastic, with multiple exposures leading to cumulative deformations rather than reversible ones. While this fact is useful for advanced fabrication techniques such as complex surface patterning [1,2], it poses severe restrictions on the development of dynamic devices.

On the other hand, azopolymer microparticles have recently been shown to display reversible behaviour when immersed into a rubbery external matrix, potentially linked to the restoring effect of the latter [3]. Here we present a novel polymeric metamaterial where high density ensembles of embedded microparticles transfer a reversible, anisotropic stretching to the overall material. The material, made entirely from commercially available polymer constituents, can switch between anisotropically stretched and in-plane expanded deformation states upon alternating exposure to linearly and circularly polarized light, with low creeping losses. The latter are measured by means of a Fourier-based algorithm, precisely tracking the deformation of planar thin films decorated with micropillar markers.

Finally, versatile soft actuators able to undergo complex deformations in 3D are proposed. Utilizing a combination of anisotropic polarization-controlled stretching and absorption gradients, the free membrane actuators smoothly undergo ample and diverse configurational changes, including twisting, roll-in, grabbing-like actuation, and even continuous, pivot-less shape rotation, examples being shown in Fig. 1.

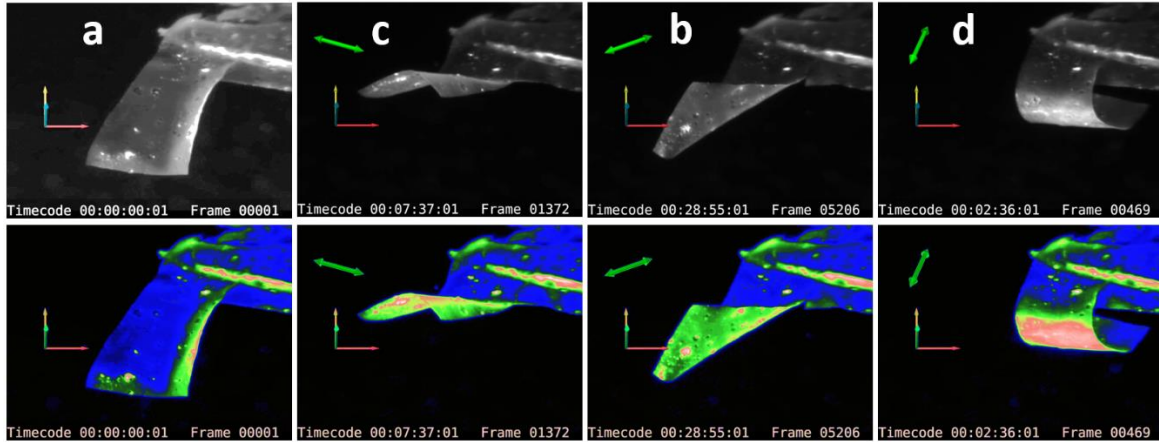


Figure 1. Examples of soft actuator 3D movements. (a) Undeformed actuator attached to a glass slide forming a rigid boundary condition. (b-d) Top irradiation with polarization (indicated by green double arrow) at an angle of 0, 45, 90 degrees with respect to the boundary condition leading to twist (b), elevation with lateral downwards bending (c) and full roll-in (d) respectively.

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# Large force-displacement actuation via liquid crystalline elastomer actuators prepared by photoalignment

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Liquid crystal elastomers (LCE) are soft materials capable of actuating loads many times their own mass.[1] Azimuthally photoaligned LCE are known to adopt conical geometries as they undergo order to disorder thermotropic transitions.[2] Stacked photoaligned LCE elements provide significant increases in absolute load and work capacity with greater thickness.[3] Molecular engineering of stacked LCE elements has recently yielded 32+ layered LCE elements that actuate at lower temperatures, carry loads greater than 1 N, and respond dynamically to transient compression.[4] Here, we use optical profilometry and transparent loading of conical LCE actuators to study the topographic response of LCE cones to compression. Topographic trends emerge, such as waveforms and particular modes of cone buckling, that depend on thickness and composition.[5] These carry implications for mechanics and further applications for stacked LCE actuators.

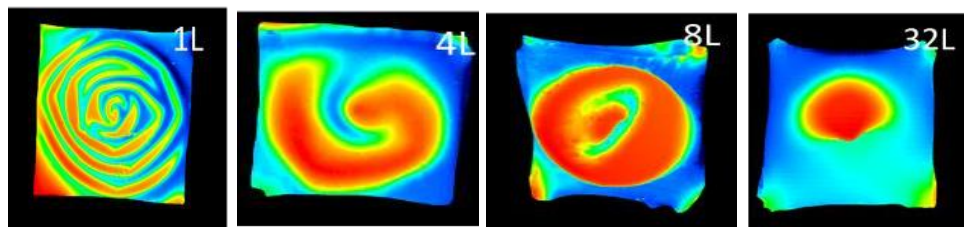


Figure 1. Topography under high compression for 1-layer, 4-layer, 8-layer, and 32-layer stacked elements of photoaligned liquid crystal elastomers.

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# **Responsive material actuators enabled by photoalignment of liquid crystalline materials**

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Liquid crystalline materials are pervasive in our homes, purses, and pockets. It has been long-known that liquid crystallinity in polymers enables exceptional characteristics in high performance applications such as transparent armor or bulletproof vests. This talk will generally focus on a class of liquid crystalline materials referred to as liquid crystalline elastomers. These materials were predicted by de Gennes to have exceptional promise as artificial muscles, owing to the unique assimilation of anisotropy and elasticity. Subsequent experimental studies have confirmed the salient features of these materials, with respect to other forms of stimuli-responsive soft matter, are large stroke actuation up to 75% as well “soft elasticity” (stretch at minimal stress). This presentation will survey our efforts in directing the self-assembly of these materials to realize distinctive functional behavior with implications to soft robotics.

# Azopolymers and azopolymer-based nanocomposite materials for applications in polarization holography and photonics

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Azopolymers are known as highly efficient photoanisotropic materials with numerous applications, e.g. for inscription of polarization-selective diffractive optical elements, photoinduced chiral structures, surface relief gratings, 1D and 2D diffractive structures, as photoalignment layers, etc. Recently, it has been shown that the optical response of azopolymers can be enhanced even further by doping with nanoparticles with various morphology. We have performed extensive studies on azopolymer-based nanocomposites and in particular on the influence of the concentration, composition, shape and size of the embedded nanoparticles on their photoanisotropic characteristics – birefringence as well as diffraction efficiency and surface relief amplitude in case of polarization holographic recording. A summary of the results from these studies will be presented, along with potential applications.

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# An overview of spatial light modulator techniques for polarization-driven writing of surface microstructures on azopolymer films

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Spatially structured optical polarization drives a photomechanical response in azobenzene polymer films, resulting in the formation of surface relief microstructures. The polarization is typically generated via multibeam laser interference, which is limited to writing structures of fixed period and orientation. Here we demonstrate that a spatial light modulator (SLM) configured as a polarization rotator is an alternative writing technique with several key advantages. The SLM can project structured polarization that is spatially addressable, enabling the facile fabrication of chirped surface relief gratings, as well as circular grating profiles. Gratings for structured color can also be written by spatially addressing the SLM with periodicities for red, green, and blue co-propagation of diffracted orders.



Figure 1. Grating micrograph of continuously chirped SRG photopatterned on azopolymer film using SLM-based system. Each horizontal stripe is approximately 0.5 mm in length with average periodicity of 2.0  $\mu\text{m}$ .

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# Active surface command layers: from polarization holography to plasmonics

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Active optical components are essential for several photonics applications ranging from sensing to light filtering. Innovative technological applications that utilize stimuli-responsive integrated optical devices is in very high demand because of the need for configurable and on-demand optical components. Photoalignment is a powerful optical technique that exploits azobenzene molecules that have boosted the utilization of liquid crystals (LCs) to convey tunability to modern devices such as smartphones and lidars. This presentation discusses several compelling opportunities that rely on utilizing an innovative class of photoalignment materials. In particular, we show how a digital spatial light polarization converter (DSLPC) enables the realization of “free form” optical components with arbitrary spatial patterns of the optical anisotropy axis orientation.[1, 2] In addition, the same class of photoalignment materials opens new and exotic opportunities for realizing unique plasmonic-based optical components [3] that can be efficiently controlled through external stimuli. Our studies open a new scenario in boosting breakthroughs in optical applications that can be easily extended to several other research applications, such as solar energy and light harvesting.

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# Reprogrammable flat optics from maskless azopolymers photo-morphing

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From natural inspiration, flat patterned surfaces can shape the wavefront of light, offering significant potential for science and technology, including augmented reality devices, holographic projectors, and space telescopes, among others. While electronics currently dominate, the future is expected to shift toward photonic technologies that use light to process information and exchange data. However, both electronic and photonic components rely on complex lithographic processes that result in inefficient use of resources and energy and the generation of hazardous waste, hindering economic growth and innovation. Replacing standard photoresists with azopolymers can be a game changer.

Amorphous azopolymer thin films form photoinduced surface reliefs under UV-visible light absorption. The free surface locally deforms according to incident light's intensity, polarization, and wavefront. This phenomenon arises from reversible surface mass migration triggered at the molecular level by azobenzene molecules isomerization cycles [1]. This unique photomechanical process allows the direct fabrication of arbitrary and reprogrammable surface geometries when advanced illumination configurations are used to project spatiotemporally shaped light patterns. As direct and reconfigurable photoresists, azomaterials form the basis for a one-step, all-optical and reversible lithographic workflow.

Here, I will report the design and the fabrication of azopolymer light-induced reliefs resulting from a computer-generated hologram illumination scheme. This technique allows the generation of grayscale intensity patterns on the azopolymer surface by phase-modulating a laser beam through a computer-controlled spatial light modulator [2]. In this way, almost arbitrary geometries in the light distribution can be achieved, allowing precise and fast control of all the lithographic parameters necessary for the realization of fully reprogrammable Diffractive Optical Elements (DOEs) [3-5]. These flat optical components, acting as transmissive phase retarders, can modulate light within a thickness comparable to the light wavelength. DOEs, implemented by digital holographic inscription of an azopolymer surface, can be arbitrarily shaped and dynamically tuned or reprogrammed to achieve the desired optical functionality. In addition, the evolving surface geometry can be analyzed and optimized right during the fabrication, resulting in unprecedented control over the final diffractive functionality.

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# Reactive mesogens for large area reflective geometrical phase optical systems

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Light reflected from a periodic helical structure such as a cholesteric liquid crystal (CLC) acquires a geometrical phase that can be used for designing flat optical components with variety of functions [1-3]. Lenses, “prisms”, and vector vortex waveplates have been demonstrated so far [4]. A key advantage of the technology compared to transmissive optics is that the geometrical phase of the reflected beam does not require meeting half-wave retardation condition for high efficiency. The periodic helical structure is obtained spontaneously due to self-organization of the liquid crystal containing chiral components. However, it is hard to ensure high quality of CLCs, particularly in large area. The pitch of CLCs for visible wavelengths is less than a micrometer, and slightest non-uniformities in the cell gap generate disclinations and are revealed as structural deformations reducing the reflection efficiency. In the case of CLC polymers, multiple layers need to be deposited, for example, by spin coating, to produce high efficiency reflection.

Reactive mesogens developed at BEAM Co. allow fabrication of reflective geometrical phase optics based on CLC polymers with no haze and high efficiency over large area [5]. Moreover, we have demonstrated such components with polarization-independent functionality by combining right-and left-hand circular CLC polymers.

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# Human Interactive Materials for soft robotic machine/ haptics applications

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We propose the use of a liquid crystal polymer network for soft robotics where the various molecular accessories are assembled in the two dimensions of a coating. For instance, the LCN surface deforms dynamically into a variety of pre-designed topographic patterns by means of various triggers, such as temperature, light and the input of electric field<sup>1</sup>. These microscopic deformations exhibit macroscopic impact on, for instance, tribology, haptics, laminar mixing of fluids in microchannels and directed cell growth. Another robotic-relevant function we brought into the LCN coating is its capability to secrete liquids under UV irradiation or by an AC field<sup>2</sup>. This controlled release is associated with many potential applications, including lubrication, controlled adhesion, drug delivery, and agriculture, antifouling in marine and biomedical devices, personal care and cosmetics. With this we bring together a tool box to form two dimensional soft robots designed to operate in area where man and machine come together.

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# Multifunctional stimuli-responsive LC polymer composites

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Stimuli-responsive liquid crystalline (LC) materials have attracted significant attention and future progress is expected in their application for optics and photonics [1]. In our research, we have developed approaches for creating novel stimuli-responsive LC polymer systems with structure and optical properties that can be easily controlled using light, electric fields, and mechanical deformation. The first type of such materials is based on highly porous cholesteric polymer scaffolds prepared by photopolymerization of mixtures containing mesogenic mono- and diacrylates, followed by removing low-molar-mass components. By filling these scaffolds with photochromic LC mixtures or LC polymers, we prepared cholesteric composites with photo- and electro-tunable selective light reflection spectra [2]. A similar approach was used to create nematic photo- and electro-switchable 2D diffraction gratings [3].

Another type of polymer composites corresponded to cholesteric LC mixtures dispersed in elastic polymer matrices such as polyurethane, plasticized polyvinyl alcohol (PVA), and polydimethylsiloxanes [4]. Stretching the films of these composites results in significant and reversible short-wavelength shift of the selective light reflection peak, with an amplitude of 100 nm or more (Fig. 1, left). Furthermore, incorporation of a chiral-photochromic dopant into cholesteric mixtures enables the creation of composite films with photo- and mechanically tunable selective light reflection (Fig. 1, right) [5].

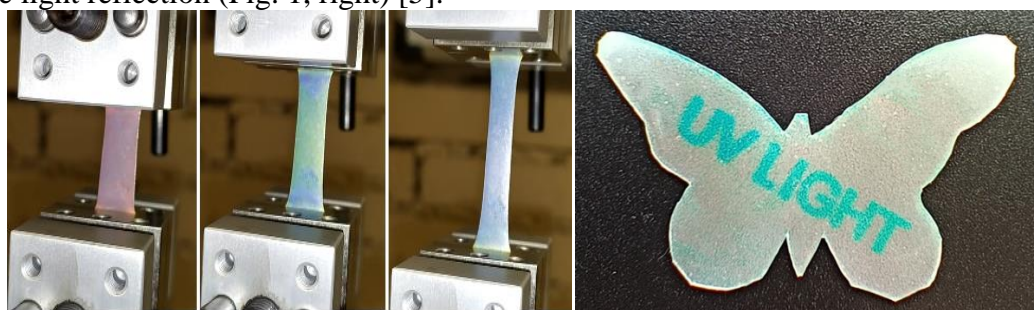


Figure 1. Photos of the mechanosensitive PVA-based cholesteric composite film before and after stretching by 0%, 75% and 150% (left); photochromic elastic composite based on PVA after UV-irradiation through a mask (right).

The LC composites obtained in our study can be considered as promising systems for developing new materials for photonics, optoelectronics, mechano-sensors, protection of securities and documents against counterfeiting, and other related applications.

## Acknowledgments

This research was supported by the Russian Science Foundation (22-13-00055).

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# Protonation effects on the isomerization kinetics of functionalized azo photoswitches

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Azopyridines have been described as next-generation functionalized azobenzene photoswitches which possess the chemical properties of the pyridine ring, including pH sensitivity, hydrogen- and halogen-bonding, and coordinative abilities towards metals. These properties enable the construction of supramolecular assemblies of azopyridine-containing polymers, employed for the photocontrol of liquid crystals, metal organic frameworks, composite materials actuation, micelles, and so on. However, detailed studies on the isomerization kinetics of parent azopyridines and their protonated forms are lacking, which would improve the design of these materials and extend their application to biological contexts. Moreover, there is a conspicuous lack of literature on the isomerization kinetics of azopyridines in chlorinated solvents where adventitious protonation can occur. In this work, irradiation of 4-phenylazopyridine (AzPy) in dichloromethane with 365 nm light, a typical *trans-cis* activation wavelength for azopyridines, produced bathochromic and hyperchromic shifts in the  $\pi$ - $\pi^*$  and  $n$ - $\pi^*$  bands. These bands shifted from 312.5 nm ( $24000\text{ M}^{-1}\text{cm}^{-1}$ ) and 452 nm ( $460\text{ M}^{-1}\text{cm}^{-1}$ ) to 340 nm ( $26000\text{ M}^{-1}\text{cm}^{-1}$ ) and 473 nm ( $580\text{ M}^{-1}\text{cm}^{-1}$ ), respectively. Alongside these shifts, there was a significant acceleration of the isomerization rate, which was attributed to the formation of AzPy protonated at the pyridine nitrogen due to HCl production from UV-mediated dichloromethane photodecomposition. Density functional theory calculations demonstrated that the thermal isomerization of AzPy and the protonated form, AzPyH<sup>+</sup>, proceed primarily through inversion of the pyridine ring, where the activation barrier of AzPyH<sup>+</sup> was markedly reduced, corresponding to a 9-fold acceleration in the isomerization rate. This acceleration was attributed to significant weakening of the azo bond in AzPyH<sup>+</sup> through an inductive effect, behaving similarly to fast-isomerizing push-pull azobenzene systems.

# Periodic photoalignment for aligning nematic, chiral nematic, blue phase and ferro-electric nematic liquid crystal

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By using periodic photoalignment on one or two substrates, different liquid crystal (LC) materials (nematic [1], chiral nematic [2,5,6], blue phase [3], and ferro-electric nematic [4]) can be appropriately oriented. Nematic diffraction gratings can be used for optical components, beam steering or smart windows. With chiral LC, reflective optical components with short periods and large diffraction angles ( $>60^\circ$ ) can be realized [5,6]. In addition, we will discuss an algorithm to estimate the preferred alignment direction when multiple steps with arbitrary polarization states are used to illuminate a photosensitive layer [8].

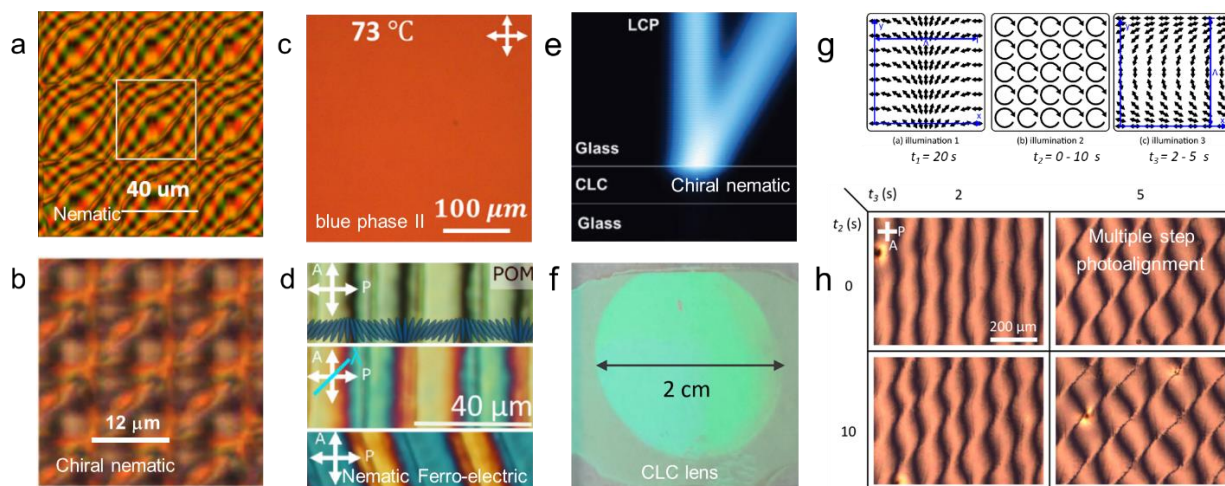


Figure 1. (a) Diffractive grating for multiple order diffraction [1]; (b) Chiral nematic grating for bistable switching [2]; (c) Homogeneous blue phase by 200 nm periodic photoalignment [3]; (d) Splay photoalignment for nematic ferro-electric LC[4]; (e) Simulation of reflection on a chiral LC layer with titled helical axis [5]; (f) chiral CLC lens for a spectrometer [6]; (g-h) modeling photoalignment for multiple illumination steps [7].

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# Lattice arrangement of blue phase liquid crystals on a patterned photoaligned surface

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We investigated the lattice alignment behavior of BP liquid crystals, which forms a three-dimensional helical periodic structure, on a photo-patterned surface. As the orientation pattern, two types of one-dimensional periodic pattern in which the alignment easy axis changes continuously and changes discontinuously was adopted. When the direction of the easy axis is changed continuously, a slant structure is formed as in the cholesteric phase. We have already reported the electrooptic behavior from a slant-structured BP lattice structure [1]. Here, we analyzed the BPII lattice arrangement in the deflector based on Bragg reflections and demonstrated that the lattice structure was slanted with respect to the substrate, which was confirmed using transmission electron microscopy. In addition, we calculated the similarity between the alignment pattern on the substrate surface and the director arrangement of BPII near the interface, confirming that the structure observed experimentally matched the most probable calculated structure [2].

On the other hand, if the easy axis direction is changed discontinuously, frustration occurs in the lattice arrangement between adjacent pattern regions. As a result, it was found that the lattice arrangement behavior differs depending on the pattern size, in this case, the pattern period [3]. That is, when the period of the pattern regions having orthogonal easy axis is large, the (110) plane is parallel to the substrate, and the [001] axis of each pattern region is aligned parallel to each easy axis direction. However, when the period was small, the (100) plane was parallel to the substrate over the entire surface of the device, and the lattice orientation was uniformly arranged in the average direction of the adjacent orientation regions.

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# Highly resolved and cross-talk free multiplexed holograms via broadband vectorial interferometry

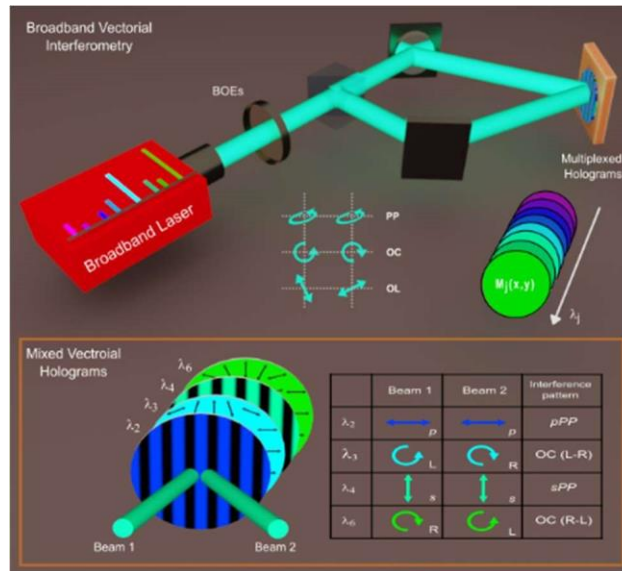
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Over the past years, there has been a growing interest in the development of ultracompact micro and nanooptics, driven by their efficient applications in augmented/virtual reality, displays, and Fourier optics. The current technologies, mainly based on meta-solutions, suffer in terms of efficiency, tunability as well as complex and multi-step fabrication methods which may limit their full-scale potential. Herein, we propose and demonstrate the capability in producing ultracompact optical platforms through single-step vectorial patterning, involving a broadband laser beam [1]. The high correlation of light fields originating from the same source and coupled in the same Gaussian mode discloses new features of the method, rooted in a selective light-matter interaction. The reported results confirm unexpected performances of the vectorial encoding where even a “depolarized” beam enables to record highly resolved, mixed, and cross-talk free multiplexed holograms. New light abilities emerge, overcoming presumed limitations in small scale and thin optics applications, and suggest original design of miniaturized multifunction polarizing and diffractive platforms. The favorable performances (large area, single step, fast and cost-effective process, in-situ optical design, etc.) of the method in printing structures with high fidelity demonstrate that vectorial encoding via multiline laser beam introduces a significant innovation to a powerful and flexible tool for devices manufacturing with nanometric spatial separation. Furthermore, it offers excellent insights towards new generation optics and provides a viable alternative to complex and multi-steps meta-solutions, giving real perspectives to the scaleup of ultracompact optical devices.





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# Photoaligning properties of the hybrid layer based on LIPSS on dielectric material covered with photopolymer

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In the last years, the use of the Laser-Induced Periodic Surface Structuring (LIPSS) of various materials (*i.e.* metals [1], dielectric materials [2, 3]) to align the nematic LCs has been studied intensively.

In our work, we investigate the nanostructured transparent dielectric materials such as Sapphire and SiO<sub>2</sub>, having different characteristics of nano-grooves such as the period  $\Lambda$ , depth  $A$ , and width of unstructured gap  $L$ , as an aligning layer by means of the combined twist LC cell method. To obtain the ‘command’ aligning layer shown in Fig. (a) and phototune the anchoring energy of the LIPSS-treated substrate, the surface was covered with the photopolymers (*e.g.* PVCN-F or azopolymer based on polymethacrylate) to form the so-called hybrid aligning layer (HAL). We found that selection of the proper UV light treatment conditions (*e.g.* relative orientation between the polarization of UV light and direction of nano-grooves, irradiation time) allows to obtain the values of the azimuthal anchoring energy within a three orders of magnitude range  $10^{-8} - 10^{-5} \text{ J/m}^2$ , see Fig. (b).

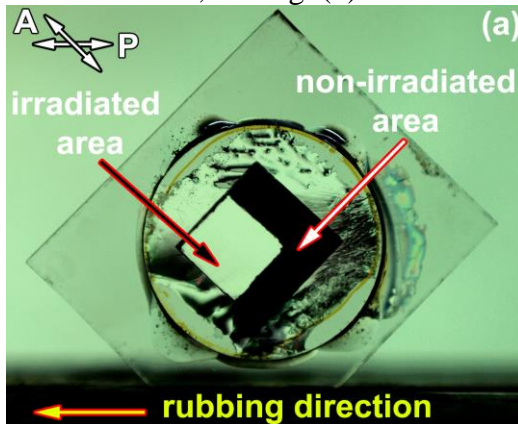


Fig. (a). Photo of the combined twist LC cell that has the HAL (square-shaped area) based on nanostructured SiO<sub>2</sub> substrate ( $\Lambda = 555 \text{ nm}$ ,  $A = 160 \text{ nm}$  and  $L = 7 \text{ }\mu\text{m}$ ) covered with polymethacrylate with azobenzene side group. HAL was irradiated by UV light ( $\lambda_{\text{max}} = 365 \text{ nm}$ ).

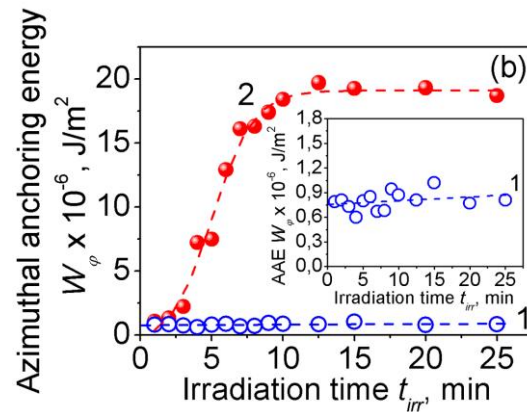


Fig. (b). Irradiation dependence of the azimuthal anchoring energy of the HAL that consists with LIPSS-treated sapphire ( $\Lambda = 980 \text{ nm}$ ,  $A = 100 \text{ nm}$  and  $L = 17 \text{ }\mu\text{m}$ ) covered with PVCN-F for the non-irradiated (blue line 1) and irradiated (red curve 2) areas.

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# Photopatterned liquid crystals to control biological systems

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Microscale biological systems such as swarms of swimming bacteria and cell tissues demonstrate fascinating out-of-equilibrium dynamics. This dynamics is difficult to control by factors other than transient gradients, such as gradients of nutrients; visual, acoustic and tactile communication channels that humans use to control large animals are not effective. To establish communication with microscale biological systems, we propose to use liquid crystals with photopatterned molecular orientation. The technique is based on the plasmonic metamasks that produce spatially-varying polarization state of the light beam [1, 2]. We report on the measurement of the azimuthal anchoring strength [3] and how the photopatterned liquid crystals can be used to command the dynamics of two living systems, (i) swimming bacteria [4-7]; (ii) tissues formed by living cells [8]. The control of biosystems by photopatterned liquid crystals might result in new approaches to harness the energy of collective motion for micro-robotic, biomechanical, and sensing devices. The work is supported by NSF DMR-2215191 and DMS-2106675 grants.

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# Solar sailing using thin geometric phase elements

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The physics of the sun remains poorly understood, and the constant threat of coronal mass ejections poses a risk of disrupting Earth's electronics-based infrastructure. To enhance the capabilities of helio-scientists in developing more precise physics-based models and CME forecasts, a network of sun-orbiting observatories at high inclination angles is necessary. Solar sails have the potential to reach inclination angles that afford direct measurements of solar activity above the solar poles. This talk will discuss recent research that aims to utilize advanced diffractive films such as geometric phase gratings to create highly efficient solar sails capable of placing a constellation of satellites in various solar polar orbits. Diffractive solar sails provide unique propulsion, navigation, and attitude control advantages compared with conventional reflective solar sails and tip vanes. What is more, the radiation pressure on other geometric phase structures such as bi-gratings afford stable beam-riding opportunities for laser-driven sails.

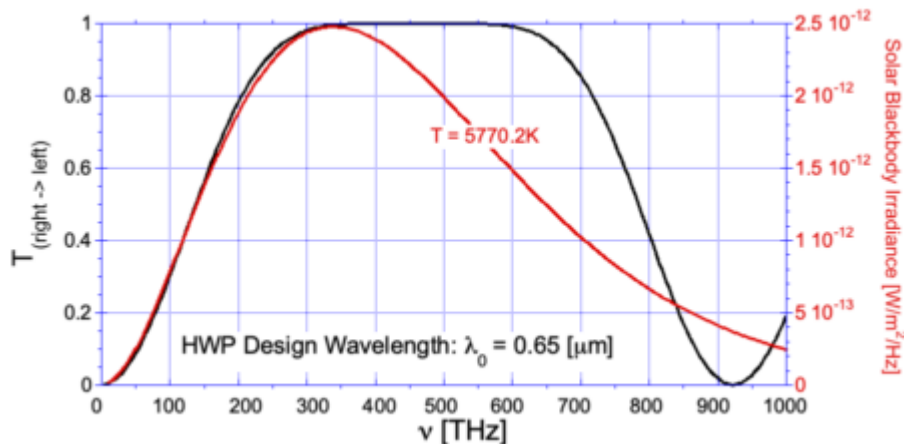


Figure 1. Achromatized geometric phase grating matched to the solar spectrum.

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# Photopatterning of novel surface topographies of canal and well structures on liquid-crystalline polymer films

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Precise control of surface topographies has attracted much attention for developing optical and mechanical applications. In past decades, surface processing technologies based on photoinduced reactions in liquid crystals (LCs) have offered the advantages of fine control with non-contacting operation and reversibility of surface topographies. One representative example is the utilization of photoresponsive dyes in LCs, which can induce various surface morphing behaviors based on phase transitions, Marangoni flow, and molecular orientation changes [1]. Recently, we have proposed a dye-free approach to form surface topographies in LC polymer films simply by photopolymerization with structured light [2–4]. Irradiation with structured light on monomers induces a polymer concentration gradient, causing mutual molecular diffusion and forming surface relief structures. Interestingly, this approach can induce molecular alignment simultaneously with surface structures. Here, we report the direct formation of the novel surface topographies with the depth and width in micrometers. Photopolymerization with specific periodic light patterns formed unusual surface topographies, including wells, canals, and random structures. Moreover, the real-time observation of the photopolymerization process clarified the formation mechanism. In the films with canal and well structures, molecular alignment was simultaneously induced along the structures. They have great potential for the development of stimuli-responsive optical and mechanical devices.

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# Magnified replication of on-axis pancharatnam-berry phase optical elements

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In this work, a method of polarization holographic replication is demonstrated with a 4f optical relay. Such a system produces magnified copies of an on-axis liquid crystal Pancharatnam-Berry phase optical element. The ~100% first-order diffraction efficiency of the Pancharatnam-Berry phase optical element permits replication of an on-axis holographic reference, such as a lens. The resampling of the optical Fourier transform allows for the holographic reference to simultaneously undergo aperture enlargement and magnification of power upon replication. The use of free-form holographic patterning enables pre-compensation of the wavefront error introduced on replication. The necessary pre-compensation phase required to generate the profile of interest in the liquid crystal optical element is discussed.

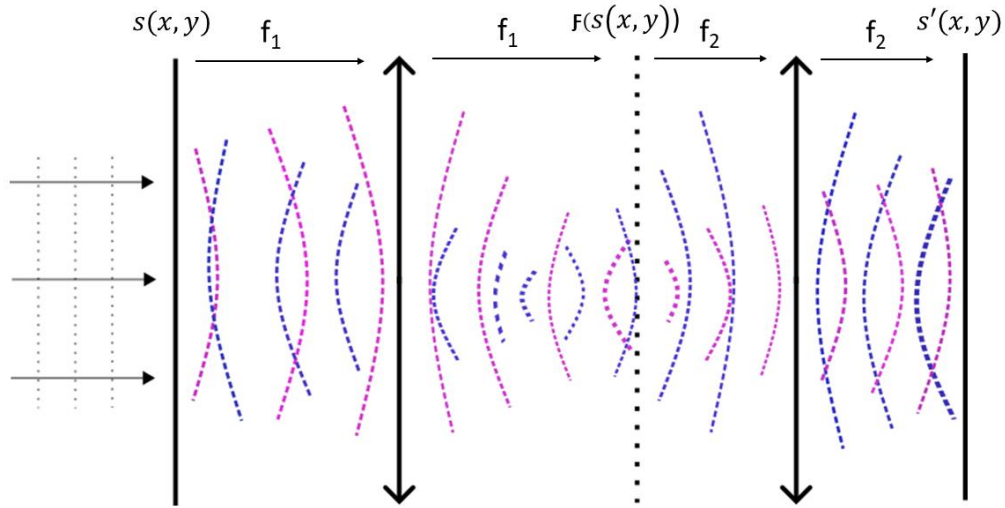


Figure 1. Illustration of RHCP (magenta), LHCP (blue) wavefronts generated by PBOE (plane  $s$ ) relayed by 4f optical system to produce a polarization field (plane  $s'$ ).

# Looking for planets with liquid crystal polymer optics

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Several new astronomical observational modes are being enabled by planar optical components made of patterned liquid crystal polymers. These components include diffractive waveplates for spectropolarimetry, focal-plane optical vortex phase masks for stellar coronagraphy and exoplanet searches, pupil-plane phase masks for beam shaping, “phase-knife” waveplates for stellar nulling interferometry, and even patterned masks for novel wavefront sensors. In some cases, such devices provide an improvement over classical optical approaches, while in other cases, completely novel functionalities arise. Several of these techniques have already been deployed in optical instruments on ground-based telescopes, and further advances should allow such approaches to be used on future space-based telescopes such as NASA’s planned Habitable Worlds Observatory. A brief summary of these novel instrumental modes is provided.

An especially interesting field of astronomy these days is the search for planets around nearby stars (i.e., “exoplanets”), and observational techniques are being pushed to ever greater contrast and sensitivity in order to be able to detect Earth-analog exoplanets around nearby stars, which are on the order of a part in ten billion as bright as their host stars. Indeed, the detection and observation of terrestrial exoplanets is the highest priority of the most recent Astronomy decadal report. As a result, instrumental approaches that can be used with large telescopes to see Earth-like exoplanets are of great interest.

The problem is that, from our point of view, most exoplanets are located at very small angular separations from host stars that are far, far brighter, making them very difficult to pick out of the stellar glare. This huge contrast ratio can be overcome by using an optical instrument called a stellar coronagraph to suppress the much brighter starlight. Coronagraphs are essentially spatial filters: they selectively block or redirect the on-axis starlight, while letting any light from off-axis exoplanets through. One of the most promising types of coronagraph is the optical vortex coronagraph, in which an optical vortex phase mask that is centered on a star’s very well-corrected diffraction pattern modifies the Airy pattern in just such a way that in a downstream pupil plane, all of the starlight appears outside the pupil, where it can be blocked, while the exoplanet light remains inside the pupil, and is transmitted.

One way to make the needed optical vortex phase mask is to use geometric phase, i.e., by making a spatially-variant half-wave plate, in which the optical axis orientations follow the azimuthal pattern of an optical vortex. Such spatially-variant patterns can be provided by liquid crystal polymer optics, but the needed level of starlight rejection requires very high accuracy phase masks. Steady progress is being made, and optical vortex phase masks manufactured recently have enabled the rejection of simulated starlight in the lab to roughly the part in a billion level, i.e., within an order of magnitude of the needed performance level.



# Reaction-diffusion spatiotemporal supramolecular dynamics in photoactive chiral liquid crystals

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Frustrated films of chiral nematics are known to host a variety of localized elastic structures that consist of spatially inhomogeneous distributions of the liquid crystal orientation, which can be classified according to their topological features [1]. The sensitivity of liquid crystals to external stimuli (thermal, electric, magnetic, optical) makes it easy to produce a mechanical work on the director field leading to on-demand emergence of such elastic quasiparticles, while their very nature usually depends on the actual experimental conditions. It is also possible to destabilize the initially uniform director field of a frustrated chiral nematic film by acting at the molecular level, via photochemical reactions. The principle involves modifying the helical twisting power of a photosensitive chiral molecular dopant under irradiation, which can induce various localized elastic structures at very modest optical power budget [2]. Recently, the emergence of localized spatiotemporal dynamics of the director field has been unveiled in such a framework, for which the role of molecular diffusion processes has been discussed [3]. Still, a quantitative description of the phenomenon lacks to date and here we report on a reaction-diffusion model combining photochemical reaction, diffusion of ground state and excited molecular species, as well as elastic distortions. This makes it possible not only to obtain a quantitative description of all the experimental observations reported to date, but also to predict new behaviors, the first experimental observations of which we will present here.

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# Polarization-holographic imaging spectropolarimeter

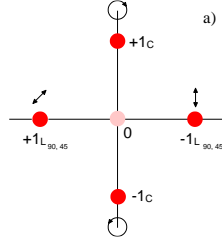
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We have developed the high-efficient polarization-holographic diffraction element by recording several polarization- holographic gratings with different profile of anisotropy and gyrotropy in the band. Such an element gives the possibility of a complete analysis of the polarization state of light in real time. Currently, we use two gratings: "C" recorded by two orthogonally circularly polarized beams and "L<sub>90,45</sub>" recorded by two linearly polarized beams with an azimuth of 45° and 90°, respectively. We have developed the technology of gratings recording on one and the same area of polarization-sensitive material so that diffracted beams are spatially separated and cross-diffraction is not observed.

In the process of diffraction this element decomposes the incident light on the orthogonal circular and linear bases and forms four diffraction orders. The simultaneous measurement of diffraction orders intensities ( $I_{+C}$ ,  $I_{-C}$ ,  $I_{90}$ ,  $I_{45}$ ) by photodetectors enables the complete analysis of the polarization state of light (the four Stokes parameters  $S_0$ ,  $S_1$ ,  $S_2$ ,  $S_3$ ) to be obtained with the developed formulas and software taking into account the values of the reaction functions of the used recording material.



Schematic pictures of diffraction on the element

$$\begin{aligned} S_{0,\lambda_i} &= k_{C,\lambda_i} I_{+C} + k_{C,\lambda_i} I_{-C} \\ S_{1,\lambda_i} &= (k_{C,\lambda_i} I_{+C} + k_{C,\lambda_i} I_{-C}) - 2k_{90,\lambda_i} I_{90} \\ S_{2,\lambda_i} &= 2k_{45,\lambda_i} I_{45} - (k_{C,\lambda_i} I_{+C} + k_{C,\lambda_i} I_{-C}) \\ S_{3,\lambda_i} &= k_{C,\lambda_i} I_{+C} - k_{C,\lambda_i} I_{-C} \end{aligned}$$

The element has a wide working spectral range (500 -1600 nm). It has angular dispersion. Total diffraction efficiency of the element at wavelength of 532 nm is close to 75% and 635 nm close to 55%, and 1550 nm close to 8%. Currently an accuracy of measuring Stokes parameters is about 0.1%.

Polarization-Holographic Element with photodetector matrix (CCD) and appropriate software represent a Real-Time Imaging Spectropolarimeter [1].

The diffraction orders contain the object image that enable the distribution of polarization to be determined in all points of the object image. The dispersion of the polarization distribution can be also obtained in the object image for any wavelengths from a working range (when narrow-band filters are used).

Spectropolarimeter is light-weight, precision, compact, relatively inexpensive, it has no internal reflections, works in real time and has the ability to analyze fast processes with variable polarization.

We have investigated the possibility of using such a Spectropolarimeter in astropolarimetry [2,3], in innovative mobile polarimetric device for determining the distribution of stressed state in various constructions and details, in tasks of remote sensing and recognition.

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# Ultralong quantum-walk photonic simulations using photo-patterned optical elements with engineered design

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Quantum walks are simple quantum-evolution processes that can reproduce the key qualitative features of many interesting complex real systems. Hence, these processes are at the root of many quantum simulations and can be used for quantum computation. A photonic quantum walk can be carried out using various possible experimental architectures. A few years ago, we have introduced an approach for which the walk degree of freedom is encoded in the internal mode structure of a single light beam [1,2]. The walk is then physically realized by a single light beam propagating through a sequence of suitable optical elements exploiting Pancharatnam-Berry (PB) phases to couple internal modes. In our initial approach, each quantum walk step was simulated using a sequence of two-three PB optical elements having relatively simple patterns. Hence, to realize a very long quantum evolution, such as a few hundred steps or longer, a similar number of optical elements would be needed. This can be effective for classical simulations, but in a genuine quantum regime this approach is too lossy. Recently, we have demonstrated a new approach in which only three PB optical elements, suitably patterned in a complex engineered way, allow the photon to undergo in a single brief optical process the evolution equivalent to hundreds of quantum-walk steps [3,4]. In this way, one- and two-dimensional ultralong quantum evolutions can be efficiently simulated.

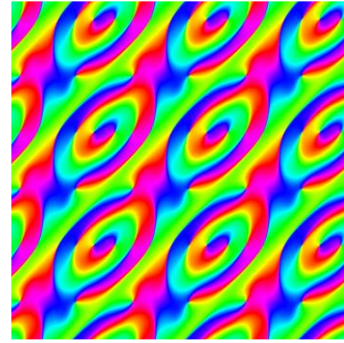


Figure 1. Optic axis pattern (in false colors) of liquid crystal devices used in the realization of two-dimensional photonic quantum walks.

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# Vector beam detection via a pair of two liquid crystal polarization holograms

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The vector beam (VB) has been expected to be used as an optical multiplexing parameter of optical communication [1], because VBs can have an infinite series of polarization topological charges (PTCs). Also, VB has two orthogonal states for the same PTC, such as the radial and azimuthal polarizations (RP and AP), which has a relative polarization angle rotation of 90deg between them. To realize optical communications using VBs, the mode sorter [2], and the fork-shaped polarization grating (FPG) [3] has been reported as a mode demultiplexer. Although, these methods can identify the number of PTCs, they cannot identify the value of initial polarization angle. In this presentation, we introduce a device consists of two kinds of liquid crystal polarization holograms (LCPHs) which can both identify the number of PTCs and values of initial polarization angle [4].

Figure 1(a) shows a schematic of the proposed device which can simultaneously detect the incidence of four types of VB. Geometric phase pattern of LCPH<sub>1</sub>/LCPH<sub>2</sub> is integrated the phase patterns of fork-shaped/normal polarization gratings whose grating vector are oriented to 0deg and 90deg directions as shown in Fig. 1(a). The details explanation of the operating principle of this device is provided in the reference literature [4]. Figure 1(b) shows numerically simulated diffraction patterns output from the proposed device in the cases where the four types of VB are individually incident. We found that only one spot has a Gaussian beam in each case, which means that the proposed device can identify the number of PTCs and values of initial polarization angle, respectively. To show the feasibility, we fabricate LCPHs by a photoalignment of liquid crystal and demonstrate its diffraction properties. Figure 1(c) illustrates a result of this experiment using 1550nm wavelength laser beam, and it shows good agreement with simulation.

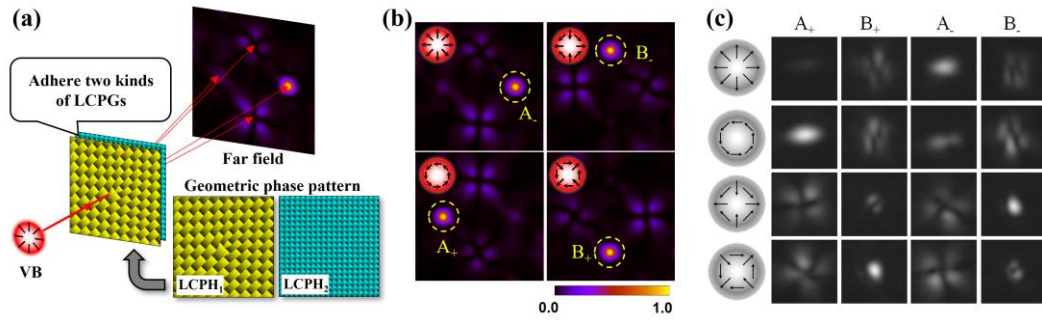


Figure 1. (a) Schematic of the proposed device. (b) Numerically simulated diffraction patterns. (c) A result of experimental demonstration.

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# **Azodye nanolayer liquid crystal photoalignment: applications in displays and photonics**

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Photoalignment and photopatterning has been proposed and studied for a long time [1]. Light is responsible for the delivery of energy as well as phase and polarization information to materials systems. It was shown that photoalignment liquid crystals by azodye nanolayers could provide high quality alignment of molecules in a liquid crystal (LC) cell. Over the past years, a lot of improvements and variations of the photoalignment and photopatterning technology has been made for photonics applications. In particular, the application of this technology to active optical elements in optical signal processing and communications is currently a hot topic in photonics research [2]. Sensors of external electric field, pressure and water and air velocity based on liquid crystal photonics devices can be very helpful for the indicators of the climate change.

We will demonstrate a physical model of photoalignment and photopatterning based on rotational diffusion in solid azodye nanolayers. We will also highlight the new applications of photoalignment and photopatterning in display and photonics such as: (i) fast high resolution LC display devices, such as field sequential color ferroelectric LCD; (ii) LC sensors; (iii) LC lenses; (iv) LC E-paper devices, including electrically and optically rewritable LC E-paper; (v) photo induced semiconductor quantum rods alignment for new LC display applications; (vi) 100% polarizers based on photoalignment; (vii) LC smart windows based on photopatterned diffraction structures; (viii) LC antenna elements with a voltage controllable frequency.

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# Polarization-independent diffractive waveplates

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The sign of geometrical phase changes with the handedness of circular polarized light. This feature has great applications, for example, allowing switching light propagation direction by switching its polarization. This opportunity first proposed in [1] is in the heart of emerging technologies of non-mechanical beam steering, augmented reality lenses with switchable focal length, etc.

However, this feature is a flaw for some other applications that need to control unpolarized light... As usual with liquid crystal devices, the phase of an unpolarized light could be modulated with the aid of a pair of components, each controlling the phase of one polarization state only. This is not so straightforward to realize for diffractive waveplates since the diffraction causes the beams to propagate into different directions! We will discuss the architectures of diffractive waveplates allowing undiffracted propagation of one of the circular polarization handedness due to so-called anti-Bragg condition while the light of orthogonal polarization undergoes full Bragg diffraction [2]. Experimental results demonstrating polarization-independent diffraction will be presented.

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# Perspective of broadband vectorial interferometry in photopatterning complex hierarchical structures

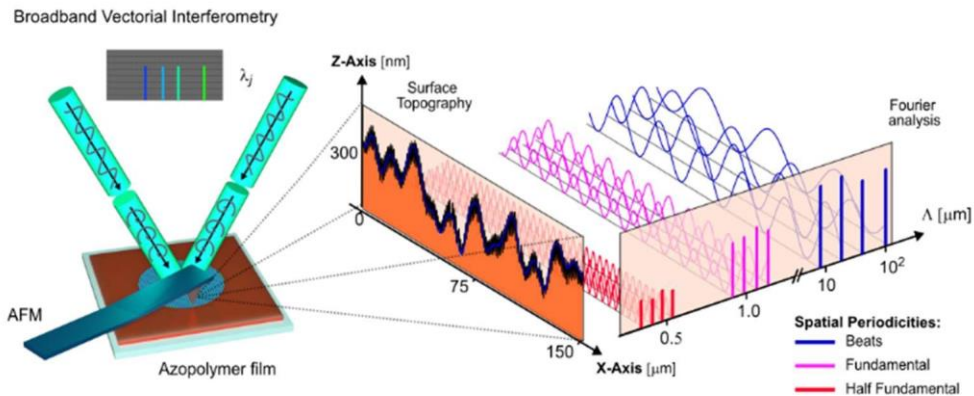
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The ever-growing demand for sub-micrometer structured surfaces with hierarchical features is driven by their efficient applications in augmented/virtual reality, display technologies, and Fourier optics. These surfaces also play a significant role in the development of multifunctional platforms for biomedicine, sensors, and security. Among all, Fourier surfaces represent still a challenge. Indeed, current technologies predominantly rely on multi-step processes that incorporate lithographic methods and various thermal, chemical, or electrical treatments. Unfortunately, these methods often suffer from losses attributed to their intrinsic binary design. Recently, a broadband laser composed by close and highly correlated lines allowed us for the simultaneous photo-inscription of extremely precise hierarchical Fourier surfaces via vectorial interference [1]. By adjusting parameters such as the number of lines, their amplitude, and polarization, polychromatic light enables the precise encoding of multiple sinusoidal profiles with nanometric spatial resolution. The occurrence of beat phenomena within the bulk of the photoresponsive medium [2] results in the formation of a previously unobserved hierarchical topographical structure. Such approach, unconventional for holographic techniques where monochromaticity of light is a key element, holds great promise for in-situ design of hierarchical Fourier surfaces and the scaling up of customized structured platforms. Moreover, it offers clear advantages in terms of scalability, reconfigurability, and tunability.



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# Mono-molecular photo alignment layer for liquid crystals using dip-coating

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Developing a reliable monomolecular alignment layer, which provides sufficient anchoring energy to the liquid crystals and ensures robust alignment, has been a long-standing challenge. A thin alignment layer is particularly beneficial for various applications, especially flexible substrates. However, the coating process often presents issues and consumes significant resources. In this presentation, we will introduce a new photo-alignment material specifically designed for liquid crystal optical devices. The alignment materials used have molecules containing at least one anchoring group and one  $\alpha,\beta$ -unsaturated carbonyl group. These groups can include derivatives of carboxylic acid, amides, nitriles, or ketones. When dipwashed onto the substrate surface of the transparent electrode (such as an ITO layer), the anchoring group (in this case, phosphonic acid) forms a single molecular self-assembled layer. These alignment layers exhibit excellent LC alignment when exposed to linearly polarized light. We were able to achieve variable pretilt angles ranging from near-vertical alignment ( $\sim 90^\circ$ ) to almost planar alignment ( $\sim 1-5^\circ$ ). The azimuthal and polar anchoring energy of these thin monomolecular alignment layers depends on the irradiation doses and possesses a wide processing window. We measured the VHR and RDC for these alignment materials as  $> 98\%$  and 3.8 mV, respectively, which is comparable with the conventional alignment layers. During this talk, we will provide an overview of the materials used for the alignment layer in liquid crystal devices, which offer a cost-effective solution with a simple fabrication process.

## Acknowledgments

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# Mechanically tunable geometrical phase optics

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Variety of all-optical, electro-optical, and opto-mechanical processes help controlling light beams very efficiently, while novel optical materials and technologies bring dramatically down the size and weight of optical and photonics components themselves. By that, the new, fourth-generation (4G) optics made a breakthrough in SWaP reduction [1] by overcoming fundamental drawbacks and limitations typical to other technologies of planar optics, including metasurfaces.

4G optical components, in essence diffractive waveplates (DWs) that spatially modulate so-called geometrical phase of light due to spatial modulation of their optical anisotropy axis, are distinguished by their continuous and smooth structure, with no thickness variations, no ridges nor nano-antennas.

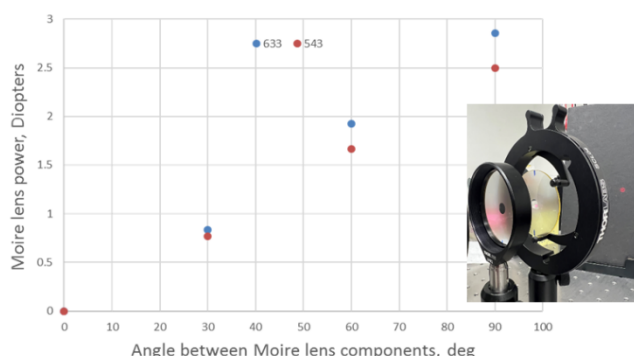


Figure 1. Measured Moire lens power as a function of azimuthal angle measured for two wavelengths, 633 nm and 532 nm.

In this paper, we discuss mechanical means for controlling light with combinations of 4G optical components due to shifts in their positions across the beam. Variable compound optics, for example lenses controlled by translation between the parts are well known as Lohman or Alvarez lenses [2,3], while the variable optics utilizing azimuthal rotations of the components with respect to each other are known as Moire optics [4]. The concepts can be extended to any other refractive device including prisms, a blazed grating, a spiral phase plate, etc. [5,6].

Technology applications span from ophthalmic lenses [7] and to variable aberration correctors for laser beams [8].

The fundamental advantage of 4G optics compared to other techniques of Alvarez or Moire systems consists in the feasibility of improving performance due to the opportunity of close positioning of the two flat, large aperture, and thin components of the system, and their light-weight making possible using piezo or other fast and low-torque micro-positioners for controls. Compared to non-mechanical systems, the absence of electrodes makes the mechanical systems highly transparent and resistant to high power laser beams such as in case with 4G Risley “prisms” [1,9].

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# Liquid Crystal Materials and Photoalignment Layers for High-Peak-Power Laser Applications

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Liquid crystal (LC) technology has experienced explosive growth from its humble origins as an unimportant laboratory curiosity discovered in 1888 to today's multibillion-dollar information display industry spawned by the invention of the twisted nematic LC display in the 1970s. The inherent anisotropy in the optical and physical properties of LCs, the ability to tune these properties through molecular design and chemical synthesis, the development of cost-effective and scalable methods for manufacturing LC optics with high optical quality and contrast, and their remarkable resistance to laser-induced damage have made LCs of considerable interest for numerous challenging applications in optics and photonics. A singularly significant example is the application of LCs in passive circular polarizers and wave plates for beamline polarization control in the 60-beam, 40-TW OMEGA Nd:glass laser system at the University of Rochester's Laboratory for Laser Energetics (LLE). This solid-state glass laser system, used to support the US Department of Energy's Inertial Confinement Fusion and High-Energy-Density physics research missions, contains more than 365 large-aperture (100- to 200-mm-diam) LLE-fabricated LC devices and represents a 30-year track record of proven performance for the technology. Many of these LC devices have been in continuous service on OMEGA for over 15 years without suffering degradation in their performance.

Following a brief overview of the history and events that led to the development of large-aperture, high-peak-power passive LC optics for OMEGA, research activities at LLE to design, develop and synthesize "next-generation" LC materials and photoswitchable alignment layers for high-peak-power passive and optically rewritable LC optics (tunable/switchable polarizers, optical modulators, polarization rotators/converters, vortex polarizers, laser beam deflectors, and beam amplitude/phase shapers) ranging from the near-UV to the near-IR will be described. Several new photoalignment polymers designed and synthesized at LLE that employ azobenzene, spiropyran, and spiroxazine chromophores on an acrylate backbone have shown *exceptional* 1053-nm laser-damage resistance, in some cases reaching 90 to 100 J/cm<sup>2</sup> at 1.4-ns pulse width. Optical patterning experiments conducted on these photoalignment materials have shown one example in which written optical patterns have remained stable for more than four months under ambient conditions.

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# Photoalignment and photopatterning by scanning wave photopolymerization

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Hierarchical control of two-dimensional (2D) molecular alignment patterns over large areas is essential for designing high-functional organic materials and devices.<sup>1,2</sup> However, even by the most potent current methods, dye molecules that discolor and destabilize the materials must be doped in, complicating the process. We present a dye-free alignment patterning technique, based on a scanning wave photopolymerization (SWaP) concept, that achieves a spatial light-triggered mass flow to direct molecular order using scanning light to propagate the wavefront.<sup>2</sup> This enables one to generate macroscopic, arbitrary 2D alignment patterns in various optically transparent polymer films from various polymerizable mesogens with high birefringence by single-step photopolymerization without alignment layers or polarized light sources.<sup>2,3</sup> SWaP also produces microscale well and canal structures in the polymer surface.<sup>4</sup> This dye-free inscription of microscopic, complex alignment patterns over large areas provides a new pathway for designing higher-performance optical and mechanical devices.

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# **Photoaligned and photoaligned graphene electrode for polymer-sustained liquid crystal devices**

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In this presentation, we demonstrate polymer-sustained alignment of liquid crystal display (LCD) prototype device manufactured using graphene as a transparent conductive electrode. Electro-optical performance of graphene-based VA, IPS and FFS prototype LCDs will be comparable with the optical transmittance on ITO electrode cells. In short, graphene electrode is demonstrated to be compatible with processing standards of modern LCD industry and has a great potential for liquid crystal displays and other electro-optical and optical applications.



# Digital holographic microscopy for photolithographic surface patterning

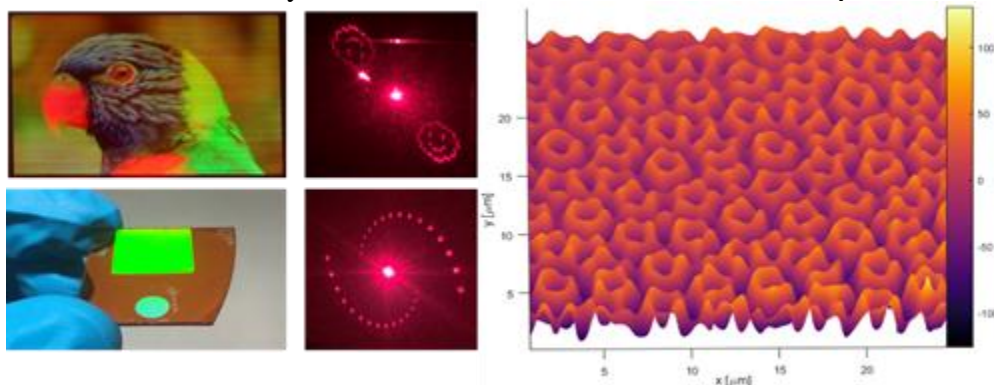
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In contrast to the advances in miniaturization of electronic components, optical components are often still very bulky. Control over material properties such as topography and refractive index is the key element for miniaturized optics, the fourth generation optics [1]. We show that surface patterning of photochromic materials using a digital holographic microscope (DHM) with integrated laser interference system provides a path to rapid fabrication of diffractive optical elements (DOEs) [2]. The light-induced mass-migration of molecules, such as azobenzene, enables the inscription of topographies in a one-step photolithographic process, which poses a challenge to existing microfabrication methods. This method also enables real-time observation of surface modifications, thus the study and control of the material and inscribed patterns.



In this contribution, we show how thin films containing azobenzene can be patterned with DHM by superimposing surface relief gratings (SRGs). Computer-controlled scanning of the sample position and tuning of interference conditions enable patterning of large areas with customized topographic structures. We show how such light-configurable topographies could benefit new metamaterials and applications such as waveguide couplers in near-eye displays. As an example of control, we present tailored topographies with highly controlled diffraction points and a true-color hologram generated by a precise superposition of SRGs. In the latter, we combine SRGs with three different periods to diffract the main colors for additive color mixing. The topographies are then stitched pixel by pixel according to a reference image to form a large hologram.

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# Ultrafast switching of infrared liquid crystal diffractive waveplates

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Switchable geometrical phase optics is revolutionizing critical modern technologies such as AR/VR and adaptive ophthalmic lenses, LiDARs for auto-navigation, and optical communication [1,2]. Those and other applications require fast switching between optical states of different focal lengths, between different line of sights and field of regards, and beam shape. Liquid crystals are the materials of choice for many photonics devices due to their high electro-optical coefficients. However, optical switching cycle of LCs is typically limited by relaxation time that increases proportional to the square of cell thickness,  $t \sim L^2$ . This conflicts with the requirement of having the LC layer thickness large enough to meet half-wave retardation condition for high efficiency of diffractive waveplates, particularly, for infrared wavelengths.

We thus experimented with LCs known to provide high speed switching – ferroelectric LCs (FLCs) and deformed helix ferroelectric LCs (DH-FLC) showing that:

- (a) these materials are capable of photoaligning at high spatial frequencies for fabrication of diffractive waveplates;
- (b) they are capable of maintaining the structure of diffractive waveplates even for thickness values producing half-wave retardation for infrared wavelengths; and
- (c) switching at sub millisecond scale can be obtained with low voltages.

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# Hydrazone-based functional materials

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For the past few years, we have been developing structurally simple, easy to make, modular, and tunable hydrazone-based adaptive materials [1]. This presentation will focus on our recent advances (Figure 1) with the use of photochromic hydrazones in manipulating the properties of liquid crystals [2] and liquid crystalline elastomers [3].

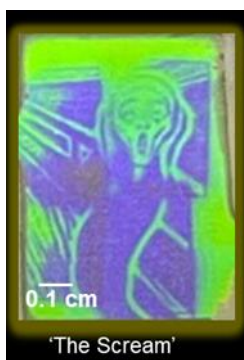


Figure 1. Edvard Munch's "The Scream" patterned in a LC film.

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# High refractive index azomaterials through hierarchical supramolecular chemistry

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The all-optical generation and reshaping of diffractive optical elements (DOE) on thin azomaterial films with precise morphology and functional reliability demonstrate possibilities not available through traditional photolithographic methods [1]. A successful approach may rely on optimizing the light pattern and illumination strategy for inducing surface structuring through mass transport. Additionally, it may depend on a material design that is both flexible and efficient, allowing for easy modification of the material's response and properties as needed. In this context, supramolecular chemistry offers straightforward synthetic procedures and a diverse range of non-covalent interactions with varying strengths to adjust material response. Additionally, molecular components can be substituted easily, and the composition can be optimized with minimal effort [2].

The strong molecular recognition of the 2-aminopyrimidine/carboxylic acid supramolecular synthon, supported by multiple hydrogen bonding, was initially introduced as a design element to build polyacrylic acid-based supramolecular azopolymers. These polymers demonstrate excellent stability towards azobenzene loading, greater mass transport efficiency compared to single H-bonding systems, and resilience towards azo removal after patterning. Subsequently, the multiple hydrogen bonding approach was refined and incorporated into the development of photostructurable high refractive index azomaterials. The goal was to demonstrate the feasibility of creating a thin film diffractive device with a specific target diffraction efficiency through the use of lower surface relief gratings, ultimately reducing the exposure time required for photopatterning and overall thickness.

To this end, a supramolecular branched system [3] has been developed based on H-bonding, and functionalized with stronger metal-carboxylate type interactions. This generates a combination of orthogonal and hierarchically ordered interactions between low molecular weight building blocks. The resulting material's molecular structure can be modified and its photoresponsiveness can be changed by simply adjusting the composition of the building blocks in solution. Thanks to this design, it was possible to synthesize molecular components with a high density of sulfur and rigid groups exhibiting high molar refractivity, including heterocycles comprising -C-S(II)-C-, -C=N-N-, and -C=N-C- bonds. These components were used to develop materials with a refractive index greater than 1.78, high solubility, processability, and tunable photopatterning efficiency, enabling optimal control of SRG morphology. Holographic lenses with the same diffraction efficiency as a standard acrylate azopolymer were easily produced with a lower profile.

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# Review of modeling efforts on bulk and surface restructuring of glassy azopolymers

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Polymers that can change their molecular architecture under such an external stimulus as light have attracted great attention in the scientific community. Particularly known are photosensitive azopolymers, which have many applications in different fields due to their high versatility. In modern engineering, a special interest has been evoked by a unique opportunity to control the deformations of glassy azopolymer films depending on the light irradiation pattern. Multiple theoretical interpretations have been developed to explain the surface restructuring, but only a few of them consider the features of underlying molecular architecture.

We have been also working on this problem by pursuing the orientation approach, which establishes a clear relation between the light characteristics and the molecular properties of azo-containing materials. In particular, an effective potential was derived that causes biaxial alignment of the chromophores under the elliptically polarized light [1]. Furthermore, we provided crucial evidence that reorientation of polymer backbones along the light polarization should be the main reason for bulk and surface deformations in azopolymer films [2, 3].

In this presentation, modeling efforts on bulk and surface restructuring under interference pattern irradiation will be reviewed. It is planned to explain the influence of initial polymer orientation and light-induced softening [4], and to highlight the importance of self-induced polarization rotation in complex irradiation patterns. Overall, a versatile modeling approach for practical applications in modern optics and photonics is being developed in close collaboration with experimental groups.

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# Birefringence control of photoalignable liquid crystalline polymers and application to polarization optical devices

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Photoalignment is a promising technique for fabricating birefringent optical films and polarization sensitive diffraction devices. Many types of photosensitive materials have been developed for the photoalignment. In these cases, generated birefringence of the photoaligned film is one of the important issues to fabricate thinner optical birefringent devices. Among them, we have carried out systematic study of photoalignable liquid crystalline polymers (PLCPs), which directly attain molecularly oriented structure of their own by the use of photo-exposure and thermally stimulated self-organization [1-3]. Herein, we introduce our PLCPs which include high photoreactivity and controllability of the generated birefringence and their application to optical devices.

We previously reported that a PLCP with 4-oxycinnamic acid (CA) side groups indicated a high photosensitivity and high molecular photo-alignability, [2] but the generated birefringence was below 0.14. In contrast, a PLCP with *N*-benzylideneaniline (NBA) derivative side groups showed photoinduced molecular orientation, and *in situ* decomposition of NBA followed by recondensation with higher inherent birefringence molecules attained molecularly oriented film with high birefringence ( $\Delta n \sim 0.4$ ) [3,4]. However, required exposure energy to orient the NBA side moieties exceeds 1 J/cm<sup>2</sup>.

In this context, we synthesized new PLCPs containing CA, 4-oxybenzoic acid (BA), and 4-(4'-oxybenzoyloxy)benzaldehyde (PA) side groups. Thermally stimulated photoinduced molecular reorientation is attained with low exposure energy (< 50 mJ/cm<sup>2</sup>) similarly as the previously reported PLCPs with CA side groups [2]. Then, *in situ* condensation of oriented PA side groups with high-inherent birefringent phenylamine derivatives results in a formation of oriented NBA-derivative mesogenic side groups with high birefringence (Fig. 1). The generated birefringence of the oriented film improved from 0.12 to 0.4 when the oriented fluorene is introduced.

Using our PLCP films, polarization optical elements such as polarization gratings (PG) and polarization Fresnel lenses have been fabricated (Fig. 2), where the diffraction efficiency of 100 % is achieved when using thinner film (< 2  $\mu$ m) [5].

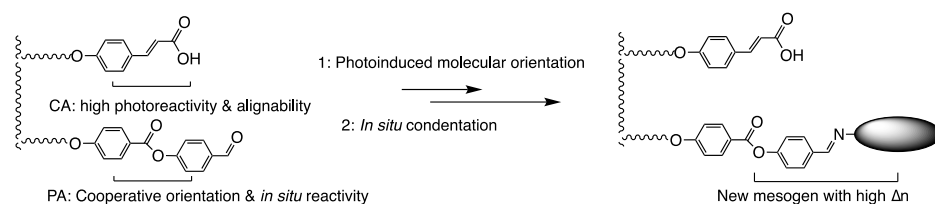


Figure 1. PLCPs and birefringence control of this study

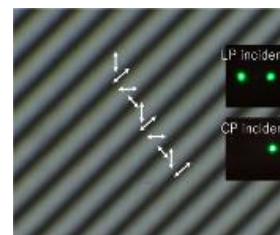


Figure 2. POM photograph of PG using PLCP thin film. Insets show 1<sup>st</sup> order diffraction.

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# Electropolymerization process as a new tool to obtain high ordered alignment layers

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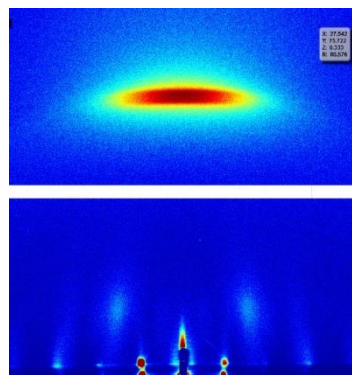
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We are studying a new electropolymerization process with calamitic liquid crystals [1-4], which is carried out in the absence of solvents or added electrolytes. The process is achieved by applying a direct electric field to monomers with liquid crystalline properties in sandwich glass cells covered with ITO electrodes. The major advantage of this process is the ability to obtain highly oriented films of side chain liquid crystalline polymers (SC-LCP) when using cells with planar alignment.

Based on a homologous series of 12 acrylates (A6En, with n=1-12) [2] we explore the occurrence of high order smectic phases after the polymerization process. The results shown that electropolymerized A6E4 monomer yielded the best aligned smectic B phase at room temperature, which was selected for this study. The obtained data as well as the SAXS characterization will be presented.

GISAX patterns for EPA6E4 at room temperature, in the SmB.



Our polymerization process was also performed by UV to compare them. Photoalignment and electropolymerization are two distinct processes, but they can be used together in certain applications, particularly in the field of liquid crystal devices and optoelectronics. They are separate techniques that can be synergistically employed in specific applications, especially in the development of advanced liquid crystal devices and optoelectronic systems. The precise implementation and potential benefits depend on the desired functionality of the final device.

**Acknowledgments:** Fondecup project EQM200266.

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# **Spectrally selective diffractive waveplates**

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Waveplates can be combined to produce practically any desired retardation spectra [1,2]. Typically, this opportunity is made possible by combinations of optical axis alignment angles for waveplates of different dispersion spectra and retardation to obtain achromatic half-wave retardation plates and quarter-waveplates. Liquid crystals introduce an additional control opportunity for spectral and polarization engineering of diffractive waveplates related with the opportunity of twisting orientation of their optical anisotropy axis and combining such twisted and uniform layers, for example, for obtaining broadband waveplates [3].

Due to the waveplate nature of geometrical phase optics as first highlighted in [4], the new, 4th generation of planar lenses, prisms, etc., can be designed to perform differently for different spectral ranges, for example, focusing light of certain wavelengths while not affecting propagation of light of other wavelengths. We will discuss, additionally, polarization engineering with combination of waveplates, particularly, switchable waveplates, making possible designing switchable or spectrally tunable filters and a wide variety of optical components.

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# Single-molecule fluorescence and nonclassical light from photoalignment agents

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Photoalignment agents have been successfully employed to align *single* molecules of dyes with dipole moments [1, 2] and *single* quantum dots-in-rods [3] in desired direction. This alignment provided definite linear polarization of fluorescence from each single emitter [1-3] that is important for quantum key distribution based on single (antibunched) photons. In this paper we present our studies of two photoalignment agents by a confocal fluorescence microscopy at a single-photon level. Two polymer solutions were spin-coated on cover-glass slips: (1) a Staralign-2100 linearly photopolymerizable polymer (Rolic Technologies Ltd.) and (2) PAAD-22 (BEAM, Co). Figure 1, LEFT shows typical single-emitter behavior with stripes and semicircles during a raster scan of a Staralign-coated sample. Single-emitter behavior was also observed with a sample with PAAD-22 (CENTER). Figure 1 shows not only single-emitter behavior, but even emission of nonclassical light. The histogram (RIGHT) shows a dip at zero interphoton time indicating photon antibunching. Fluorescence lifetime from these measurements (at level  $1/e$ ) was evaluated as 2.1 ns from fitting. The data was collected from a single emitter marked by a cross at the bottom left corner of a central confocal fluorescence micrograph. For single-photon device fabrication these single emitting centers should be bleached by additional exposure with linearly polarized UV light to avoid misinterpretation in characterization of single-photon sources based on single emitters aligned with these agents.

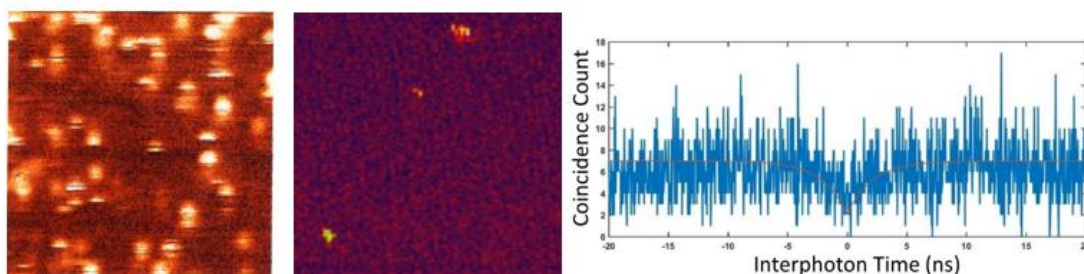


Figure 1. Confocal fluorescence microscope raster scan micrographs of samples coated with Staralign-2100 (LEFT,  $10\ \mu\text{m} \times 10\ \mu\text{m}$ , 532 nm laser excitation) and PAAS-22 (CENTER,  $40\ \mu\text{m} \times 40\ \mu\text{m}$ , 633 nm laser excitation). RIGHT: Photon antibunching histogram for a single emitter of a left bottom corner of a PAAS-22 micrograph. A dip at zero interphoton time indicates photon antibunching.

The authors thank K.L. Marshall for providing photoalignment materials and a clean room facility of the Laboratory for Laser Energetics, M. Johnston and O. Ta for assistance.

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# **Intuitive understanding of the connection between polarization volume holograms and Pancharatnam phase device**

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Two classes of traditional diffractive devices are Bragg and Rahman-Nath diffractors. It is well known that these two classes of diffractive devices are similar in their physical properties, but have quite different optical properties.

Two classes of diffractive devices that are analogous to the above, are polarization volume holograms (PVHs) and Pancharatnam Phase devices (PPDs). A familiar form of a PVH are cholesteric liquid crystals when fabricated as a polarization and wavelength sensitive reflector; and a familiar form of a PPD is a device that whose optic axis spirals about a layer normal and can deflect transmitted light with high efficiency. While quite different optical effects, it is observed that by varying the physical properties of a PPD device its optical properties can continuously change to be that of a PVH.

While others working in the area of optics of polarization based diffraction have noticed the apparent analogous behavior of a PVH and a Bragg diffractor; and a PPD and a Rahman-Nath diffractor, an intuitive explanation of the transition between PPD and PVH behavior has been lacking.

The optical properties of PVH and PPD devices can be calculated with the rigorous coupled wave analysis method, and the finite difference time domain method. Both methods provide highly accurate modeling of the optical properties of PVH and PPD devices, and show the continuous evolution of one type to the other. However, these numerical methods do not allow for a simple explanation of the nature of the transition. In other words they cannot give a straight, simple answer as to why a PPD becomes a PVH as its thickness is changed.

Following a beam propagation approach that had been used to previously to illuminate the distinction between a Bragg and Raman-Nath behavior of isotropic diffractors by Gaylord and Moharam [“Planar Dielectric Grating Diffraction Theories, Appl. Phys. B 28, 1-14 (1982)], we developed a beam propagation method for birefringent materials that was able to provide a simple, and clear, explanation of the underlying distinction between PPD and PVH devices.

The result is that the distinction between PPD and PVH devices is related to the relative phase of the one of the diffracted orders of a PPD device to the incident beam, as light propagates through the birefringent material. Specifically if the phase of both diffracted orders maintain a near constant phase relationship with each other, energy is transferred between the incident beam direction and that of the diffracted orders. However if the angle of the incident beam relative to a normal to the axis of index variation is quite different from one of the two diffractive orders, their relative phase can change rapidly and no energy is exchanged between that diffracted order and the incident beam. Therefore, in a PVH, only one diffracted order is observed.

# Achromatic planar liquid-crystal optics

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Liquid crystal has been widely used for display applications, covering from microdisplay light engines for VR and AR to large-screen TVs. Besides displays, liquid crystal also exhibits great potential for diffractive optical elements. Planar liquid-crystal optics offers several advantages, such as high diffraction efficiency, easy fabrication, polarization selectivity, ultrathin formfactor, and dynamic switching, making them a promising candidate for AR/VR applications. However, its severe chromatic aberrations impose a big challenge for full-color display applications. To overcome this longstanding chromatic aberration issue, we demonstrate an achromatic planar liquid-crystal device by stacking three planar liquid crystal optical elements with specifically designed spectral response and polarization selectivity. The concept is validated by both simulations and experiments. Potential applications for VR-enabled metaverse, spatial computing, and digital twins that have found widespread applications in smart tourism, smart education, smart healthcare, smart manufacturing, and smart construction are foreseeable.

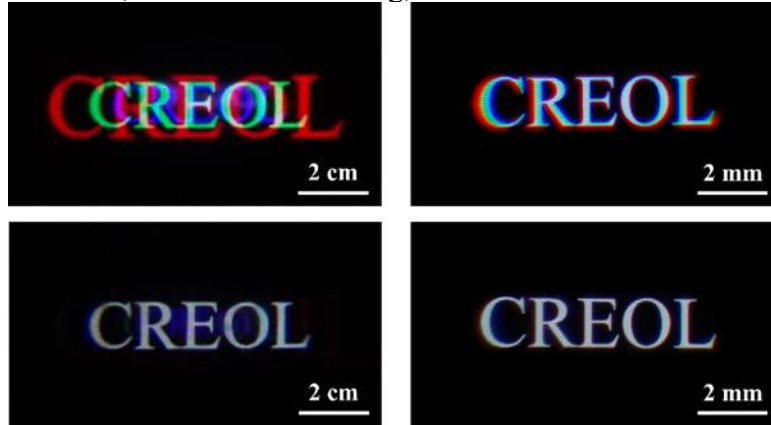


Figure 1. Image performances of conventional planar LC lens (top) and achromatic LC lens (bottom).

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# Cholesteric liquid crystals with controlled molecular orientation for flexible optical sensors

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A programmable liquid crystal elastomer (LCE), with controlled and patterned molecular orientation, shows stimuli-responsive behavior and enables to realize high-performance applications such as reconfigurable photonics/electronics, soft robotics, and flexible sensors [1,2]. LCEs immediately change their macroscopic shape and molecular orientation as well as materials properties when an external stimulus is applied to them. However, the recovery response after stimulus removal is relatively slow. Recently, we have proposed a multilayered concept in which an LCE film is sandwiched between other materials showing a desired mechanoresponsive behavior. This approach allows us to tune the recovery response of both the macroscopic deformation and molecular orientation change even without requiring molecular structure modification of LCEs [3]. Here, as an LCE film, we utilized cholesteric liquid crystals (CLCs), which intrinsically form a helically twisted molecular orientation with the spatial distribution of refractive indices. The helical structure of CLCs is extremely sensitive to external stimuli; thus, CLCs can quantify and visualize stimuli by measuring the wavelength of reflected light (proportional to the helical pitch), serving as flexible optical sensors [2–4]. Notably, changing the layering materials alters the molecular-level recovery response. Therefore, we have demonstrated highly sensitive, stretchable mechano-optical sensors with fast and/or ultraslow recovery response, quantitatively detect an applied strain with high spatiotemporal resolution, even by using a commercially available camera.

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# **Approaches to controlling the self-assembly of liquid crystals in the bulk**

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Photoalignment is a highly adaptable technique for manipulating the self-assembly of liquid crystals by subjecting photosensitive surface coatings to polarized light. However, the inherent dependence on surface interactions generally limits this technique to 2-dimensional directed self-assembly patterns. In order to realize 3-dimensional control of the self-assembly of liquid crystals new techniques are required. Towards that aim, we will present our work investigating alternative approaches to affecting the self-assembly of liquid crystals in the bulk, including so-called bulk-photoalignment, as well as creating polymeric 3-D alignment scaffolds through polymer-stabilization combined with solvent exchange.

# **Application of the fluctuation microscopy**

## **- Direct imaging of the spatial distribution of elastic constant, anchoring condition, slippery interface...**

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The fluctuation microscope we are developing is a new microscope that directly visualizes the "dynamic heterogeneity" of inhomogeneous motion of internal degrees of freedom in a material as a two-dimensional image. Figure 1 shows a photograph of the fluctuation microscope we have developed. Based on an inverted microscope, a high-brightness and high-stability laser (532nm) for illumination is introduced. The image of the scattered light is recorded as frame data by a high-speed camera (up to 1,500,000 f/s), and an autocorrelation function for each pixel is calculated by a high-speed algorithm to obtain a two-dimensional image of the dynamic heterogeneity. Furthermore, by analyzing multiple consecutive frames, the time variation of dynamic inhomogeneity can be recorded as a movie. Figure 2 shows an actual observation of the self-organized dynamic heterogeneity (256x128 pixel) created near the glass transition point of a liquid crystal gel (25°C). The colors represent the variation of the relaxation time of the sample at each point, with warmer colors indicating slower dynamics in motion and the presence of dynamic inhomogeneities ranging from 100 ms to several seconds.



Fig.1 Photograph of fluctuation microscope.

The developed fluctuation microscope is completely different from various types of conventional optical microscopes and confocal microscopes in that it can obtain two-dimensional images of the interior of a sample by contrasting the dynamic heterogeneity inside the sample, which cannot be effectively observed with ordinary microscopes, such as in isotropic or in transparent solutions.

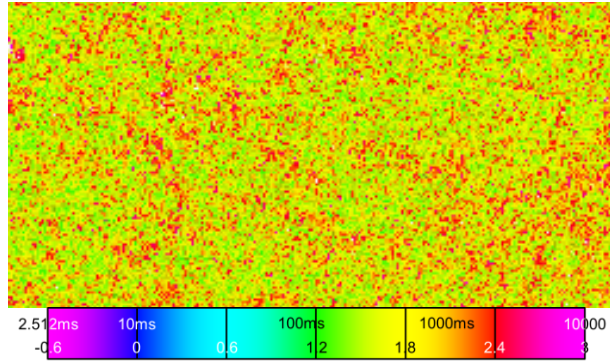


Fig.2 2D Image of dynamic heterogeneity observed near glass transition of LC gel.

Therefore, it is very useful for the study of pre-transitional phenomena near phase transitions or separations, sol-gel transitions, liquid-glass transitions, and so on. In addition, in-situ observation under external force can be performed, making it possible to study mechanical phenomena and fracture processes under an applied mechanical field, or switching behaviors and elastic constant distribution under an applied electric field. The interfacial effects can also be studied, such as slippery interfaces and anchoring transitions. Thus, the fluctuation microscope can be widely used for various material research.

# Surface and bulk photo patterning of azobenzene molecules

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Azobenzene molecules are very efficient light-to-mechanical energy convertors [1]. Huge energy densities (eV at nano metric scale) can be obtained and manipulated by using light and particularly its polarization.

In this talk, we shall review the behavior of such molecules in various host media, including liquid crystals, polymer stabilized and elastomer liquid crystals, thin solid surfaces (as command layers) as well as polymers and reactive mesogens. Various applications of such guest-host systems will be presented. One of the examples is the “truly holographic recording” that allows the storage and reconstruction of light’s polarization state, Fig.1, [2].

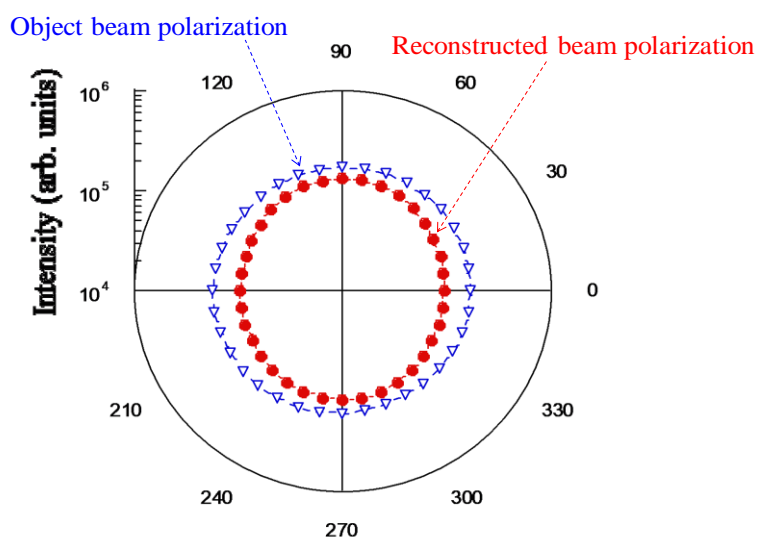


Figure 1. Demonstration of truly holographic recording that is allowing also the reconstruction of the polarization state of light.

Other applications, such as rocking filters [3], asymmetric diffractive optical elements [4] and guided structures [5] will be presented also.

Finally, we shall address an emerging area of their application that is the the photo dynamic therapy. We shall show briefly how we use azobenzene molecules to control the motility of bacteria [6].

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# Paving the pathway to the holy grail of optics

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The future of optics is being shaped by a new technology uniquely capable of providing high efficiency over broad bands of spectra in thin layers and in large aperture sizes – all beyond capabilities of metasurface optics [1-5]. The technology of diffractive waveplates using liquid crystalline materials to modulate geometrical phase has proven to not only possess any of those features but even the combination of those features, furthermore offering opportunities for switching and tuning of optical properties and functions in thin films. We discuss the opportunities and demonstrate the feasibility of overcoming polarization dependence for broad spectral bandwidths maintaining high efficiency. The materials, designs, and manufacturing techniques used in the development bring us closer to the elusive goal of creating a single, flat surface that can replace the complex and bulky optics used in many applications today.

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# Area 3D printing with liquid crystal light valves and laser damage to photo and rubbed alignment materials

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The Selective Laser Melting (SLM) method of 3D printing, whereby 3D metal objects can be printed from a digital file with unprecedented design flexibility, is spurring manufacturing innovations in medical, automotive, aerospace and consumer technology industries. Because SLM is based on scanning a laser beam spot over successive layers of metal powder, the process is relatively slow compared to most traditional metal manufacturing methods such as casting and forging (hours to days), thus limiting wider spread use. Seurat Technologies has developed the Area Printing™ method for 3D metal printing. This method is similar to photolithography but uses an optically-addressable light valve (OALV) as the photomask, to selectively melt and sinter areas in a powder bed, and print large areas for tiling each layer. We pattern near infrared light by imaging blue light onto transmissive OALVs, which consists of polarization steering nematic liquid crystal sandwiched between a photoconductor and a transparent conductor for high resolution reprogrammable masking.

We report on materials and fabrication considerations to mitigate high fluence laser induced damage on liquid crystal materials including rubbed polyimide, rubbed nylon, and photoaligned azobenzene. Photoalignment is, in principle, more laser-damage resistant because of the no-touch processing in the clear aperture.<sup>1</sup> Laser-damage thresholds depend substantially on the presence of aligned liquid crystal material; substrates with alignment materials but not formed into a liquid crystal cell were not damaged at all. We collected statistically significant data for each material in a liquid crystal cell configuration and found that nylon outperformed photoalignment, which outperformed polyimide. Further, we compared the polarization dependence of the damage threshold with respect to the alignment direction in ordinary and extraordinary modes at a near-infrared wavelength. Studies on polarization dependence on laser-damage of liquid crystals with alignment layers have been very limited,<sup>2</sup> and none include photoalignment. Polarization effects were smaller than the choice of material and only the photoalignment material demonstrated polarization sensitivity at our statistical power level. These data inform the design of high-power beam-shaping devices for applications in fusion, 3-D printing, and directed-energy applications.

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# 3D morphing of azopolymer-based microstructures: in between top-down and bottom-up approach

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Surfaces endowed with three-dimensional (3D) microstructures have captured the interest of the scientific community because of the possibility to introduce intriguing properties that are not present in their planar counterparts. They are extensively used in various fields such as photonics, surface wettability, bio-interfaces, and energy harvesting and storage [1]. Generally, there are two types of techniques for 3D microfabrication: the top-down and the bottom-up approach. On one hand, the top-down approach, such as the conventional photolithography and the imprinting lithography, is well-known for its fast and large-scale production. However, these methods require post processing steps, and often result in 3D structures with limited complexity. On the other hand, while the bottom-up approaches such as direct laser writing and 3D printing bring complexity and accuracy to the 3D structures, they suffer from the slow speed, high-cost procedure, and sophisticated tools. Recently, the direct and reversible light-induced mass-transport in azomaterials has been explored as a very powerful alternative.

To create unique and complex 3D microstructures using azopolymers, an initial 3D microstructure is usually fabricated and subsequently exposed to light in order to shape it into various 3D architectures. The photo-driven deformation of azopolymer-based microstructures exhibits a distinctive mechanism in which the initial structure morphs without requiring removal of any material, as in the top-down approach, or addition of any new material, as in the bottom-up approach.

Here, we use the light-induced mass transport in azopolymer to create complex and anisotropic textures [3]. By exploiting the polarization-driven transport of the material, a 2D square array of micropillars was reshaped into a grating-like structure with a programmable amplitude, height, and orientation controlling the polarization direction and exposure dose. In addition, the approach can be generalized by using the different light penetration depth in azopolymer volume at different light wavelengths [4]. By exposing the azo microvolumes with light of different wavelengths, we fabricated different 3D microstructures from the same original with tunable structure-dependent properties, such as wettability, with a single exposure with a low-intensity light and simple set-up.

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# Spectroscopic studies of photoinduced interference colors

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Our investigation of the mechanism of the phenomenon of vector polyphotochromism led to the opinion that it has an interference nature. This paper presents the experimental data on the correspondence of this effect with the manifestation of the photoelasticity of the polymer component of the material. We verified the accordance of the transmission spectra of polarized light on the one hand of a photoactivated polarization-sensitive material and on the other hand under the influence of a mechanical stress. The profiles of the obtained polarization spectra match each other.

Earlier, the photoinduced interference fringe pattern was detected in our laboratory, which was preliminarily named as vector polyphotochromism [1]. This kind of photochromism manifests itself in the form of the possibility of tuning the polarization spectral characteristics over the entire visible range. We continue to investigate this phenomenon and are studying the mechanism of its formation.

The light-induced effect was found in some highly efficient polarization-sensitive polymeric materials with azocomponents as vectorial selective receivers of the recording light. The results depend on the illumination dose of the linearly polarized actinic light. When radiant exposure exceeds certain value, a sharp change in the transmission spectrum of the probing linearly polarized nonactinic beam is observed after analyzer. The effect is an interference nature and depends on the path difference between the ordinary and extraordinary rays in material with photoinduced anisotropy. It is found that in this case birefringence has made the main contribution in photoanisotropy [2].

It is noteworthy that when material is exposed to the actinic light, its spectral properties change in the same sequence as observed in the Michel-Lévy interference color chart [3], [4]. Due to interference between of the ordinary and extraordinary rays within optically anisotropic medium a fringe pattern develops along with the irradiation of polarization-sensitive material. It is important to note that at the beginning of the sequence of changing color of photochromic area is the same one that the Michel-Lévy interference color chart starts. Further development of polyphotochromism is in a precise sequence in accordance with this chart. Thus, it became necessary to check the accordance of the transmission spectra of polarized light in one case of a photoactivated material and on the other hand under the influence of a mechanical stress.

**Conclusion:** The observed behavior of the material in the same way, both under the influence of light and under mechanical action, suggests that the phenomenon of the vector polyphotochromism is based on the photoelasticity of the polymer component of the material.

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# Broadband orbital angular momentum shaping from bragg-berry cavities

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Liquid crystal materials are widely used for the modulation of light beams because of their high optical transparency, large anisotropy, and flexible orientational structure. Cholesteric liquid crystals (CLCs) possess a supramolecular spiral structure forming Bragg reflection for circular polarized light. Light reflected from CLC cells acquires the Bragg-Berry phase modulation: the geometrical phase according to the patterned photoalignment on the cell substrate [1]. This effect is used to create broadband reflecting phase plates for the generation of optical vortices, which are light beams with angular orbital momentum.

The combination of the CLC q-plate and the ordinary mirror allows one to obtain spin-to-orbital angular momentum mapping caused by the interference of two optical vortices with opposite charge signs [2].

To further enhance this scheme, we have introduced a tunable retarder between the Bragg-Berry q-plate and the mirror. This retarder serves as an anisotropic defect layer, allowing multiple reflections in the CLC-mirror cavity. As a result, a pronounced modification of the angular distributions of phase and amplitude profiles of the reflected light beam is obtained. In particular, by adjusting the retardance of the defect layer, we can achieve optical beams with an azimuthal singularity in the phase gradient distribution, which can be considered as a number of vortex components with high charges. These beams are of great interest because of the phase divergence in the form of linear singularity, which, to the best of our knowledge, has not been previously observed. Both experimental results and theoretical considerations are presented.

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# Morphology-related diffraction behavior of surface relief gratings on azopolymer films

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The superposition of diffraction gratings with different geometries in a single and multiplexed grating gives an enhanced control over the distribution of the light propagating far from it. By engineering the geometric parameters (periodicity, orientation, groove depth, and groove profile) of the superimposed gratings, the diffraction angle and the relative efficiencies for each diffraction order can be controlled. A similar device obtained by combining sinusoidal gratings, is referred to as Optical Fourier Surface (OFS). A grating component for an OFS can potentially be fabricated by standard interference lithography by exposing a photoresist to a sinusoidal fringe pattern. However, grating multiplexing using such a method is either impossible or it requires multiple and challenging lithographic steps. Conversely, by using azomaterials films, the diffraction gratings can be fabricated directly as surface reliefs (known as Surface Relief Gratings – SRGs), eliminating several subsequent steps required by the photoresist [1]. The use of azopolymers as materials for diffractive components is particularly attractive due to the possibility of making the grating reconfigurable and then usable for multistep patterning processes that combine the pre-existing texture with a new one. Multiplexed surfaces can then be designed as complex diffractive gratings [2]. However, describing their diffraction behavior requires an accurate analysis of the surface morphology which can deviate significantly from the ideal sinusoidal pattern of the illuminating interferogram.

Here, we exploit the dynamic surface relief on the azopolymer to design and characterize multiplexed gratings via sequential two-beam interference lithography. A large-scale three-component 1D multiplexed grating, capable of diffracting polychromatic light with controllable relative color intensities, is fabricated using a switchable interference setup able to inscribe reprogrammable gratings of different periodicities on the azopolymer surface [3]. In addition, we extensively investigate the relationship between the surface profile and the diffraction behavior of interference-induced SRGs in azopolymer. Scalar diffraction theory, combined with the Fourier decomposition of the SRG profile, is used to fully describe the diffraction behavior of engineered high-performance diffraction gratings [4].

Our results could be used to accurately describe the diffraction behavior of SRGs and to engineer multiplexed diffraction gratings, to open new paths for future flat and lightweight optical components.

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# Studies of soft-bonded azo dye materials for sunlight-triggered controlled photo-dis-assembly

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Photo-reversible azobenzene (azo) and polymer self-assembled materials have received great interest for study recently, for applications as optical devices, photo-actuators, bio-compatible active interfaces, and in drug delivery and other reversible disassembly/reassembly systems. The goal of our research here is to develop novel sustainable, low-toxicity, and bio-compatible photo-reversible materials based on natural polymers harvested from highly abundant biobased sources such as cellulose, chitosan, and alginate, which offer competitive mechanical and physical properties compared to traditional oil-based polymers. We describe here a new process of preparing photo-reversible materials by soft-bonding the natural polymers with small bi-functional aqueous photo-switchable azo dyes as reversible cross-linkers. These two water-soluble components can be assembled onto a substrate into multi-layers held together by electrostatic interactions, or as polyelectrolyte complex bulk materials, both of which become stable to water. On triggering the characteristic *trans-cis* isomerization of azo dyes however, a controlled disassembly of the material can be preferentially induced using low intensities of visible light while being gently washed, returning the material back to its water-soluble components, fully reversibly with 100% recovery of the undegraded starting materials.

Physical chemistry characterization of the nature of the soft-bonding, and computational treatment of the isomerization mechanism is challenging however, and is thus far lacking in the materials development cycle, yet is crucial for rational development of these next generation photo-materials. We present here new research towards elucidating the structure of the soft-bonding self-assembly of these materials, and a density functional treatment of the isomerization energetics on irradiation, and thus insight into the mechanism of the controlled disassembly. Solid State NMR methods in particular, hold great promise for shedding light on both the structure, and function, of these new light-reversible soft-bonded assemblies.

## References

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