

Light-driven components and systems made of smart polymer materials

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Abstract

Liquid crystal elastomers (LCEs) are a group of smart materials that can reversibly change their shape, color, refractive index, and other physical properties under external stimuli. The properties of these materials are closely tied to their molecular alignment. Research on LCEs includes the synthesis of new compounds that enable materials to respond to different ranges of stimuli or exhibit specific parameters (e.g., optomechanical), the development of methods to control geometry and alignment, and the integration of LCEs into systems, such as e.g. actuators or optical components.

This dissertation describes light-driven systems based on thermoresponsive, dye-doped LCEs. It presents new methods for fabricating and aligning the elastomer-based components, as well as millimeter-scale devices that use LCEs as actuators heated through absorption of light. The work consists of a cycle of five articles dedicated to this topic.

The automated station for producing LCE films with predefined alignment (RoboLEC), described in the first publication, enables the fabrication of dye-doped materials suitable for use in light-driven systems. The user loads glass slides coated with a photoalignment layer, uploads images that define the alignment direction, and supplies a UV-curable adhesive with microspheres of a specified diameter to set the sample thickness. The process runs fully automatically, and the number of samples is limited by the supply of these materials.

The second article presents an overview of LCE results obtained within the research group in which I conducted my research. In addition to previously published outcomes, it introduces a previously unreported robot that mimics water-strider locomotion and moves on the water surface. Its leg motion is driven by an LCE “muscle”. Muscle deformation is induced by laser pulses at a frequency of a few hertz. Although the robot is about 6,000 times slower than its natural counterparts, to the best of my knowledge it is the first report of a water-strider robot at natural scale.

The third article describes a robot that imitates snail locomotion at natural scale. The device travels over surfaces coated with synthetic “mucus” (e.g., glycerine), enabling motion at various orientations relative to gravity, including vertical and upside-down. Locomotion is driven by a deformation wave propagating along the robot’s body, generated by laser scanning of the LCE.

The fourth and fifth articles present a rotary and linear motor, respectively. In both designs, motion is driven by scanning a laser beam over LCE elements, which allows the operating direction of each motor to be controlled. The rotary motor employs an LCE ring whose alignment forms a +1 topological defect, whereas the actuators in the linear motor feature a periodically varying, stripe-domain alignment along their length, produced using the novel rubbing-overwrite method described in the fifth publication.

Key words: liquid crystal elastomers, smart materials, photoresponsive polymers, actuators, soft robots, light-driven systems, motors

Streszczenie

Ciekłokrystaliczne elastomery (LCE, ang. liquid crystal elastomers) stanowią grupę inteligentnych materiałów, które pod wpływem zewnętrznych bodźców mogą w sposób odwracalny zmieniać kształt, kolor, współczynnik załamania światła oraz inne właściwości fizyczne. Cechy materiału są ściśle związane z uporządkowaniem jego cząsteczek. Kierunki badań nad LCE obejmują zarówno syntezę nowych związków umożliwiających otrzymywanie materiałów reagujących na odmienny zakres bodźców lub o określonych parametrach (np. opto-mechanicznych), jak i opracowywanie metod nadawania im zadanego kształtu i uporządkowania oraz integrację LCE jako siłowniki lub elementy optyczne w rozmaitych układach (np. robotach).

Niniejsza rozprawa dotyczy zasilanych i sterowanych światłem układów opartych na termoresponsywnych LCE domieszkowanych barwnikami. Praca prezentuje nowe metody wytwarzania i porządkowania materiałów oraz milimetrowych rozmiarów urządzenia, które wykorzystują LCE jako elementy wykonawcze ogrzewane poprzez absorpcję wiązki laserowej. Zaprezentowany cykl obejmuje pięć artykułów poświęconych tej tematyce.

Opisana w pierwszej publikacji stacja do automatycznego wytwarzania folii z LCE o zadanym uporządkowaniu (RoboLEC) umożliwia produkcję domieszkowanych barwnikami materiałów, co umożliwia ich wykorzystanie m.in. w sterowanych światłem układach. Użytkownik wprowadza szkiełka pokryte warstwą fotoporządkującą, wgrywa obrazy definiujące kierunek orientacji oraz podaje klej utwardzany promieniowaniem UV z mikrokulkami o określonej średnicy, determinującymi grubość próbki. Proces przebiega w pełni automatycznie, a liczba próbek jest ograniczona zapasem ww. materiałów.

Drugi artykuł przedstawia przegląd wyników w dziedzinie LCE uzyskanych w zespole badawczym, w którym prowadziłem prace. Oprócz wcześniej opublikowanych rezultatów zaprezentowano niepublikowaną wcześniej konstrukcję robota odwzorowującego ruch nartników, poruszającego się na powierzchni wody. Napęd kończyn zapewnia „mięsień” z LCE. Deformację mięśnia wywołują impulsy lasera o częstotliwości kilku Hz. Mimo że robot jest około 6000 razy wolniejszy od swoich naturalnych odpowiedników, zgodnie z moją wiedzą publikacja ta stanowi pierwsze doniesienie o robocie imitującym nartnika naturalnej wielkości.

Trzeci artykuł opisuje robota naśladującego ruch ślimaków, również w naturalnej skali. Urządzenie porusza się po powierzchniach pokrytych syntetycznym „śluzem” (np. gliceryną), co umożliwia ruch pod różnymi kątami względem pola grawitacyjnego, w tym pod górę i „do góry nogami”. Ruch wywołuje fala deformacji przesuwająca się wzdłuż ciała robota, wytwarzana poprzez skanowanie wiązki lasera.

Czwarty i piąty artykuł prezentują odpowiednio miniaturowy silnik obrotowy i liniowy. Obie konstrukcje wykorzystują wiązkę lasera skanującą elementy wykonane z LCE, co pozwala na zmianę kierunku pracy każdego z silników. Silnik obrotowy wykorzystuje pierścień z LCE o uporządkowaniu tworzącym defekt topologiczny o ładunku +1, natomiast siłowniki zamontowane w silniku liniowym mają uporządkowanie, w którym domeny okresowo zmieniają się wzdłuż materiału, uzyskane zaprezentowaną w piątej publikacji nowatorską metodą nadpisywania rubbingu.

Słowa kluczowe: ciekłokrystaliczne elastomery, inteligentne materiały, światłoczułe polimery, siłowniki, miękkie roboty, układy sterowane światłem, silniki

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Abbreviations

CLCE	cholesteric liquid crystal elastomer
DIW	direct ink writing
DLP	direct laser processing
DLW	direct laser writing
LC	liquid crystal
LCE	liquid crystal elastomer
LCN	liquid crystal network
POM	polarised optical microscopy
UV	ultraviolet

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Introduction

In 1971, Julian Antonisz created an animated short film titled *Jak działa jamniczek*, which can be translated as *How Does a Dachshund Work* (although the author did not provide an official English title). The film was made using unorthodox, non-camera techniques such as drawing directly onto the film reel. In an unusual and artistic manner, the movie presents animals, particularly dachshunds, as highly complex machines designed by nature. At one point, the narrator proposes: “Let’s try to imitate nature; let’s build a muscle engine” (originally in Polish: “Starajmy się raczej naśladować przyrodę, zbudujmy silnik mięśniowy”), while a mechanical construction is shown in which pistons and hydraulic cylinders are replaced by muscles.

Ten years later, in 1981, likely unaware of Antonisz’s film, Finkelmann, Kock, and Rehage published an article considered to be the first experimental report of a new class of materials: liquid crystal elastomers (LCEs) [1]. These belong to a broader category known as smart materials, which respond to specific environmental triggers by changing their physical properties, such as shape, color, or stiffness [2, 3]. Today, LCEs are widely regarded as ideal candidates for artificial muscles [4–7], and many examples of nature-inspired systems employing LCEs [8–11] have been developed, reflecting the spirit of the *How Does a Dachshund Work* concept. This arises from the unique combination of elastomer elasticity and the anisotropic order typical of liquid crystals (LCs) within LCEs. In response to external stimuli such as temperature [12] or light of a specific wavelength [13], these materials can undergo a reversible shape change driven by a phase transition between the anisotropic and isotropic states. The geometry of deformation (such as bending or shrinking) is governed by the molecular alignment of the material, which is determined during the fabrication process. Upon activation, the material contracts along the direction of alignment and expands perpendicularly, so uniformly aligned LCEs behave similarly to a muscle (see Fig. 1).

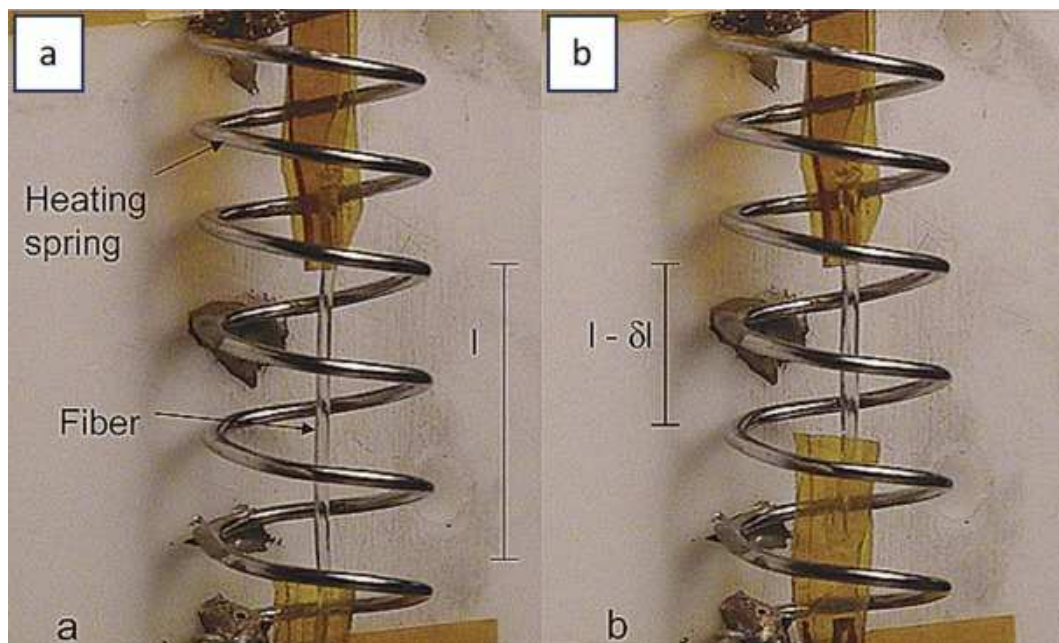


Fig. 1 Thermal actuation of a uniaxially aligned liquid crystal elastomer fiber loaded with a 200 mg weight. (a) At room temperature (nematic phase), the fiber is fully extended (12.6 mm length, 0.3 mm diameter). (b) Heating above the nematic–isotropic transition induces 40% contraction, lifting the loaded weight. The deformation is fully reversible upon cooling. Image adapted from [12].

Since their introduction in the early 1980s, LCEs have attracted steadily growing scientific interest. Initially investigated within materials science and polymer chemistry, research has expanded into other fields such as mechanical engineering, optics, soft robotics, and applied physics (see Fig. 2). This interdisciplinary expansion has been supported by two key developments: cost-effective, scalable

fabrication methods and advanced alignment strategies enabling precise control over molecular orientation and complex shape programming. Today, LCE research spans a wide range of applications, including devices with tunable optical properties [14–17], self-oscillating systems [11, 18, 19], and soft robots [20–22]. The increasing importance of these materials is reflected in the growing number of scientific publications and their citations over the past two decades (see Fig. 2). While initial efforts focused on phase transitions and mechanical characterization, recent work has shifted toward functional integration, patterned alignment, and enhanced responsiveness to diverse stimuli including thermal, photonic, magnetic, and chemical triggers.

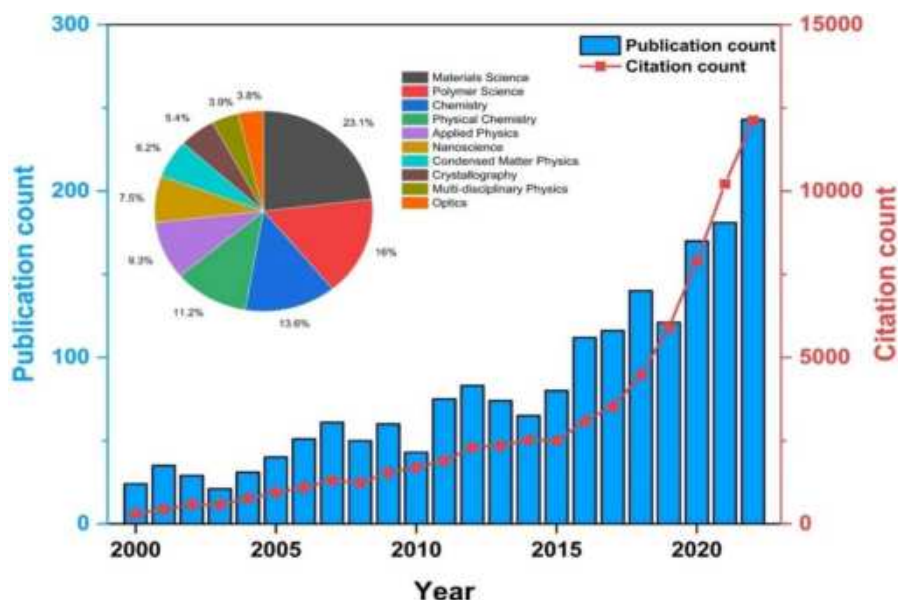


Fig. 2 Field distribution and annual output of LCE research from 2000 to 2022. Nearly 2 000 papers on LCEs have been published, accumulating around 70 000 citations by 2022. Approximately half were published between 2018 and 2022, reflecting a rapid rise in research activity. Image adapted from [23].

This dissertation contributes to the evolving research landscape on LCEs by focusing on thermally responsive LCEs activated wirelessly through light absorption. It addresses two primary objectives:

1. development of fabrication methods enabling programmable shape change and precise molecular alignment across multiple length scales in LCE elements, and
2. integration of LCE elements into soft, untethered systems such as light-driven actuators, motors, and robots.

Together, these objectives reflect a progression from fundamental material design to the implementation of functional devices.

Following this introduction, the chapter *Fundamental properties of LCEs* provides essential background on the key physical and chemical characteristics of liquid crystal elastomers. The chapter *Theoretical foundations* briefly explains the principles underlying LCE deformation. The next chapters present the article-cycle results and comparisons with related work. Publications that are part of the cycle are cited as **Publication I–V**, and the label **Publication** is capitalized and bolded at every reference. *Methods of LCEs fabrication*, reviews various fabrication techniques and highlights the contribution of **Publication I** to advancements in this area. The chapter *Light-driven systems* presents different applications of LCEs as light-responsive actuators, focusing in particular on motors and bio-inspired soft robots. It also outlines the contributions of **Publications II–V** to achieving precise molecular alignment and integrating LCEs into untethered, light-driven systems. Following that, each publication in the cycle is included in its original published form. The *Conclusions* chapter synthesizes the main findings, discusses their broader implications, and outlines directions for future research. Finally, the sections *Author’s contribution* and *Co-authors statements* outline my role in each publication and include confirmations from co-authors who led the projects or contributed most significantly.

Fundamental properties of LCEs

LCEs consist of long molecules known as polymer chains and mesogens (see Fig. 3), typically rigid aromatic or cycloalkyl cores connected to the polymer backbone via flexible spacers, usually alkyl or alkoxy chains [24]. The mesogens, or mesogenic units, are the components responsible for the material's liquid crystalline behaviour. While the mesogenic cores promote orientational order, the flexible spacers provide the necessary chain mobility. Together, these features enable the material to self-organize into mesophases such as the nematic or smectic state [3]. In most LCEs, the mesogens are rod-like, as illustrated in Fig. 3, but other geometries have also been reported, for example discotic mesogens [25]. In this work, however, I focus specifically on rod-like mesogens, since this was the configuration present in the materials I studied.

Mesogens can be incorporated into the polymer in three main architectures (see Fig. 3):

1. Side-chain, end-on: mesogens attach to the backbone via flexible spacers at the end of the mesogenic unit.
2. Side-chain, side-on: flexible spacers are linked to the middle of the mesogen, resulting in perpendicular orientation relative to the backbone.
3. Main-chain: mesogens are integrated directly into the polymer backbone. This configuration offers a strong link between molecular order and mechanical deformation.

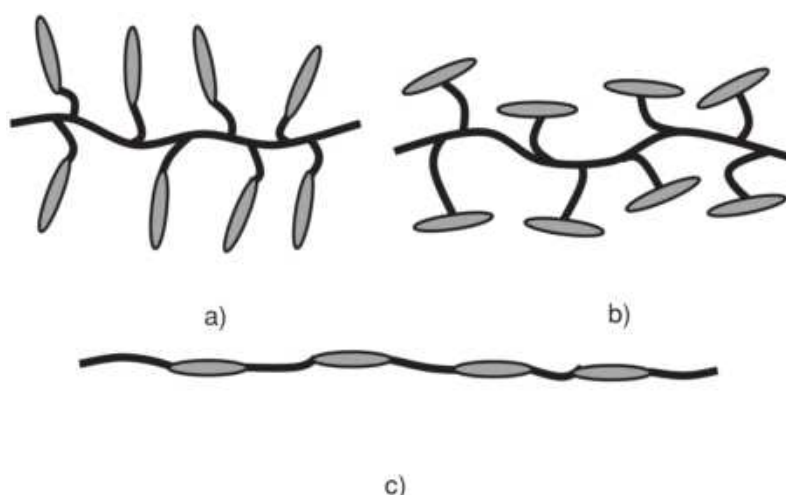


Fig. 3 Three mesogen-backbone connection architectures: (a) side-chain, end-on; (b) side-chain, side-on; and (c) main-chain. Mesogens shown as grey ellipsoids. Image adapted from [26].

In my research, a side-chain, end-on LCE system was employed. The LCE was prepared from a monoacrylate mesogenic monomer, C6BP (4-methoxybenzoic acid 4-(6-acryloyloxy-hexyloxy) phenyl ester; Synthon Chemicals), and a diacrylate mesogenic crosslinker, RM257 (1,4-Bis-[4-(3-acryloyloxypropyloxy)benzoyloxy]-2-methylbenzene; Synthon Chemicals), using Irgacure 369 (2-benzyl-2-(dimethylamino)-4'-morpholinobutyrophenone; Sigma-Aldrich) as the photoinitiator. The C6BP–RM257 mixture was selected primarily for its prior validation by my supervisor at the European Laboratory for Non-Linear Spectroscopy (LENS) in Sesto Fiorentino, Italy, ensuring a reliable and reproducible formulation. In addition to being relatively inexpensive, the mixture is compatible with surface alignment techniques, which was the only alignment method used in my research. Although this thesis does not include original direct laser writing (DLW) experiments, the formulation is also compatible with that technique [14]. Since our laboratory is equipped with a DLW system manufactured by Nanoscribe, using a common precursor mixture across projects enhances both experimental flexibility and resource efficiency. Both surface alignment and DLW are briefly introduced in the *Methods of LCEs fabrication*.

In LCEs, mesogens can be organized into various mesophases, most commonly nematic, smectic A, smectic C, or cholesteric (see Fig. 4), each contributing to specific material properties. In the nematic phase (as in a C6BP–RM257 mixture), mesogens exhibit long-range orientational order without positional order: their long axes align on average along a common direction (the director), while their molecular centers of mass remain randomly distributed. When positional order exists, the mesophase is classified as smectic, with layers perpendicular to the director in smectic A or tilted relative to the layer normal in smectic C.

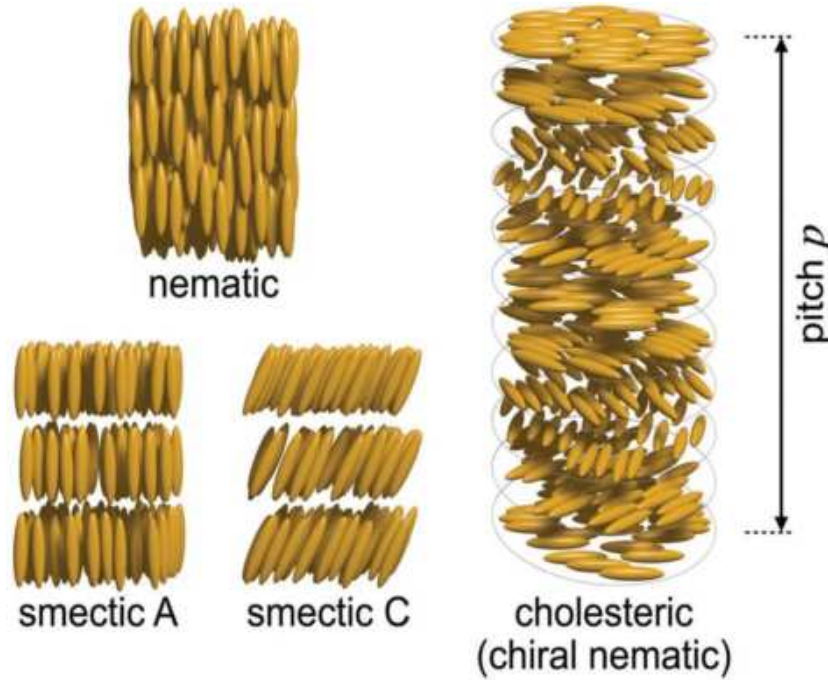


Fig. 4 Most commonly studied LCE mesophases: nematic, smectic A, smectic C, and cholesteric. Adapted from [27].

In cholesteric (chiral nematic) LCEs, the director rotates helically along an axis, forming a periodic structure in which one full 360° rotation defines the pitch, the distance along the helical axis over which the director completes a full turn (see Fig. 4). This periodic structure acts as a Bragg reflector, selectively reflecting light at wavelengths determined by the pitch [17]. Such materials exhibit tunable mechanochromic responses: stretching alters the pitch, shifting the reflected wavelength and thus producing visible color changes under strain (see Fig. 5).

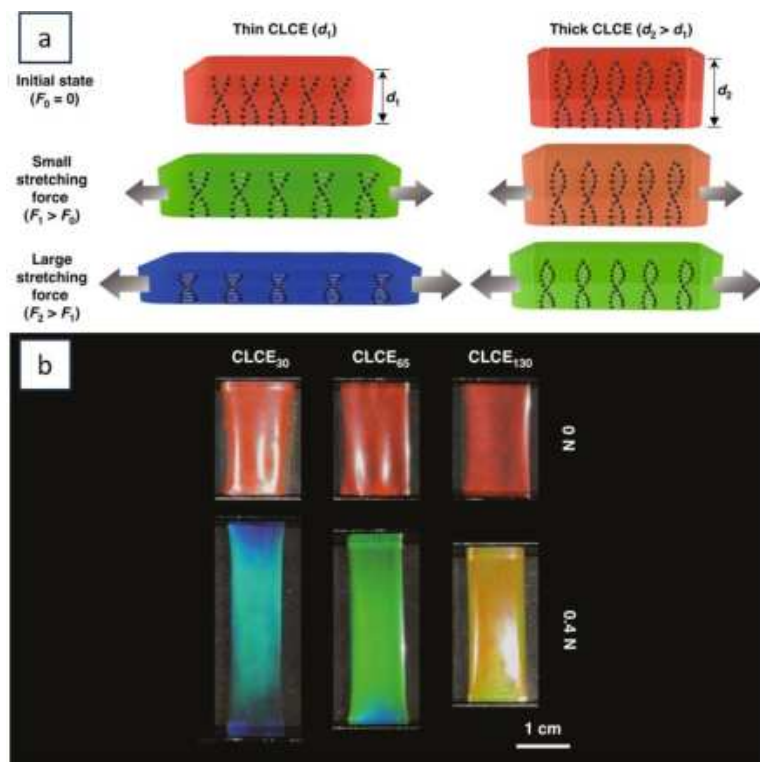


Fig. 5 Color change in cholesteric LCEs (CLCEs). (a) Pitch modulation under uniaxial stretching depends on initial sample thickness. (b) Experimental demonstration of color change in cholesteric LCEs under a tensile force of 0.4 N. The numbers in the headings correspond to the sample thicknesses: 30, 65, and 130 μm , respectively. Adapted from [17].

To convert these polymers into solid materials with stable internal structure, they must be crosslinked. Crosslinking introduces bonds between polymer chains, forming a network that preserves the anisotropic organisation of mesogens. The degree of crosslinking plays a critical role in determining the mechanical properties of the material [24].

The term liquid crystal elastomer refers to loosely crosslinked networks that exhibit soft, rubber-like elasticity and large, reversible deformations [3]. In contrast, liquid crystal networks (LCNs) are more densely crosslinked, resulting in stiffer, often glassy or brittle materials. In such systems, polymer chain mobility is strongly restricted, and deformation under stimuli is significantly limited [28, 29]. However, likely because LCEs were developed earlier, the term “LCE” is often used more broadly to refer to a wider range of materials within this spectrum [24]. Notably, the International Liquid Crystal Elastomer Conference (ILCEC), held every two years and focused on this research area, includes studies of both LCEs and LCNs, despite the specific use of “elastomer” in its name. In this thesis, especially since crosslinking was not the primary focus of my research, I adopt this convention and use “LCE” as a general term referring to crosslinked liquid crystal polymer networks that exhibit reversible deformation in response to external stimuli, regardless of their crosslinking density. For clarity, each article included herewith specifies the composition of the precursors used.

LCEs exhibit reversible deformation driven by a phase transition from an anisotropic (ordered) to an isotropic (disordered) state. In the isotropic phase, polymer chains adopt a randomly coiled, spherical configuration, whereas in the anisotropic phase, they form ellipsoidal shapes, either prolate or oblate, depending on the material [30, 31]. In this work, I focus exclusively on LCEs with a prolate configuration, as this was the structural form present in the materials investigated during my research. In such systems, the transition to the isotropic phase induces contraction along the director and expansion perpendicular to it. Upon removal of the stimulus (e.g., cooling) or application of another stimulus (e.g., light of a different wavelength), the original alignment and dimensions are restored (see Fig. 6a). The magnitude and geometry of actuation depend directly on both the degree and direction of molecular alignment (see Fig. 6b). Well-aligned samples exhibit large, anisotropic mechanical responses,

therefore, measuring reversible strain serves as a practical metric for assessing alignment quality [32]. For this reason, uniformly aligned nematic and smectic LCEs are widely used as actuators [26].

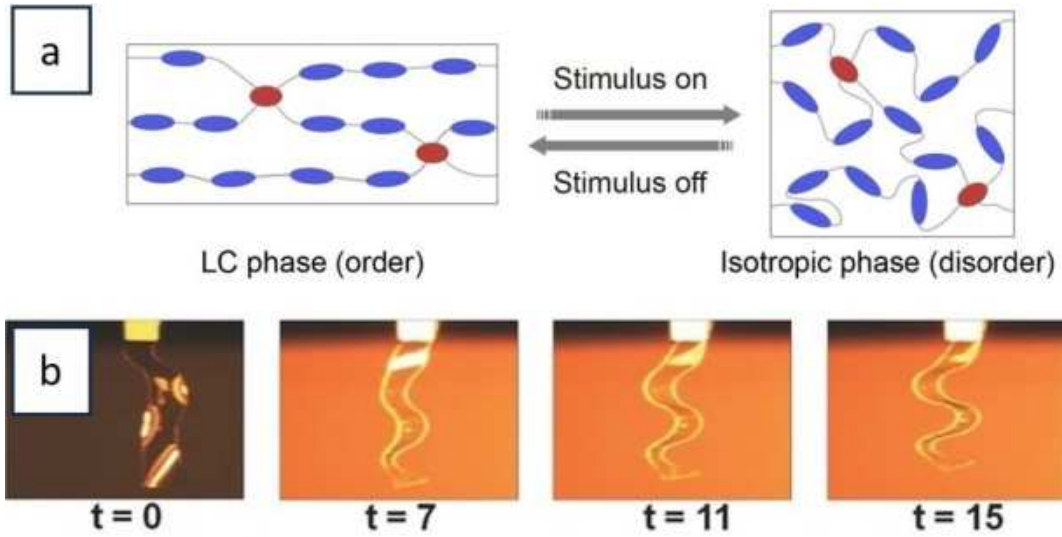


Fig. 6 (a) Diagram illustrating phase-transition behavior of LCEs. In response to an external stimulus, LCEs reversibly contract along the polymer chains and expand in perpendicular directions. (b) Accordion-like deformation of an LCE film with patterned molecular alignment captured after t seconds of infrared irradiation. The film measures 30 mm by 5 mm and is 15 μm thick. Adapted from [33] (a) and [34] (b).

Molecular alignment is quantified using the orientational order parameter, S , defined as:

$$S = \frac{1}{2} (3 \cos^2 \theta - 1) \quad (1)$$

where θ is the average angle between a mesogen's long axis and the director. An order parameter $S = 0$ corresponds to a disordered (isotropic) system, while $S = 0.5$ (with an average angle $\theta \approx 35^\circ$) is considered moderate alignment [35], and $S = 1$ represents perfectly aligned mesogens. In well-aligned nematic LCEs, order parameter values can reach or even exceed 0.8 [36]. This parameter can be determined using techniques such as polarized infrared or UV-vis spectroscopy or X-ray scattering. As a key metric of molecular orientation, the order parameter is crucial for assessing alignment quality and, by extension, the efficiency and reliability of actuation in LCE-based devices.

One of the most accessible ways to measure molecular alignment, both qualitatively and quantitatively, is through birefringence, defined as the difference between the refractive indices parallel and perpendicular to the director. Aligned LCEs exhibit strong optical anisotropy, making birefringence a powerful tool for assessing molecular orientation. This optical property correlates linearly with the liquid crystal order parameter [35], and it is routinely observed using polarized optical microscopy (POM) [28, 37, 38]. For example, in the nematic phase, under POM with crossed polarizers, regions aligned at about 45° to the polarizer axes appear bright due to polarization rotation, while regions aligned parallel to one of the polarizers appear darker (see Fig. 7). These characteristic patterns, known as “textures”, reveal alignment of the material. External stimuli, such as heat, causes changes in texture brightness or color, indicating changes in the order parameter and thus in birefringence [35, 39]. Due to its simplicity and non-destructive nature, POM was the primary technique for characterizing alignment in LCE samples fabricated by various methods throughout the experimental work presented in this dissertation.

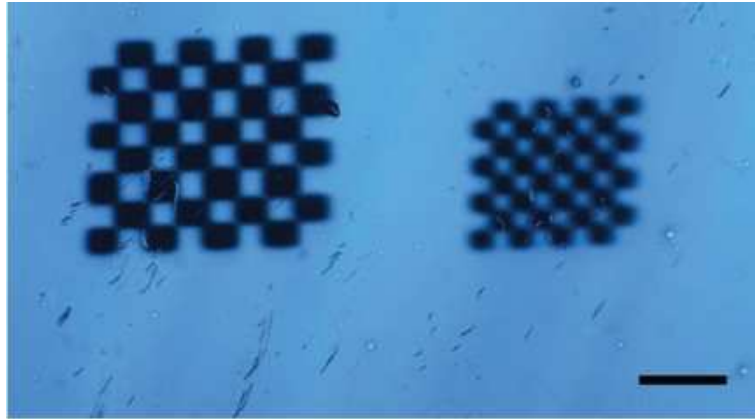


Fig. 7 POM image of checkerboard patterns in LCE film in the nematic phase, showing areas aligned 45° to the polarizers (bright) and regions aligned parallel to one of the polarizers (dark). Scale bar: 500 μm . Adapted from [37].

Theoretical foundations

Pierre-Gilles de Gennes was awarded the Nobel Prize in Physics in 1991 “for discovering that methods developed for studying order phenomena in simple systems can be generalized to more complex forms of matter, in particular to liquid crystals and polymers” [40]. During the 1970s, he provided a theoretical framework to describe how order–disorder transitions arise across a broad class of phase transitions, with particular focus on liquid crystals and polymers in solution. Although not the central focus of the Nobel Prize, de Gennes also theoretically predicted the existence of LCEs in 1975 [41]. In a brief note, he proposed a material that would couple the elastic properties of a solid with the orientational order of a nematic liquid crystal.

Following de Gennes’s work, two quantitative models describing how the mechanical properties of LCEs depend on molecular order were independently developed by two groups in the late 1980s [42, 43]. These approaches extended the classical concept of rubber elasticity by assuming that the polymer network consists of chains with anisotropic statistical properties, arising from the orientational order characteristic of the nematic phase. This work led, in 1994, to the introduction of the concept of *soft elasticity* [44], along with the *Trace formula* for the free energy density of nematic LCEs [45]. The theoretical framework briefly outlined in the following is described in detail in the monograph by Warner and Terentjev [35], which served as the primary source for my understanding of LCE behavior.

In the *Trace formula*, the statistical properties of an aligned polymer network are described by the step length, which represents the effective segment length over which a polymer chain can bend. This is a statistical measure of chain flexibility and typically spans many monomer units. In isotropic rubber, the step length is uniform in all directions. However, in uniaxial nematic LCEs, the alignment of mesogens introduces anisotropy, which requires a tensorial description. The step length tensor $\underline{\ell}$ is generally defined as [35]:

$$\ell_{ij} = l_{\perp} \delta_{ij} + (l_{\parallel} - l_{\perp}) n_i n_j, \quad (2)$$

where: $l_{\parallel}$ and $l_{\perp}$ are step lengths along and perpendicular to the director, respectively; δ_{ij} is the Kronecker delta; and n_i is the i -th component of the unit director vector $\mathbf{n}$.

With the step length tensor capturing the anisotropic chain statistics associated with mesogen alignment, the elastic behavior of nematic LCEs can be described using the *Trace formula*. This expression for the free energy density generalizes classical rubber elasticity to account for anisotropic deformations and serves as the starting point for modeling the mechanics of LCEs [45]:

$$F = \frac{1}{2} n_s k_B T \cdot \text{Tr}(\ell_0 \cdot \underline{\lambda}^T \cdot \ell^{-1} \cdot \underline{\lambda}), \quad (3)$$

where: n_s is the number of network strands per unit volume, k_B is Boltzmann constant, T is the absolute temperature, ℓ_0 and ℓ are step length tensors in the reference (initial) and current (deformed) state, respectively. $\underline{\lambda}$ is a deformation tensor capturing the magnitude and direction of deformation. Its diagonal components represent the ratios of the current length to the reference length along the principal axes. For example, in a uniaxially aligned nematic LCE with the director along the z -axis, heated up to the isotropic state, in the absence of external stress, the deformation must preserve volume and maintain the symmetry of alignment, so the largest deformation occurs along the z -axis. If the deformation along z is denoted by λ , constant volume implies $\lambda_x \lambda_y \lambda_z = 1$. By symmetry $\lambda_x = \lambda_y = 1/\sqrt{\lambda}$. Therefore, the deformation tensor can be expressed as:

$$\underline{\lambda} = \begin{pmatrix} \frac{1}{\sqrt{\lambda}} & 0 & 0 \\ 0 & \frac{1}{\sqrt{\lambda}} & 0 \\ 0 & 0 & \lambda \end{pmatrix} = \underline{\lambda}^T. \quad (4)$$

For the case where the director is aligned along the z-axis, the step length tensor ℓ_0 takes the form:

$$\ell_0 = \begin{pmatrix} l_{\perp} & 0 & 0 \\ 0 & l_{\perp} & 0 \\ 0 & 0 & l_{\parallel} \end{pmatrix}. \quad (5)$$

Because in the deformed state the material becomes isotropic, the step length tensor takes the form $\ell = a \cdot I$, where a is the (isotropic) step length value and I is the identity tensor. The *Trace formula* for the free energy density simplifies to:

$$F = \frac{1}{2} n_s k_B T \cdot \left(\lambda^2 \frac{l_{\parallel}}{a} + \frac{2}{\lambda} \frac{l_{\perp}}{a} \right). \quad (6)$$

Thus the material adjusts its volume to minimize the free energy. By solving the condition $\partial F / \partial \lambda = 0$, one obtains the spontaneous strain along the z-axis [41, 42]:

$$\lambda = \left(\frac{l_{\perp}}{l_{\parallel}} \right)^{\frac{1}{3}}. \quad (7)$$

For polymer chains organized into a prolate shape, the step length perpendicular to the director $l_{\perp}$ is smaller than the parallel component $l_{\parallel}$ [35], resulting in a stretch ratio $\lambda < 1$, which indicates material contraction along the director, as expected. Based on this observation, the ratio $l_{\perp}/l_{\parallel}$ can be estimated from experimental measurements of thermally induced deformation.

Methods of LCEs fabrication

Because of the crucial role of molecular order in LCEs, molecular alignment methods are very important and are constantly being developed. Without any alignment applied, mesogens would spontaneously self-organize only into micrometer-sized polydomain regions (see Fig. 8a), resulting in a material that responds isotropically upon stimulation [46]. Fabricating aligned LCEs generally involves two key steps: first, the mesogens must be oriented in the desired direction, second, that alignment must be locked in place via cross-linking, which leads to formation of a three-dimensional polymer network. One of the major challenges is achieving precise control over mesogen alignment, particularly when spatial variation is required within the bulk of the material [47].

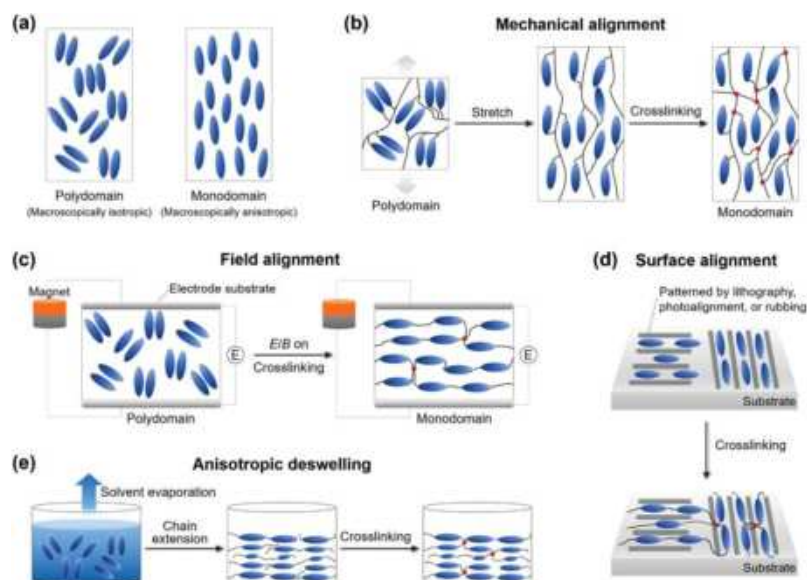


Fig. 8 (a) Comparison between polydomain LCE. (b–e) Schemes of different alignment methods for LCEs: (b) mechanical alignment, (c) electric or magnetic field alignment, (d) surface alignment, (e) anisotropic deswelling used for cholesteric LCEs. Adapted from [48].

Mechanical alignment

The first monodomain LCE was obtained through a mechanical alignment method using a two-step polymerization process [49]. Initially, a partially cross-linked prepolymer is produced (step one), forming a soft, gel-like material. In the second step, this prepolymer undergoes uniaxial mechanical stretching, aligning the mesogens, which is then immediately followed by a second cross-linking reaction, e.g. by UV irradiation, to “freeze” this orientation in place (see Fig. 8b). While this approach is highly effective for producing uniform uniaxial alignment over bulk samples, it falls short when more complex or spatially varying alignment geometries are needed. Nonetheless, researchers are extending these boundaries by stretching the prepolymer within specially designed molds to preprogram the desired deformation profile before the final cross-linking stage [50].

Anisotropic Deswelling

Two-step polymerisation is also used in the fabrication of cholesteric LCEs. The method, known as anisotropic deswelling [51], relies on solvent evaporation that is restricted to the vertical direction. In the first step, a weakly crosslinked gel is formed in a homogeneous solution, resulting in an initially isotropic material. The gel undergoes deswelling exclusively along the vertical axis, while its lateral dimensions in the horizontal plane remain unchanged. This directional contraction flattens the overall shape of the

polymer network and promotes macroscopic planar alignment of the cholesteric structure. As the cholesteric phase emerges from the isotropic state, the alignment is fixed during the second step of polymerisation, which completes the crosslinking reaction (see Fig. 8e). This technique enables fabrication of uniform cholesteric LCE films with well-defined planar helicoidal alignment, where the helix axis is oriented perpendicular to the film surface. Although highly effective in producing high-quality alignment, the method is time-consuming due to slow solvent evaporation and poses challenges for large-scale fabrication.

Field alignment

Aligning liquid crystals via electric field is a foundational technique in liquid crystal displays (LCD) technology. Displays utilize arrays of tiny electrodes and color filters, typically three subpixels (red, green, and blue), each with its own electrode pair, to generate electric fields that reorient LC molecules, enabling rapid switching at refresh-rate frequencies [52]. Field alignment is also adapted for LCE fabrication: liquid LCE precursors are sandwiched between two glass slides coated with transparent electrodes [53] or positioned within a magnetic-field generator [54]. Once the field aligns the mesogens, cross-linking fixes their orientation (see Fig. 8c). However, maintaining this alignment requires very strong fields, which restricts the method to thin films. For example, inducing reliable alignment in a 50 μm -thick LCE film has been shown to require a magnetic field of 1.1 T over 12 hours [55]. Similarly, electric-field-based approaches often demand high voltages, such as $6.8 \text{ V } \mu\text{m}^{-1}$ for a 40 μm film [53]. The exact values of the required field strength depend on various material parameters, such as magnetic susceptibility [47] or molecular dipole moment [56]. Consequently, field-aligned LCEs are typically limited to micrometer-scale thicknesses, making them unsuitable for 3D, volume elements.

Surface alignment

Surface alignment is another technique widely used in both LCDs and LCEs. In LCDs, when no electric field is applied, liquid crystal molecules are structured by surface anchoring on alignment layers. This enables switching between surface-induced and field-induced orientations, which is essential for modulating pixel brightness and color [52]. In LCE fabrication, surface alignment is typically achieved by sandwiching a liquid crystal precursor between two glass slides coated with alignment layers. The mesogens align according to the orientation defined by these layers and retain this configuration after cross-linking (see Fig. 8d), which is typically performed through photopolymerization or reactive precursors [48]. One common method for preparing alignment layers involves rubbing a polymer coating, usually polyimide or polyvinyl alcohol, with a cloth, to align the polymer chains through localized melting [57]. Rubbing provides strong anchoring and reliable uniaxial alignment, but it is limited to simple geometries [58] and may introduce surface defects or static charges [59, 60].

To achieve complex director fields different surface alignment techniques, like photoalignment [37], are employed. In this method, a thin photosensitive layer is deposited on the substrate and irradiated with linearly polarized light. The light induces anisotropy in the layer, causing the mesogens to align either parallel [61] or perpendicular [62] to the polarization direction, depending on the type of photosensitive material used. This method avoids mechanical contact and, combined with spatially modulated light polarisation, enables high-resolution alignment patterns (see Fig. 9).



Fig. 9 POM image of a liquid crystal elastomer film fabricated using photoalignment. The alignment pattern was generated by projecting images with predefined linear polarization onto a photosensitive layer using a laser projector. White arrows indicate the polarizer and analyzer axes, and yellow arrows the local mesogen orientation. Scale bar: 3 mm. Adapted from [37].

Lithographic patterning represents another surface-alignment technique capable of producing spatially controlled director configurations in LCEs. In this method, the substrate is microstructured, typically via photolithography, to generate grooves or channels that affect the orientation of mesogens. The resulting director field conforms to the orientation imposed by the topographic features of the substrate [63]. When the surface pattern incorporates rotational symmetry or discontinuities, the induced alignment may give rise to singularities - locations where the director cannot be defined continuously. Such singularities are known as topological defects and are characterised by their topological charge, which quantifies the net rotation of the director around a closed loop that encircles the defect core. A topological charge of $+1$, for example, corresponds to a complete 360° counterclockwise rotation [64]. Fig. 10 presents an example of a surface-aligned LCE containing an array of $+1$ and -1 defects, produced using a lithographically patterned substrate.

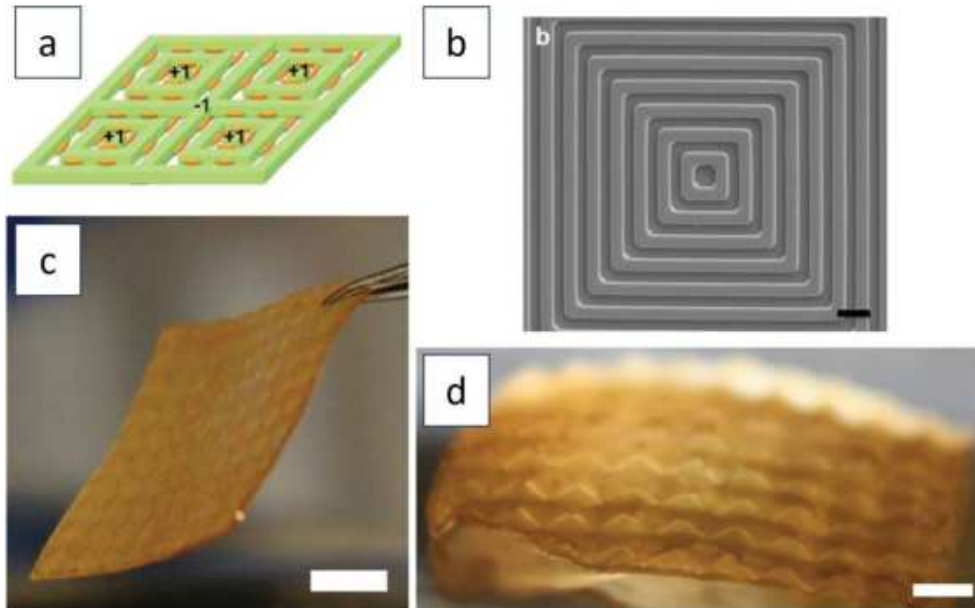


Fig. 10 LCE film with “ $+1$ ” and “ -1 ” topological defect array. (a) Schematics of mesogen alignment with defects labeled. (b) Scanning electron microscopy image of the pattern fabricated by lithographic methods in epoxy resin (channel depth: $1.5\ \mu\text{m}$). (c) LCE film at room temperature and (d) at $200\ ^\circ\text{C}$, aligned using an array of patterns shown in (b). Scale bars: (b) $5\ \mu\text{m}$, (c) 3 mm, (d) 1 mm. Adapted from [65]

Overall, rubbing, photoalignment, and lithographic patterning provide complementary methods for aligning mesogens in LCEs, each suited to different alignment requirements and material geometries. However, the influence of surface alignment typically extends only to depths of around 100 μm [66], which limits its effectiveness in thicker LCE samples. Controlling alignment throughout the bulk remains challenging, and in practice, the only well-established way to vary the director orientation across the sample thickness is through twisted nematic configurations, in which the liquid crystal molecules are aligned at different angles on the top and bottom surfaces, causing the director to rotate gradually through the film. Therefore, structures with different geometries or with spatially varying alignment throughout the bulk require alternative fabrication approaches.

Direct Laser Writing

DLW is a high-resolution fabrication technique based on two-photon photopolymerisation. This nonlinear process requires a pulsed laser tightly focused into the volume of a photocurable LCE precursor, where two-photon absorption initiates polymerisation within voxels (volumetric pixels), typically smaller than 1 μm across [48, 67]. Complex 3D microstructures are built by scanning the beam with galvanometric mirrors or moving the sample on a piezo stage (see Fig. 11a). Alignment is typically induced via surface treatment, though alternative methods such as electric field alignment [16, 68, 69] have also been demonstrated, allowing mesogen orientation control during printing. After printing, unpolymerised precursor is removed using an appropriate solvent, leaving behind the solid, patterned structure. Although DLW requires expensive equipment and precise optimisation of printing parameters such as laser power and writing speed, it enables fabrication of complex geometries and offers exceptional versatility for designing microscale, shape-changing LCE devices (see Fig. 11b).

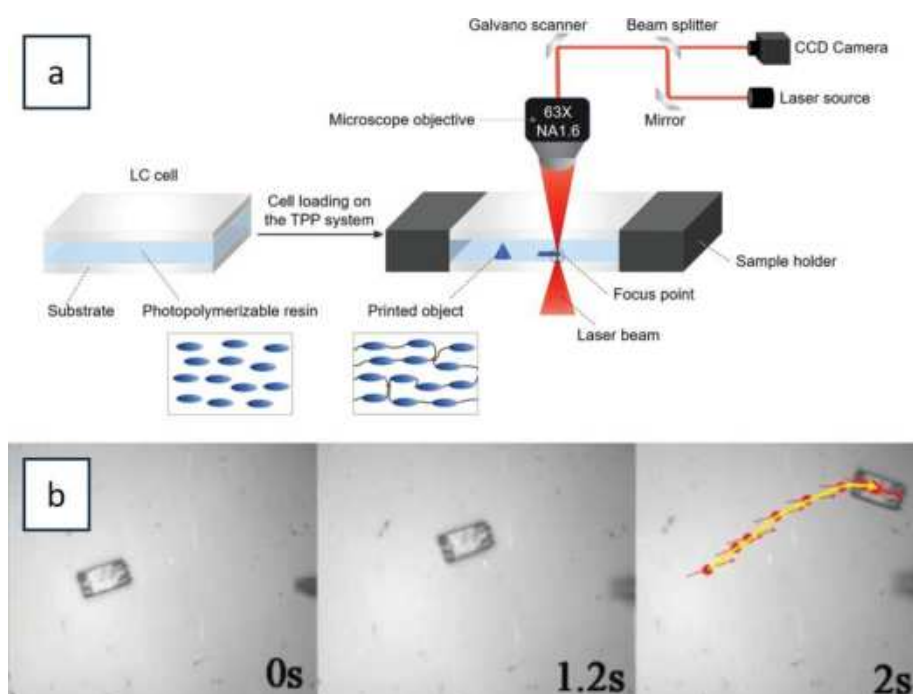


Fig. 11 (a) Schematic of the DLW. A sandwich-like cell filled with photocurable resin is mounted in a sample holder. The pulsed laser beam is focused into the resin by a microscope objective. Since two-photon polymerisation (TPP) occurs only at the focal volume, controlled movement of the focus using a galvo scanner enables printing of predefined 3D shapes. A CCD camera provides live visualisation of the process. (b) Motion of a 60 μm -long microrobot fabricated via DLW. The microrobot was actuated using a chopped laser beam (532 nm, 50 Hz, 10 W mm^{-2}). The final image shows the robot's trajectory during motion. Adapted from [48] (a) and [20] (b).

Digital Light Processing

Digital Light Processing (DLP) is a layer-by-layer additive manufacturing technique that enables rapid fabrication of complex LCE structures. It relies on spatially controlled photopolymerization: a digital micromirror device projects patterned UV light into a vat of photocurable LCE precursor, selectively curing one layer at a time. After each exposure, the platform moves vertically to allow the next layer to be projected and cured (see Fig. 12a). Compared to Direct Laser Writing, DLP offers significantly faster printing speeds and is suited for creating millimeter-scale structures with high efficiency. While conventional DLP setups typically produce isotropic crosslinking, several strategies, such as applying external magnetic field [70] or inducing shear flow [4], have been developed to induce anisotropic mesogen alignment during printing. These approaches enable fabrication of LCE structures with designed actuation behavior, however, only relatively simple alignment geometries have been demonstrated to date (see Fig. 12b). Moreover, the orientation parameter of the obtained LCEs is relatively low (e.g. 0.33 for magnetic field alignment reported in [70]).

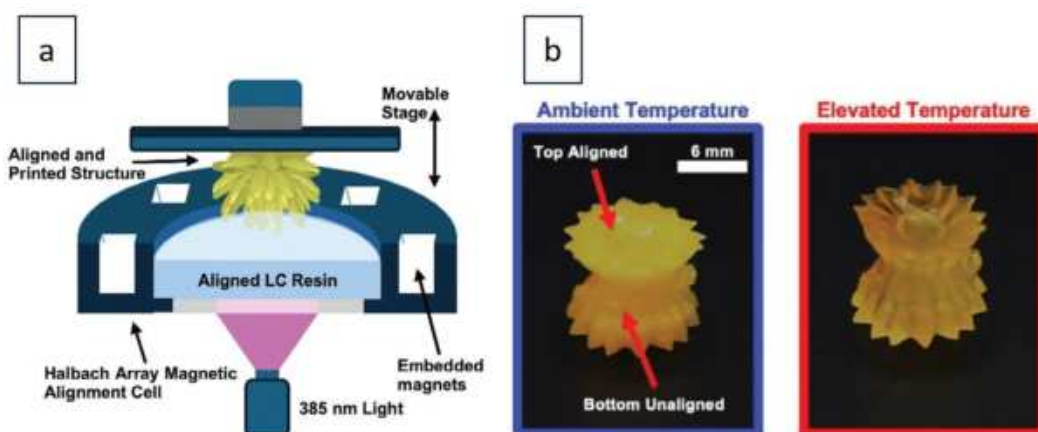


Fig. 12 (a) Schematic of DLP printing with magnetic-field-induced alignment of LCE. Each layer is polymerized by UV light projected from below the vat filled with liquid crystal precursor. Successive layers are built by vertically translating the stage and updating the projected pattern. The cubic magnets arranged in a Halbach array generate a uniform magnetic field of 100 mT. (b) Example of an LCE structure printed via DLP. The blue frame shows the structure at room temperature, and the red frame shows it after heating (exact temperatures not provided by the authors). The structure is mirrored: the top “flower” was printed with horizontal alignment, while the bottom was printed without alignment. Upon heating, only the aligned top flower exhibits significant deformation. Adapted from [70].

Direct Ink Writing

Spatial control of molecular alignment in LCEs can also be achieved using an extrusion-based 3D printing technique known as direct ink writing. During extrusion at temperatures within the nematic phase, mesogens align along the printing direction under shear forces [47]. This alignment is subsequently fixed by ultraviolet irradiation, which initiates cross-linking and solidifies the structure (see Fig. 13).

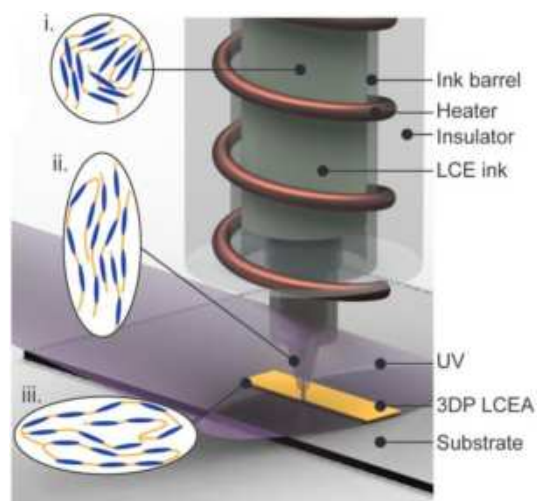


Fig. 13 Schematic illustration of direct ink writing of an LCE. (i) The disordered precursor is stored in a heated ink container. (ii) Shear-induced alignment occurs as the material is extruded through the nozzle. (iii) The extruded filament is cured on the substrate using ultraviolet UV light, forming a crosslinked and molecularly aligned structure of a 3D-printed LCE actuator. Adapted from [71].

The resulting shear-induced orientation allows precise control over molecular alignment within each printed layer, with spatial resolution primarily constrained by the capabilities of the printing system and the rheological properties of the ink. The printed LCE structures exhibit directionally dependent actuation behaviour governed by the programmed mesogen orientation, enabling complex and programmable shape transformations (see Fig. 14). Effective implementation of this method requires careful optimisation of LCE precursor properties, such as viscosity, which often necessitates temperature control of the nozzle, ink container, and printing substrate [47].

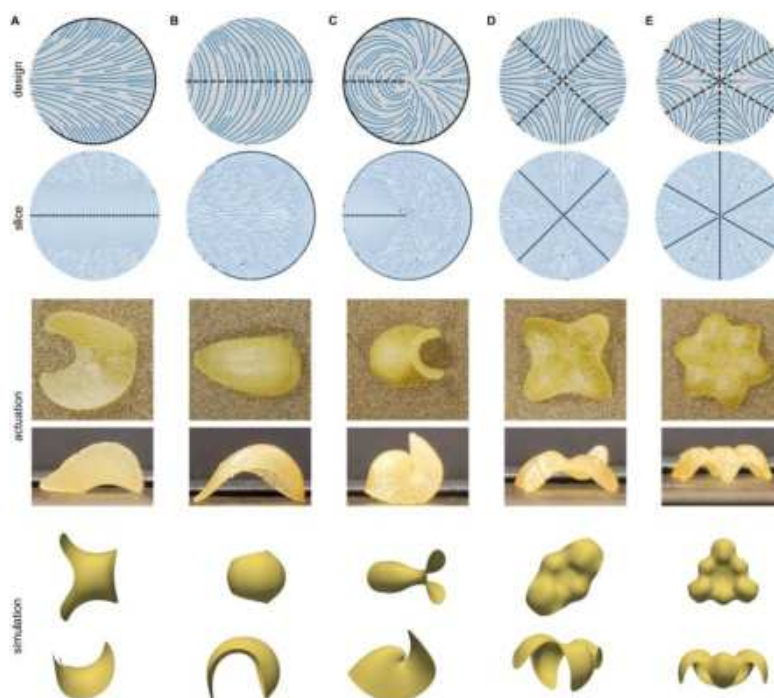


Fig. 14 LCEs with complex director fields fabricated using the DIW method. The “design” row shows the intended director orientation, “slice” shows the programmed print paths, “actuation” presents photographs of the LCEs under thermal actuation (140–150°C), and “simulation” displays computational predictions of the resulting deformations. The initial radius of each structure was 10 mm, and the printed line thickness was approximately 480 μm . Adapted from [72].

Automated LCE Film Fabrication

Thanks to techniques based on commercially available technologies, such as DLW, DLP, and DIW, the fabrication of 3D LCE structures has been largely automated. Companies such as Nanoscribe and Femtika offer ready-to-use DLW systems designed for scientific applications. In contrast, there is still a notable lack of automated methods for producing LCE films with controlled molecular alignment which would be a valuable tool in research where many samples with different characteristics need to be fabricated. This is particularly relevant given that, in many cases where three-dimensional structuring is not required, the simplest fabrication method involves polymerizing the material within an alignment cell and then cutting it to the desired shape.

The system described in **Publication I** (RoboLEC-Robot for LCE fabrication) addresses this gap by introducing a custom-built workstation (see Fig. 15a) capable of fabricating LCE films with pre-programmed molecular alignment. The device was constructed by modifying a commercially available 3D printer to produce sandwich-like alignment cells. The user supplies glass slides coated with a photoalignment layer, an LCE precursor mixture, and glue doped with microspheres that define the film thickness. A set of user-designed images is also uploaded to determine the spatial alignment pattern.

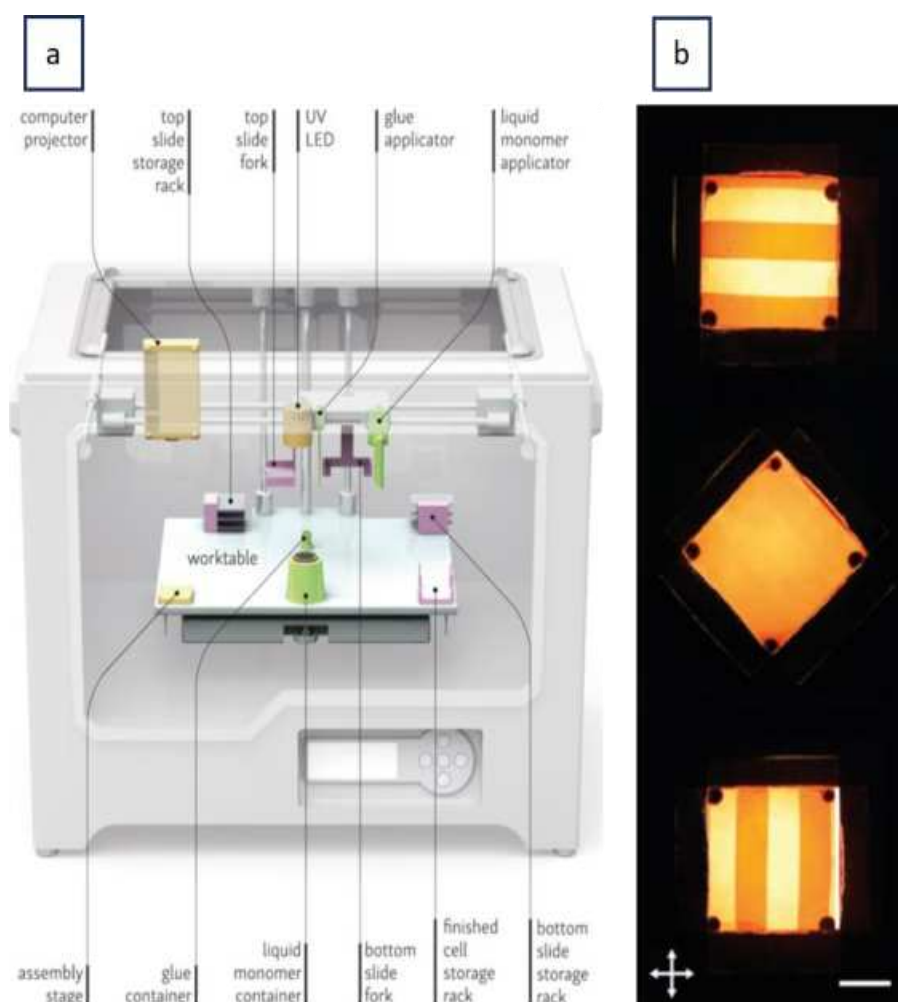


Fig. 15 (a) Workstation for LCE film fabrication with labelled components. The device is capable of producing multiple samples in a single run, provided that the LCE precursor, microsphere-doped glue, and glass slides coated with a photoalignment layer are supplied. The user predefines the projected images to program the desired molecular alignment. (b) POM images of an LCE film with patterned alignment, fabricated using the workstation with a precursor doped with an azo dye. The black dots visible in the corners of the sample are glue deposits applied during the fabrication of the sandwich-like alignment cell. Scale bar: 5 mm. Adapted from **Publication I**.

The fabrication process begins with photoalignment: RoboLEC positions a glass slide under a computer-controlled projector that illuminates the alignment layer with polarized light according to the uploaded image files. The setup supports projection using two orthogonal linear polarizations. After preparing the slides, the system assembles the sandwich cell by joining the glass substrates with the microsphere-doped glue. The cell is then filled with the LCE precursor via capillary action, with the workstation stage heated to reduce the precursor's viscosity. Once filled, the cell is cooled below the temperature of the isotropic–liquid crystal phase transition (clearing temperature), and then cured using UV light. The polymerized cell is subsequently transferred to a storage rack. This fabrication cycle can be repeated automatically until the machine runs out of materials such as glass slides, precursor, or glue.

The only requirement for compatibility with this setup is that the precursor must be UV-curable. This condition allows for a wide range of formulations, including those doped with light-absorbing dyes for photothermal actuation (see Fig. 15b). As a result, RoboLEC provides a versatile platform for producing aligned LCE films tailored to specific functions, including soft actuators, microrobotic elements, and optomechanical systems.

Light-driven systems

Among the various external stimuli such as heat, electric and magnetic fields, humidity, or pH, light stands out as particularly advantageous for remote activation [73–75]. In the context of LCEs, photothermal actuation provides precise spatial and temporal control, as light can be delivered from a distance, focused onto small regions, or applied uniformly over large areas. Local heating via light absorption rapidly induces the anisotropic–isotropic phase transition, enabling fast and reversible deformation-actuation frequencies as high as 100 Hz have been reported in microstructures [20]. However, the use of light also introduces technical challenges, including the need for optical modulation systems, the risk of surface overheating, and the necessity of incorporating light-absorbing dopants. Despite these limitations, light remains one of the most powerful and adaptable stimuli for driving LCE-based actuators, particularly at the centimeter scale and below, where conventional wiring or thermal systems become impractical.

This photothermal strategy was employed throughout the experimental studies presented in this dissertation. A 532 nm laser served as the actuation source, while LCEs doped with azo dyes (Disperse Red 1 or Disperse Orange 3) ensured efficient light absorption and local heating. To enable dynamic activation, the laser beam was directed using a system of mirrors with electronically controlled positioning, allowing for periodic and localized heating of selected material regions. This setup facilitated the development of untethered systems capable of programmable, reversible motion.

In the following paragraphs, I describe a series of light-driven devices that utilize this actuation mechanism. These projects contribute to the realization of the concept of “material as the machine” [76, 77], where functional behavior is encoded directly into the material’s structure and response. The systems presented here are grouped into two categories: bio-inspired robots, which replicate locomotion strategies observed in nature, and motors, which generate continuous rotary or linear motion. Each project addresses distinct challenges, highlighting different aspects of photothermal actuation and demonstrating the versatility of this approach for future applications in soft robotics and microscale actuation.

Light-driven Bio-inspired Robots

The development of soft robots inspired by nature offers a powerful strategy for creating untethered, adaptive systems capable of operating at small scales. Mimicking biological organisms provides not only conceptual and aesthetic inspiration but also functional advantages. Many living systems have evolved efficient mechanisms for locomotion, manipulation, and environmental adaptation across a wide range of body structures and size scales [18]. Bio-inspired designs can also simplify control by embedding behavior directly into the mechanical structure, similar to what is observed in natural organisms. When LCEs are used as the active material, these strategies can be translated into synthetic systems that deform smoothly, respond to external stimuli, and navigate complex environments without rigid mechanical components.

Depending on the intended actuation or locomotion mechanism, different illumination strategies can be employed. Uniform, continuous light can induce simple global deformations, but with appropriately designed geometries, it may also trigger self-oscillations through self-shadowing effects, enabling locomotion without the need for modulated illumination (see Fig. 16b). In contrast, chopped light, applied as periodic cycles of illumination and darkness, can introduce temporal asymmetry that is essential for breaking motion symmetry and generating directional movement (see Fig. 16a). More sophisticated control can be achieved by scanning a laser beam across specific regions of the material, allowing for precise spatiotemporal heating and programmable actuation patterns. One of the earliest experimental demonstrations of this approach in LCEs was a robot mimicking caterpillar locomotion, which I authored as the primary contributor in a collaborative published study during my BSc studies (see Fig. 16c). This level of selectivity also enables coordination of multiple limbs or segments within a single robot.

Ultimately, light-driven bio-inspired soft robots offer great potential for applications that require autonomous operation, minimal invasiveness, and high adaptability, such as targeted drug delivery, environmental sensing, or microassembly.

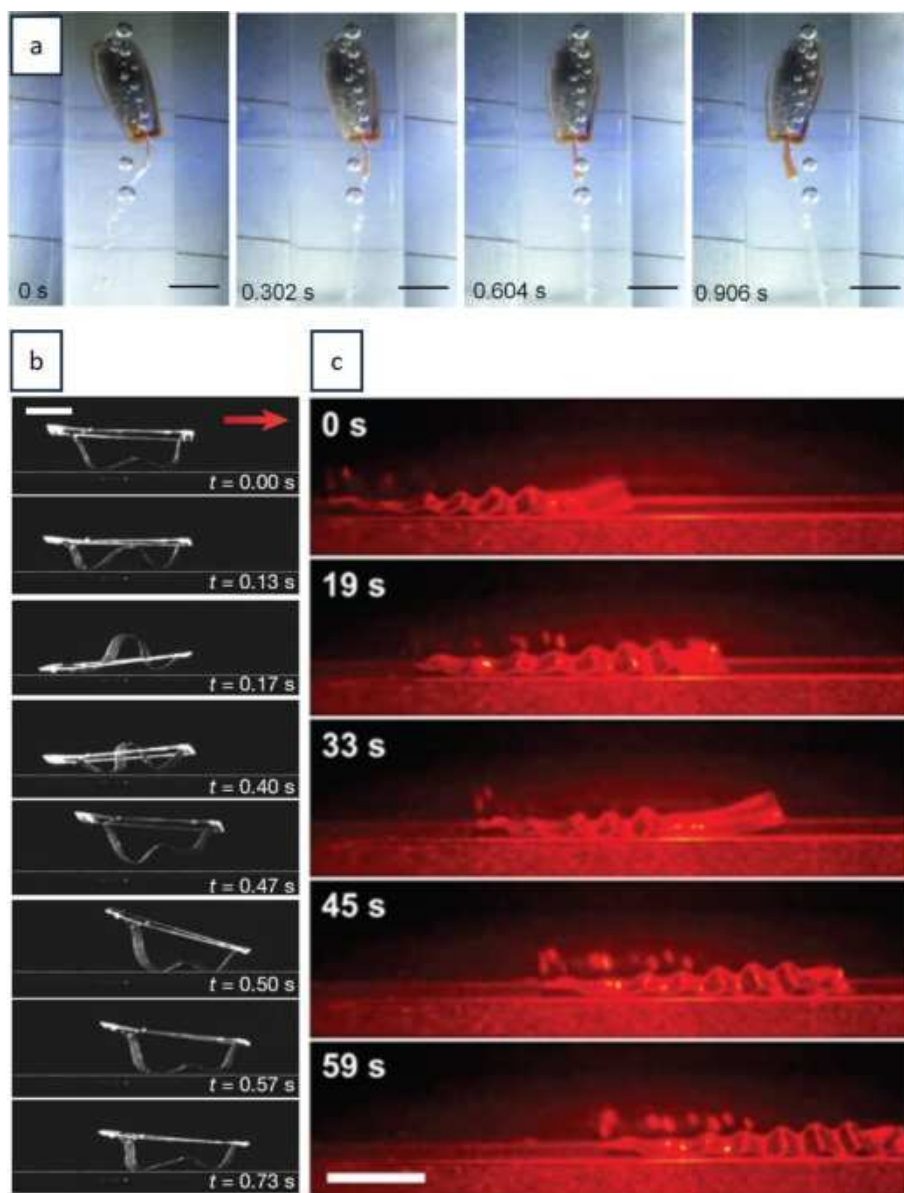


Fig. 16 Different illumination strategies used in light-driven soft robots. (a) Spermatozoid-inspired robot driven by chopped light. The robot's head and most of its tail are made of passive polymer, while the orange segment consists of light-responsive LCE. Swinging motion in a viscous liquid is induced by alternating illumination from the left and right, causing bending toward the light source due to differential light absorption across the LCE thickness. (b) LCE-based crawler moving across a glass surface under uniform illumination. The LCE film, attached to a rigid frame, undergoes self-oscillation via a self-shadowing effect. Periodic oscillations of the film drive locomotion through asymmetric frictional interactions with the glass surface. The upper glass surface is indicated by a dashed line. (c) Caterpillar-like crawling of an LCE film actuated by a scanning laser beam. The molecular alignment was preprogrammed to produce accordion-like deformation under localized heating. Scale bars: 3 mm for (a), 5 mm for (b) and (c). Adapted from [78] (a), [21] (b), and [79] (c).

These concepts were explored throughout my research, including in **Publication II**, published as an invited paper in the special issue *Polish Achievements in Materials Science and Engineering* of the journal *Materials*. The article provides an overview of the development of light-responsive LCE-based mechanisms within the research group in which I conducted my doctoral work, placing these efforts in the broader context of microscale robotics and actuator design. Although formally categorized as a

review article, it includes original results. In particular, the section describing the millimeter-scale water strider robot (see Fig. 17a) contains previously unpublished data. This project was a direct continuation of my earlier work during my bachelor's studies and represented a significant advancement in the development of light-driven soft robotic systems within the group, particularly because earlier prototypes were substantially larger (see Fig. 17b).

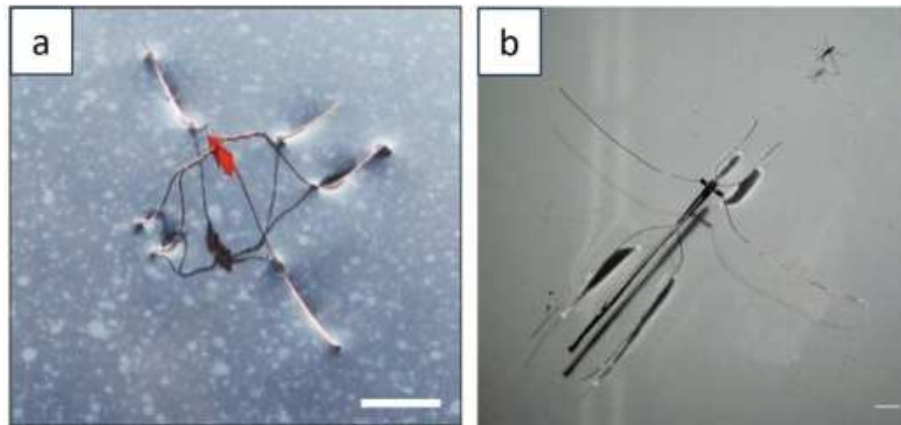


Fig. 17 Comparison between a living water strider and its robotic counterparts. (a) A light-driven water strider robot with a body and legs made of copper wire, actuated by a bending LCE film (orange) that drives the leg movement. (b) An adult water strider (smaller) next to a robot constructed from mechanical components. Scale bars: 1 cm. Adapted from [80] (a) and **Publication II** (b).

The water strider robot, despite its slow speed of 180 mm/min – approximately 6 000 times lower than that of its biological counterpart [80] – represents a significant step toward replicating the locomotion strategies of water striders at their natural scale. The design demonstrates that macroscopic soft actuators fabricated from liquid crystalline elastomers can generate sufficient force and directional control to achieve locomotion on water. Propulsion is achieved through an asymmetric actuation cycle: the LCE actuator bends and relaxes the attached copper wire legs with different timing under chopped light illumination, resulting in net movement. Importantly, the robot demonstrates the potential for coordinated multi-limb movement through controlled illumination. To the best of my knowledge, this was the first robot to mimic a water strider at its natural scale, and to date, only one other approach at this scale has been reported [11].

It may seem intuitive that research on soft robot locomotion should primarily prioritize speed and take inspiration from fast-moving organisms. However, some slow-moving creatures display highly effective locomotion strategies that are equally worth emulating. Snails, for example, can traverse a wide variety of surfaces, including vertical ones, thanks to the combined effect of their soft bodies and mucus secretion [81, 82]. Despite its effectiveness in nature, adhesive locomotion has remained relatively underexplored in robotics, with earlier implementations typically relying on mechanically driven systems [83] rather than on soft, deformable materials.

Publication III presents a light-powered soft robot based on a continuous LCE actuator that mimics the pedal-wave locomotion of terrestrial gastropods. Locomotion is achieved through a laser beam that scans along the length of the robot body, creating a moving region of localized heating. This spatiotemporal modulation of illumination induces traveling contractions in the LCE strip (see Fig. 18a), generating forward propulsion in a manner analogous to the contractile waves used by snails. A synthetic mucus layer provides both adhesion and lubrication, enabling stable crawling on various surfaces, including smooth glass, vertical walls, and ceilings. Although the robot operates at relatively low speeds, it demonstrates that mucus-assisted, light-driven crawling is possible at the millimeter scale. Its ability to navigate complex terrains (see Fig. 18b) highlights the potential of LCE-based systems in soft robotics and provides a platform for studying biologically inspired adhesive locomotion. Moreover, the observation that the robot pushes the mucus in the direction of movement inspired my colleagues to investigate this phenomenon further, resulting in a separate study on a light-driven peristaltic pump [84].

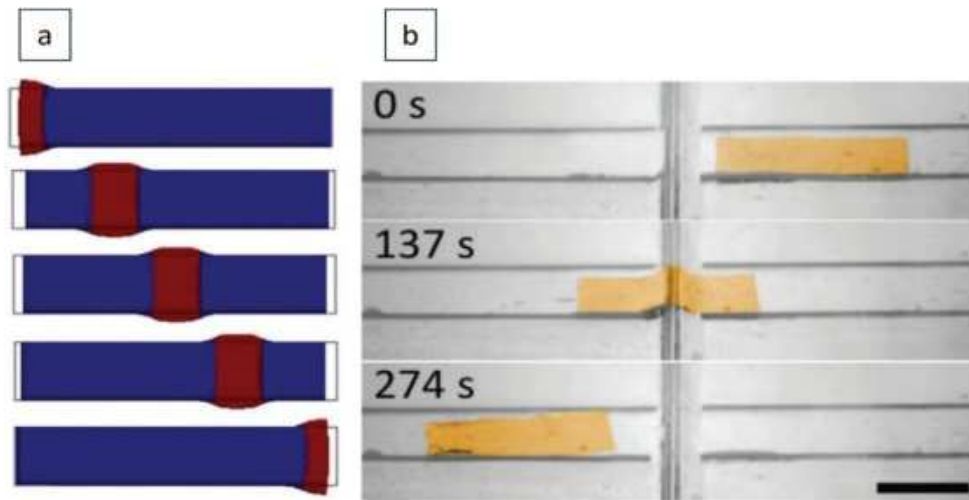


Fig. 18 Light-driven snail robot. (a) Finite element simulation of photo-thermally induced deformation propagating along the robot's body during a single laser scan. (b) The robot traversing an obstacle. Scale bar: 5 mm. Adapted from **Publication III**.

Light-driven Motors

Rotary motion is particularly challenging to achieve in soft, untethered systems due to the need for continuous asymmetry in actuation. Nevertheless, its fundamental role in mechanical systems across our civilization makes the development of rotary soft motors a problem worth solving. In my view, one of the most impressive solutions to this challenge comes from the group of Ikeda [85]. The core idea is to place a belt made of a LCE laminated film over two pulleys and illuminate one pulley with UV and the other with visible light (see Fig. 19a). Trans-cis photoisomerization induced by UV light and its reversal under visible light, combined with spatially asymmetric illumination, causes the belt to deform in a way that drives continuous rotation of the motor (see Fig. 19b).

An alternative approach involves the use of LCE fibers that contract upon light illumination. Conceptual designs based on this principle include a mechanical turbine-based engine [85] and an analogue of a four-stroke engine [86]. The proposed turbine consists of two spindles around which an LCE fiber is wound, with an inextensible wire coupling the spindles to ensure synchronized rotation. Asymmetric actuation is achieved by illuminating only the bottom part of the spindles (see Fig. 19c). In the four-stroke engine analogue, the piston and cylinder are replaced by an LCE fiber, and directional motion is induced by selectively illuminating only one side of the motor (see Fig. 19d). However, both constructions remain theoretical and have not yet been realized as functional systems.

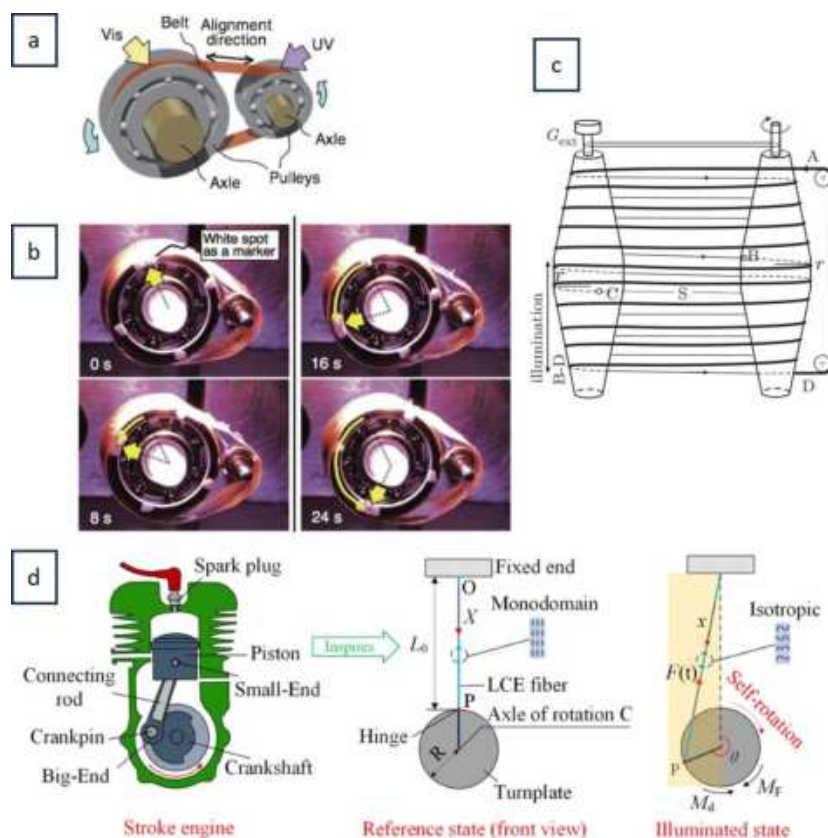


Fig. 19 (a) Schematic of a rotary motor using a belt made of LCE laminated film looped around two pulleys. Static illumination of different motor parts with visible and UV light induces continuous rotary motion through photoisomerization. (b) Experimental realization of the motor shown in (a), demonstrating rotary motion. (c) Conceptual design of light-powered turbines. An LCE fiber is wound around two spindles connected at the top by an inextensible wire that ensures synchronized rotation. Motion is generated due to the nonuniform radius of the spindles combined with asymmetric light exposure, which creates a difference in mechanical response along the fiber. (d) Schematic of a conventional four-stroke engine and its LCE fiber analogue. One end of the fiber is fixed, while the other is attached to the engine's turnplate. Illumination of the fiber induces contraction, generating torque; once the fiber rotates out of the illuminated zone, it relaxes, allowing continuous rotation as the fiber re-enters the illuminated region, thus completing the cycle. Adapted from [85] (a, b), [86] (c), and [87] (d).

Ring-shaped piezoelectric ultrasonic motors are widely used in the autofocus systems of camera compound lenses. In these devices, electrical excitation generates a traveling wave of deformation within the piezoelectric ring, which propels a rotor through frictional contact [88–90] (see Fig. 20a). **Publication IV** presents a rotary motor inspired by this design, replacing the piezoelectric actuator with a light-responsive LCE film.

The motor consists of a ring-shaped segment cut from the aligned LCE film (5.5 mm outer diameter, 1.3 mm inner diameter), mounted on a steel needle serving as its rotation axis (see Fig. 20b). To enhance frictional coupling, the ring was placed on a sandpaper surface and actuated by a laser beam scanned around the ring using a mirror mounted on an electric motor (see Fig. 20c). This setup enabled cyclic heating of selected regions, creating spatially asymmetric thermal stimulation that generated unbalanced forces and resulted in unidirectional rotation.

The molecular alignment of the LCE film was defined by +1 topological defects, with the defect center aligned with the center of the ring to ensure continuous director rotation around the axis. This alignment was achieved using photoalignment based on azopolyimide, specifically synthesized for this purpose. The mesogens were oriented either azimuthally on both surfaces (A–A configuration) or azimuthally on one side and radially on the other (A–R configuration). While the A–R configuration enabled higher rotation speeds (up to approximately 1.4 rpm), the A–A configuration provided more stable and consistent motion, reaching a maximum speed of nearly 0.3 rpm.

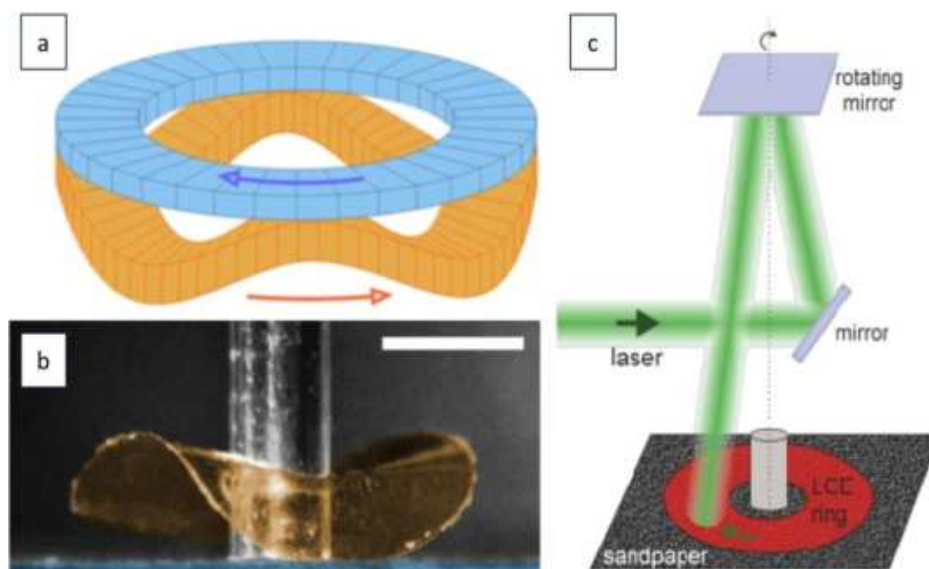


Fig. 20 (a) Schematic of a piezoelectric ultrasonic motor. Dynamic deformation induced in the stator (orange) is transferred to the rotor ring (blue) through frictional interaction. (b) Photograph of the LCE-based rotary motor used in the experiment. The LCE ring is mounted on a steel needle. Scale bar: 2 mm. (c) Schematic of the experimental setup. The LCE ring is placed on sandpaper, and a laser beam reflected from a rotating mirror scans the ring, inducing rotary motion. Adapted from **Publication IV**.

Despite its low speed and modest efficiency, this motor introduces a novel concept in micro-scale mechanics, opening the door to a new class of light-powered, friction-coupled actuators. Such systems may be especially suitable for applications requiring wireless control or operation in environments where electrical currents are undesirable or impractical.

One of the fundamental challenges in developing functional devices based on LCEs is their limited blocking stress [3], defined as the maximum stress an actuator can generate when its deformation is fully constrained. This limitation arises from the rubbery nature of the material and becomes particularly significant in applications that require reversible and controllable motion, such as linear motors or precise positioning systems. In these cases, actuators must not only generate sufficient force to initiate movement but also resist external loads when stationary. However, this remains difficult for LCEs, which primarily produce contractile strain rather than effective pushing forces.

A promising solution to this problem is presented in **Publication V**, which introduces a light-powered inchworm motor. The design is inspired by inchworm motors originally developed in the early 1990s using piezoelectric elements [91, 92]. Instead of relying on continuous mechanical pushing or pulling, these motors operate through friction-based, stepwise displacement. The LCE-based motor described in **Publication V** follows a similar principle but replaces the piezo actuators with light-responsive LCE elements. These actuators are designed to deform into an accordion-like shape (Fig. 21a), enabling significantly greater contraction than is achievable with uniformly aligned LCE elements. To enhance blocking stress by increasing friction with the shaft, a lead block with sandpaper attached to its underside was mounted on top of the actuators. In this configuration, the LCE actuators generate sufficient force to lift and shift the block during actuation, while the added friction ensures that the shaft remains securely held between steps. When illuminated with a scanning laser beam, the motor performs a cycle of gripping the shaft, shifting it forward, releasing it, and returning to its original shape (Fig. 21b). The direction of motion is determined by the direction of the laser scan.

To enable this accordion-like deformation, a new technique was developed to program the molecular alignment within the LCE film: rubbing overwriting. It involves rubbing the substrate twice: first uniformly across the entire surface, and then, for the second time, in a perpendicular direction through a mask. The second rubbing overwrites the initial alignment in the exposed areas, creating spatially patterned alignment domains (see Fig. 21c). These domains respond differently to light, resulting in the desired localized bending pattern.

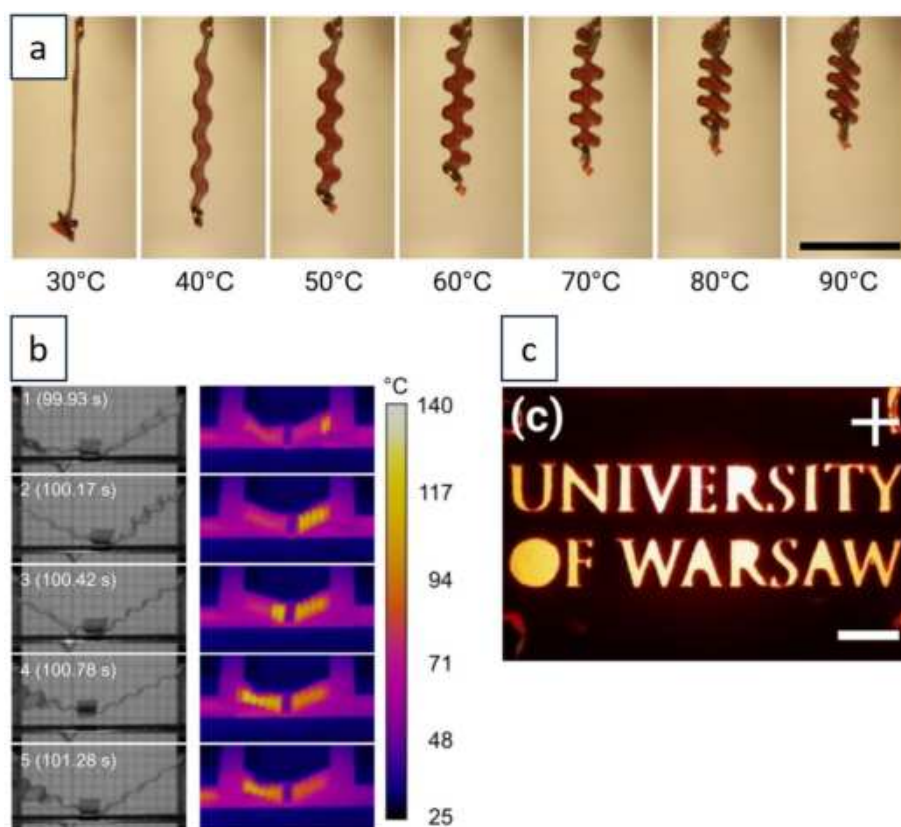


Fig. 21 (a) Accordion-like deformation of an LCE actuator at different temperatures. (b) Operating cycle of the linear motor captured using CMOS and infrared cameras (left and right, respectively): (1) grip the shaft, (2) shift the shaft, (3–4) release the shaft, and (5) grip the shaft again. The grid visible in the CMOS images has one millimeter spacing. The triangular marker positioned below the shaft allows for tracking its position. (c) POM image of an LCE film with molecular alignment patterned into the University of Warsaw logo using the rubbing overwriting technique. Scale bar: 5 mm. Adapted from **Publication V**.

While traditional rubbing is typically used to produce uniform alignment, the overwriting approach significantly expands its potential by enabling programmable, non-uniform actuation. By applying multiple rubbing steps with different masks and orientations, increasingly complex alignment patterns can be created. To the best of my knowledge, this is the first reported application of the rubbing overwriting method to program deformation patterns in LCE actuators. Previously, this technique had only been demonstrated for aligning liquid crystals [93].

Publication I

Automated photo-aligned liquid crystal elastomer film fabrication with a low-tech, home-built robotic workstation



OPEN

Automated photo-aligned liquid crystal elastomer film fabrication with a low-tech, home-built robotic workstation

Przemysław Grabowski, Bartosz Fabjanowicz, Magdalena Podgórska, Mikołaj Rogóż & Piotr Wasylczyk✉

Laboratory procedures are often considered so unique that automating them is not economically justified – time and resources invested in designing, building and calibrating the machines are unlikely to pay off. This is particularly true if cheap labour force (technicians or students) is available. Yet, with increasing availability and dropping prices of many off-the-shelf components such as motorised stages, grippers, light sources (LEDs and lasers), detectors (high resolution, fast cameras), as well as user-friendly programmable microprocessors, many of the repeatable tasks may soon be within reach of either custom-built or universal lab robots. Building on our previous work on fabrication, characterization and applications of light-responsive liquid crystal elastomers (LCEs) in micro-robotics and micro-mechanics, in this paper we present a robotic workstation that can make LCE films with arbitrary molecular orientation. Based on a commercial 3D printer, the RoboLEC (Robot for LCE fabrication) performs precision component handling, structured light illumination, liquid dispensing and UV-triggered polymerization, within a four-hour-long procedure. Thus fabricated films with patterned molecular orientation are compared to the same, but handmade, structures.

In both utopian and dystopian visions of the future of human labor, scientific jobs are usually considered to be on the safe side. Yet, on second thought, large swaths of research work may in fact be much easier to automate than we are ready to acknowledge^{1,2}.

Many laboratory activities seem to be so unique that automating them may at first appear to be well beyond what is economically justified—designing, building, testing and calibrating the (usually one-off) laboratory equipment needed would take longer than doing the task manually; this is particularly true if cheap labor is available. Yet, with increasing availability of components such as motorized stages, grippers, light sources (LEDs and lasers covering UV, visible and NIR bands), detectors (high resolution, fast cameras), and other mechanical and optical components, as well as programmable microprocessors, many daily, mundane, repeatable tasks may soon come to be automated, either by researchers themselves or by companies delivering the necessary solutions. Apart from the increased throughput, such automatization would also eliminate the human factor which often contributes to errors in complex laboratory procedures. Robotic workstations guarantee precision timing, temperature settings and reliable control of other parameters without subjective estimation.

There are two areas where automation may come into play in research: software (data acquisition, storage and processing) and hardware (sample preparation, handling, disposal). Whereas the substantial progress made in software is beyond doubt, hardware, in principle, is considered to have a much higher entry threshold. In a typical laboratory setting—be it chemistry, biology, materials science, or solid state physics—automating even part of the workflow of sample preparation and handling would require expertise in precision mechanics, electronics, robotics, perhaps combined with optics, machine vision, haptics, etc. As a result it may prove prohibitively difficult to automate even the simplest parts of the workflow, especially if cheap labor is readily available—many technicians as well as MSc and PhD students spend most of their time performing routine, repeatable tasks, like pipetting hundreds and thousands of samples for standard measurement procedures or transferring samples from one apparatus to the next.

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At the same time, many solutions and sub-systems are becoming more and more easily available at the hardware end. Precision mechanics, computer-controlled drives and actuators, sensors and cameras have entered the mass market, which has dramatically boosted availability and reduced prices.

The life sciences have already benefited from laboratory automation in processing large numbers of samples. High-throughput workstations have been demonstrated for genome screening, protein purification and drug discovery, consisting of robots preparing samples (e.g. setting up transfections), moving them around (in well plates, tubes or cell culture flasks) between liquid handlers, centrifuges, incubators and plate readers³. While such workstations require teams of engineers (and substantial resources) to design and set up, cheap automated sample preparation and imaging is also possible, with authors often sharing the designs openly.

A pipetting robot has been developed, combined with a modular optical system and open-source software for microscopy and image analysis⁴. In high-throughput chemistry research—searching for photocatalysts for hydrogen production from water—a computer controlled system has been demonstrated, with a mobile robotic arm navigating through the laboratory, handling samples between several pieces of equipment, interpreting the results and optimizing the output⁵. Similar systems have also been scaled down (in size and cost) to fit onto a single desktop⁶.

In a robotic platform developed for organic flow synthesis, a robotic arm assembles modular units (reactors/separators) and connects reagent lines and pumps to the setup; after the synthesis is completed, the robot removes the modules to their storage⁷. The Chemputer project, in turn, sought to develop a computer program that would translate abstract chemical procedures into instructions for modular hardware of a laboratory-scale synthesis robot. The robotic platform was set on a fixed fluidic backbone, made of pumps and tubes connected to different modules: a reaction flask, temperature controlled filtration setup, liquid–liquid separator and solvent evaporation module⁸.

Inspired by such examples, in order to explore current perspectives in material (polymer) science research automation, we designed and built a compact workstation for liquid crystal elastomer (LCE) film fabrication, which we called “RoboLEC” (Robot for LCE fabrication). LCE films have been used as light- or temperature-responsive actuators in soft, millimeter-scale robotics⁹ and micromechanics¹⁰. In one of the possible approaches, they are prepared by photopolymerization of liquid monomers in glass cells, where the orienting layers on the glass surface (prepared by e.g. photoalignment) determine the molecular order and orientation within the film, and thus its optical properties and photo-mechanical response¹¹. The ultimate goal here is a technology that would deliver LCE films with any pre-programmed director orientation within the material volume¹². In the case of thin films, arbitrary molecular orientation means that there may be any (position-dependent) director orientation at one film surface and a different director orientation on the other surface. Some of the automated processes realized in our device are used in the industry, in particular photoalignment and photopatterning has been introduced in fabrication of liquid crystal displays (LCDs)—see^{13–16} for a review of developments therein.

Methods

LCE film fabrication involves several stages: preparation and precision handling of glass slides that, once glued with a spacer in between, make up the cells, structured light exposure to prepare orienting layers via photoalignment¹¹, and monomer mixture polymerization by UV exposure. Some of these must be performed at controlled temperatures (typically between room temperature and 200 °C).

The concept for our RoboLEC workstation originated from the realization that most of the steps can be straightforwardly implemented in a standard extrusion 3D printer, with minor modification to the hardware. A Flashforge Creator Pro 3D printer (227 × 148 × 150 mm³ workspace) was used as the platform and was connected to the internet with OctoPrint plugin for Raspberry Pi 3 to enable the G-CODE control. The extruders and fans on the printer's head were replaced with a set of custom-made tools: two forks for glass slide handling, a glue dispenser, a heated LCE mixture applicator, and a UV (395 nm) LED for photopolymerization. Two storage racks for glass slides and one for finished cells, a stage for the alignment and assembly of cell parts, a glue container, and a heated LCE monomer container were mounted on the printer's heated worktable. A miniature computer laser projector with a set of polarizers was installed for the photoalignment pattern generation—compare Fig. 1 and Movie 1 in the Supplementary Information. Most of the added parts were either machined from metal or 3D printed from polymers (see Table 1 in the Supplementary Information for the list of components).

Results

RoboLEC's process of automated LCE film fabrication, presented in ten consecutive stages in Fig. 1a, starts with lifting the first (top) glass slide (26 × 38 × 1 mm³, one half of a standard microscope slide) from the storage rack and placing it onto the assembly stage. The glass slides have been prepared by spin-coating with a photoalignment azopolyimide (PI) solution (~1%) in N-methyl-2-pyrrolidone, dried, and heated at 130 °C for 2.5 h to evaporate the solvent¹⁷. At the assembly stage, the computer laser projector illuminates the glass coated with the photoalignment layer with arbitrary patterns through one or two perpendicular polarizers (the blue light intensity on the cell surface is 1 mW/cm²)¹⁸. The slide is then moved back to the storage rack, and the second (bottom) slide is delivered from the other storage rack to the assembly stage for the exposure of its photoalignment layer. At this point the printer's worktable and the monomer mixture applicator are starting to heat up. The UV-curable glue mixed with spacers (calibrated glass balls, typically between 5 and 100 μm diameter, depending on the desired LCE film thickness) is applied in the four corners of the bottom slide. The top slide is placed onto the bottom one and the glue is cured by means of UV LED exposure for 10–50 s with the light intensity on the cell surface equal 18 mW/cm². When the cell temperature stabilizes at 80 °C, it is filled with the liquid (molten) monomer mixture, transferred from its container on the heated dispenser tip (in several rounds, if needed for the cell to be completely filled by capillary forces). Finally, the worktable is cooled down and kept at 40 °C for 5 min to allow

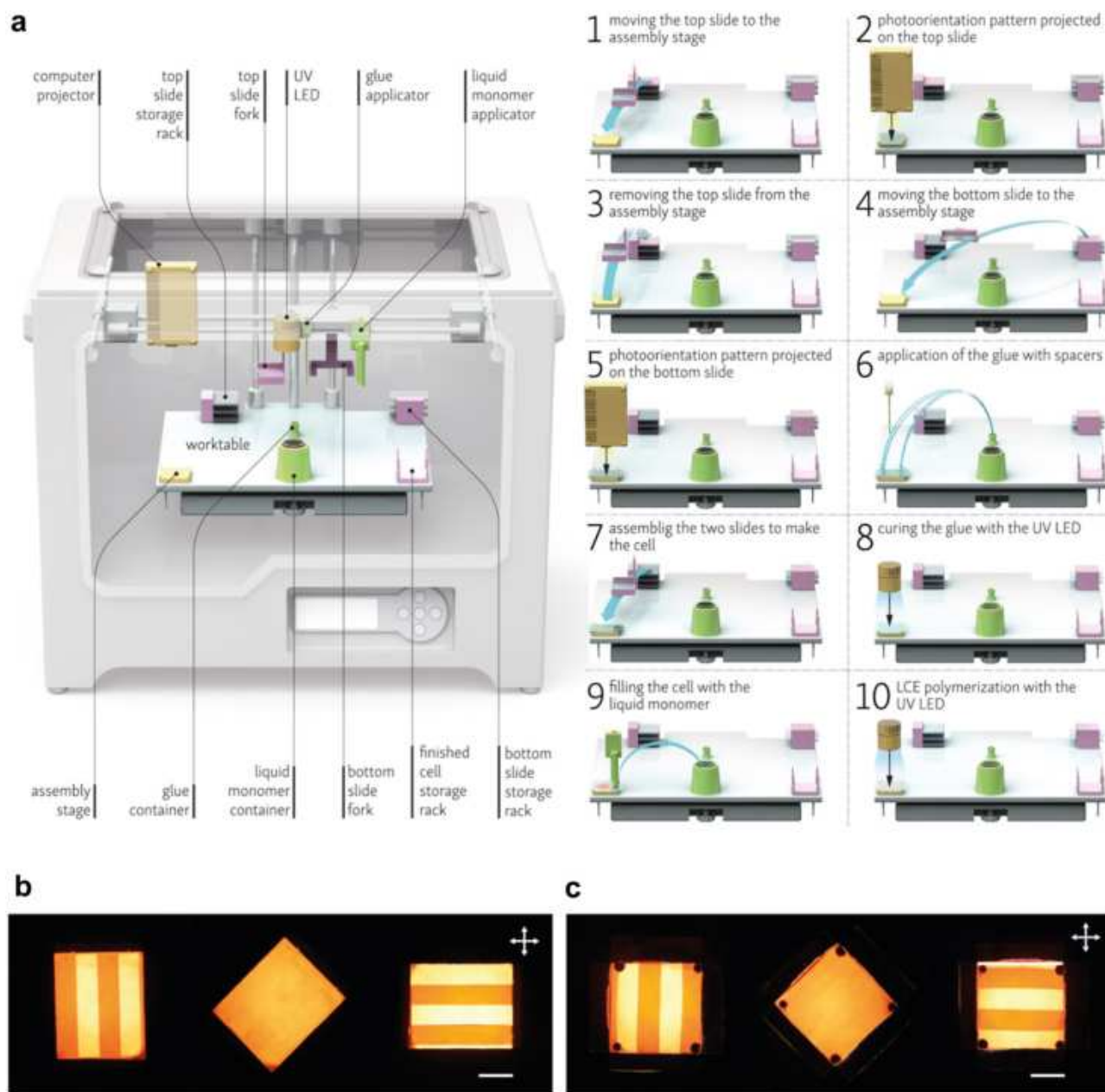


Figure 1. The RoboLEC robotic workstation for automated liquid crystal elastomer (LCE) film fabrication. The framework uses an extrusion 3D printer with several modifications: storage racks for the glass slides, containers for the UV glue and the LCE molten mixture, and a laser projector. Figure shows ten steps of the photoaligned LCE film fabrication (a), and LCE films with patterned alignment: handmade (b) and from the RoboLEC (c). Polarized white light photographs at three different orientations, the cross shows the polarizer orientation, the scale bar is 5 mm long.

the liquid crystal monomer to orient and the mixture is polymerized by illumination with the same UV LED for 6 min. After the finished cell is transferred to the storage rack, RoboLEC is ready for the next cycle. The entire cycle takes around 4 h and 40 min, of which 4 h is taken up by the exposure of the photoalignment layers and about 30 min by heating and cooling of the worktable (compare Table 2 in the Supplementary Information for a detailed break-down of the cycle). The cells remain closed at the end of the cycle, as for some applications they may need to be opened under specific conditions or just before the next step of preparation.

To demonstrate RoboLEC's capacity for automated LCE film fabrication with complex director patterns, we fabricated 50 μm thick films with twisted nematic orientation alternating between neighboring, 3 mm wide stripes. Previously such films, used to make light-driven actuators, have been prepared with manual assembly of glass cells with either photoalignment layers¹⁹ or patterned rubbing through masks^{20,21} and they have been used in millimeter-scale light-powered crawling robots²⁰ and micromechanical devices, e.g. a linear stepping motor²¹. Inspection of the hand-made film with patterned alignment and the film of the same type fabricated with

RoboLEC with polarized light microscopy shows that both films exhibit the desired molecular order (Fig. 1b,c)—sharp boundaries between areas with different orientation are visible and high contrast between them indicates high degree of molecular order. We have also tested the reproducibility of the process by running it a few times and comparing the resulting films (see Supplementary Information for details).

Conclusions and outlook

Using a few of-the-shelf components, the most expensive being the 3D extrusion printer, we were able to build a robotic workstation for LCE film fabrication. As the glass slides used for the cells have to be spin-coated with the polyimide layer for photo-alignment, and the monomer mixture has to be prepared beforehand, the process can be considered semi-automated. In the current prototype, the racks were design to hold three plates for bottom and top of the cell. Within the same mechanical framework, this can be extended to ten, which would already allow for ten cells to be made without reloading, taking more than 40 h of continuous work. For even more cells, a separate glass plate feeder (e.g. carousel) can be added. The next generation of RoboLEC may also feature a rotating polarizer for photoalignment, thus opening up the possibility of fully automated preparation of LCE films with arbitrary 2.5 D director distribution¹². This demonstrates how repeatable manual tasks—handling and assembling of small objects, fluid dispensing, image projection—can be automated with reasonable effort and expense. Thus, preparing a series of LCE films becomes possible with limited operator time needed to prepare the components (glass plates, glue with spacer(s), monomer mixture) and to set up the process. While the time required to prepare an LCE film remains the same as with the manual handling, the robotic system offers the capability of an unsupervised search for the optimum parameters—a procedure often involved in materials science—as it can produce several films with varying parameters, such as compound concentration, thickness, polymerization time, or temperature at different stages of the process and, with some modification, characterize their parameters, e.g. molecular orientation, with polarized light transmission measurements.

More broadly, our results show that automation of repeatable laboratory tasks is now within reach of even small research teams. This may evolve in two directions: one involves custom-made systems, designed for specific, unique applications, while the other, even more interesting, may offer flexible, modular systems that could integrate various units into automated workstations. Time will tell how these two branches will evolve and be adopted, both by the laboratory equipment manufacturers and by their potential users.

Data availability

The datasets used and/or analyzed during the current study are available from the corresponding author on reasonable request.

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Author contributions

P.G. coined the idea and with B.F. built the setup, performed the experiments and analyzed the data. M.P. and M.R. developed the photopatterning technique. P.W. supervised the project and wrote the manuscript.

Competing interests

The authors declare no competing interests.

Additional information

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Publication II

**From Light-Powered Motors, to Micro-Grippers, to Crawling Caterpillars, Snails and Beyond—
Light-Responsive Oriented Polymers in Action**

Review

From Light-Powered Motors, to Micro-Grippers, to Crawling Caterpillars, Snails and Beyond—Light-Responsive Oriented Polymers in Action

Mikołaj Rogóż , Zofia Dziekan, Klaudia Dradrach, Michał Zmysłony, Paweł Nałęcz-Jawecki, Przemysław Grabowski, Bartosz Fabjanowicz, Magdalena Podgórska, Anna Kudzia and Piotr Wasylczyk * 

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Abstract: “How would you build a robot, the size of a bacteria, powered by light, that would swim towards the light source, escape from it, or could be controlled by means of different light colors, intensities or polarizations?” This was the question that Professor Diederik Wiersma asked PW on a sunny spring day in 2012, when they first met at LENS—the European Laboratory of Nonlinear Spectroscopy—in Sesto Fiorentino, just outside Florence in northern Italy. It was not just a vague question, as Prof. Wiersma, then the LENS director and leader of one of its research groups, already had an idea (and an ERC grant) about how to actually make such micro-robots, using a class of light-responsive oriented polymers, liquid crystal elastomers (LCEs), combined with the most advanced fabrication technique—two-photon 3D laser photolithography. Indeed, over the next few years, the LCE technology, successfully married with the so-called direct laser writing at LENS, resulted in a 60 micrometer long walker developed in Prof. Wiersma’s group (as, surprisingly, walking at that stage proved to be easier than swimming). After completing his post-doc at LENS, PW returned to his home Faculty of Physics at the University of Warsaw, and started experimenting with LCE, both in micrometer and millimeter scales, in his newly established Photonic Nanostructure Facility. This paper is a review of how the ideas of using light-powered soft actuators in micromechanics and micro-robotics have been evolving in Warsaw over the last decade and what the outcomes have been so far.

Keywords: light-responsive materials; liquid crystal elastomers; micro-robotics; soft robotics; actuators

1. Introduction

A one-millimeter spider that can walk, jump, fly (on a gossamer), spin a super-strong web, catch prey and replicate for millions of generations (adapting along the way to the changing environment, if necessary), not only inspires awe, but also makes us realize how disappointing and, in most cases, futile are our attempts to mimic nature in the small scale. There is a glaring gap between the scale of meters or centimeters, where industrial mechanisms, machines and robots have a well-established footing, and the molecular scale, where we have gained enough understanding to make molecules dance at our will [1]. The smallest practical electrical motors available are approximately 1 mm in diameter, and given drivetrains of similar scale, a centimeter-long solar-powered walker might ultimately be possible [2]. With piezo drives, some spectacular results have been demonstrated, including a several-second, self-sustained flight [3] on the scale of a small dragonfly (albeit with very limited control).

The challenges in building small autonomous mechanisms are threefold:

- How to design the mechanics to overcome all the problems with friction, van der Waals forces, fluid viscosity and moving with very low Reynolds numbers [4]? The laws governing mechanics and motion in the micro-world are different from those

we are used to: gravity and the electromagnetic forces are governed by the same constants, but their relations change with scale. As a result, new challenges as well as new opportunities arise: crawling on an upside-down glass ceiling is easy for a millimeter-scale snail robot [5], while a micron-size walker may struggle to lift its legs, glued to the surface by van der Waals forces [6,7];

- How to fabricate motors, gears and other elements on the sub-millimeter and smaller scales, reliably and cheaply?
- How to supply the mechanisms with energy, either remotely or from an onboard source?

For around ten years now, our group at the Faculty of Physics at the University of Warsaw have been trying, with varying success, to approach microscale mechanics and micro-robotics with soft, light-responsive materials: liquid crystal elastomers. In what follows, we present our achievements to date and sketch out possible avenues of future work.

1.1. Liquid Crystal Elastomers as Light-Powered Actuators

Apart from traditional engines and gears, soft actuators capable of powering simple mechanisms and machines, driven by various stimuli, have been demonstrated in small scales [8]. In dielectric elastomer actuators (DEAs), a compressible membrane is sandwiched between flexible electrodes that interact via electrostatic forces [9], resulting in large strains, although with very high voltages needed [10]. Another approach to electrically driven actuators uses conjugated, conductive polymers [11], and asymmetric swelling of gels in ionic solutions can generate shape changes as well [12]. Magnetically driven actuators have also been developed, mostly based on soft polymers with suspended magnetic particles and spatiotemporally varying magnetic fields to control them [13,14]. Material deformation can also be induced by chemical reactions, e.g., selective modification of chemical bonds in polymers [15]. Pressure-driven soft actuators rely on structures with spatially varying stiffness, deforming upon expansion of gas- or liquid-filled chambers [16]—they can be scaled down [17], but still need substantial tubing to deliver the pressurized medium.

It was probably De Gennes who, around 1975, first came up with the idea of anisotropic crosslinked polymer networks with embedded orientational order of nematogenic monomers [18]. This, in turn, triggered various attempts towards fabricating, understanding and using stimuli-responsive-oriented polymers, in particular liquid crystal elastomers (LCEs). LCEs are solid, elastic polymers with a well-defined alignment of molecules (this alignment orientation is often described by the so-called director), just as in the familiar liquid crystals. If cleverly designed, both at the molecular (chemistry) and mesogenic (orientation) level, LCEs may exhibit rather unique mechanical properties, in particular very large, reversible and fast deformations in response to an external stimulus: heat [19]; light triggering photochemical reactions [20,21]; light inducing photo-thermal heating [19,22,23]; electric field [24,25] or the presence of a solvent [26].

Due to the tight packing of rod-shaped molecules, arranged in cross-linked polymer chains, the order (anisotropic) $\leftrightarrow$ disorder (isotropic) transition reduces the effective chain length along the alignment direction (the director), at the same time increasing the spacing between the molecules in the perpendicular directions (Figure 1A). As a result, the material deforms in a manner determined by: (a) the spatial distribution of the director; (b) the spatio-temporal distribution of the stimulus, e.g., light intensity; (c) the properties of the material itself (stiffness, elasticity, thermal conductivity) and (d) the surrounding environment (e.g., in our LCEs, the response will usually be different in the air (good thermal insulator) and under water (good thermal conductor)). If stimulated by light, additional degrees of freedom in the mechanical response may involve the light spectrum [27,28] or polarization [29,30]. Thus, LCE elements of various sizes and forms [31–33] can be used as micro-actuators in various configurations as well as remotely powered “muscles” for robots (Figure 1B, cf. [34] for a review of LCE actuators and [7] of LCE soft robots).

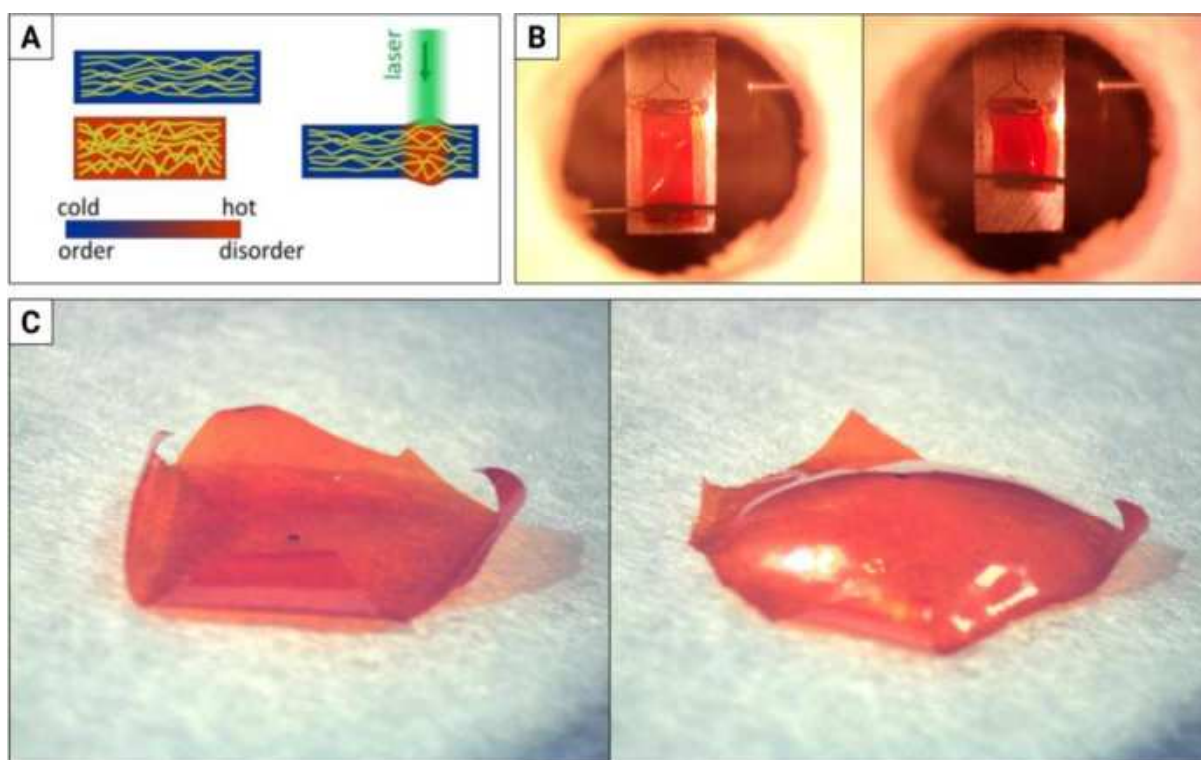


Figure 1. Light-responsive LCE—from a simple contracting strip to films with arbitrary molecular alignment. (A) A piece of material with cross-linked polymer chains (olive) arranged in one direction undergoes a phase change in response to temperature increase, either in the entire volume (left) or locally, when illuminated with a green laser beam (right): the molecular order decreases, the polymer chains effectively shorten along the director and macroscopic deformation results. (B) A 10 mm long strip of LCE film heated in an oven from 29 °C to 150 °C shortens by approximately 35%. (C) A 22 × 22 mm piece of flat LCE film with azimuthal director orientation (the black dot marks the center of rotational symmetry) buckles when heated on a hot plate from 22 °C to 80 °C.

From the material science and chemistry perspective, LCEs can be categorized by the synthesis pathways—the monomer may be a precursor and a synthesis from low-molar-mass LC is carried out, or a polymer chain (macromer) is grown first and then the material is crosslinked. In terms of the crosslinks' density, they can be loosely crosslinked liquid crystal polymers, called elastomers, or a highly crosslinked polymer network. As for the location of the mesogenic moieties, they can be built into the polymer backbone in the so-called main-chain LCE, or covalently bonded with the polymer backbone as side groups in the side-chain LCEs.

Following the first experimental demonstration by Finkelmann et al. [35,36], both main-chain and side-chain LCEs were synthesized via different synthesis routes. In the so-called “Finkelmann method”, elastomers with Si-O in the polymer backbone are obtained, capable of high strains, but hydrolyzation reaction is time-consuming and sensitive to impurities and the presence of oxygen. Much later, the “click-chemistry” found its way into the LCE synthesis. These reactions are not sensitive to oxygen and allow for the control of the LCE properties by tuning the structure of the crosslinked liquid crystal polymer [37,38]. The most notable examples are the Michael addition and thiol-ene reactions, leading to LCEs with mechanically programmable alignment, either by shear thinning (in 3D printing) or by surface anchoring [39]. It is also possible to quickly fabricate LCE's thin films—in a matter of hours—from low-molar-mass LCs, through acrylate homopolymerization. A side-chain LCE is formed with a relatively high number of crosslinks and the alignment in thin films can be induced by rubbing or photopatterning. Additionally, in these materials

a sensitizer (e.g., dyes) can be introduced into the matrix before as well as after polymerization and crosslinking.

There are two approaches to actuate LCEs with light. Additional molecules (e.g., dyes) can be embedded into the matrix that absorbs light of a specific wavelength. Most often azobenzene moieties are used, likely due to the extensive studies related to the photoisomerization of azobenzene itself and azobenzene derivatives since the 1960s. An azo-moiety can be built into the polymer backbone (as a monomer or a crosslinker [40–42]) and when exposed to light (typically in the UV band), isomerization occurs (trans molecules, supporting the nematic phase, bend and the order parameter in the polymer matrix decreases). After the irradiation stops, the bent molecules go back to their previous state, yet this reversal is slow (it can be sped up with exposure to visible light). Because of the slow response of these reactions (seconds to minutes), some light-responsive LCEs make use of molecules or nano-particles that absorb light and generate heat (which, in turn, reduces the order parameter and induces conformational changes) [43]. Usually, they are dispersed in the LCE matrix: dye molecules, carbon nanotubes [44] or gold nanoparticles [45,46], but coating an LCE actuator surface with NIR-absorbing polymers also works [47]. A combination of photochemical and photothermal responses in light-responsive liquid crystal polymer networks for soft actuators was also presented [48].

1.2. LCE Fabrication in Different Scales

LCE elements and structures are often fabricated by UV light-induced polymerization of the liquid monomer, crosslinker and photoinitiator mixture, typically in a glass cell, tens of micrometers thick. Properly prepared orienting surfaces—either rubbed mechanically or coated with photo-orienting layers—guarantee the director orientation within the LCE film in 2 or 2.5 dimensions, if the opposing layers have different properties (Figure 1C). Different elements, typically of the order of a few millimeters, can then be cut with a blade or a laser beam [49]. More complex director patterns—and thus film deformations—are available with patterning of the orienting layers [50–52]. Another fabrication method involves shaped molds with the director orientation induced by a magnetic field [53]. In extrusion 3D printing, the molecules are oriented by rheological effects while they flow through a small nozzle [54,55]. The most advanced technology so far—3D laser photolithography (direct laser writing)—uses a focused infrared laser beam that is scanned across a drop of liquid monomer, triggering polymerization via two-photon absorption in a volume (voxel) as small as a fraction of a cubic micron [56].

1.3. Problems Yet to Be Solved

While several simple devices have been demonstrated with contracting, bending, twisting or even more complex photo-responsive LCE actuators, typically only their basic response to the light stimulus has been studied. At the same time, even monochromatic light (e.g., in the form of a laser beam(s)) offers a number of degrees of freedom that can be used to control the mechanisms: average power; pulse duration; pulse energy and the light intensity spatial distribution. If the development of mechanisms and machines based on photo-responsive elastomers is to extend beyond single contracting or bending strips of a stimulus-responsive material, a better understanding of the photo-mechanical response of LCEs will be needed, including the interplay of the energy flow and heat dissipation with different light pulse durations and the influence of the light spatial intensity distribution.

Our studies of the millimeter-scale LCE actuators [57] hint that: (a) if used in the transient regime, the actuator response time is independent of laser power; (b) for pulsed laser actuation, long laser pulses result in a smaller actuator response, compared to short laser pulses delivering the same energy; this is true down to a certain pulse duration, determined by the time constants of heat transfer to the environment; (c) when part of the actuator is illuminated with a laser beam of constant total power, but of varying size, the photo-mechanical response is independent of the illuminated area.

Two of the biggest problems we have discovered so far are as follows:

- light absorption in the LCE films decreasing over time, mainly due to the light-absorbing dye bleaching; this can be potentially bypassed by using quantum dots or other, more robust absorbers, well known from fluorescence microscopy, where dye bleaching has been studied and addressed for some time;
- the wear and tear of the actuator after many cycles of operation; this calls for further insights into the mechanical properties of the light-responsive elements, probably at the level of polymer chemistry.

2. Micro-Motors—Direct Conversion of Light Energy into Mechanical Work

“Can light drive a motor?” was the opening question asked by Ikeda et al. [58] in 2008. Most energy-harvesting systems convert (solar) light energy either to heat (in solar thermal collectors) or to electricity (in solar cells). Direct conversion of light energy into mechanical energy has been demonstrated, from the molecular to macroscopic scales [59–61]. Ultimately, this may enable mechanical devices remotely powered with light (delivered either via free space or through optical fibers), where using electrical cables is not possible.

Yamada et al., in [44], demonstrated a light-driven motor using the contraction of an LCE/polyethylene laminated film irradiated with UV and visible light. The motor presented by Geng et al. [62] used a looped strip of hydroxypropyl cellulose film that deformed under humid air to drive a rotating element. The LCE rod described in [63] can roll with either a light beam or a heated surface as the energy source, but it remains an open question if this deserves to be called a “motor”. In a similar fashion, various LCE tubes and helical ribbons have been demonstrated as drives for centimeter-scale light-powered rolling vehicles [64].

Historically, we first developed micro-robots—the water strider, the caterpillar [50] and the snail [5]—only later to realize that the very same caterpillar, if held in place, can make a rotary motor [65] (if shaped into a ring—“eating its tail”) or a linear inchworm motor (if working in a team of two) [52].

2.1. Rotary Motor

Many high-end camera lenses use so-called ultrasonic motors (USMs) to drive the auto-focus mechanism. Typically, piezoelectric USMs are composed of a ring-shaped rotor and stator (Figure 2A). The voltage applied sequentially to the rotor segments induces travelling wave deformations that couple by friction to the stator, thus setting the former in motion [66–68].

Inspired by this design, we built a light-driven micromotor, where the travelling wave deformation results from the photo-mechanical response in a 5.5 mm diameter LCE ring, illuminated with a laser beam moving around the disc circumference (Figure 2B) [65]. In our experiments, we tested LCE discs with different director distributions, in particular, one with the azimuthal orientation on both sides (A-A) and one with the azimuthal and radial orientations (A-R). The A-R disc proved to be 10 times faster ($\omega = 5.88$ rad/min) and 14 times more efficient at converting laser beam revolutions to disc (rotor) revolutions—see [69] for the video of the rotating motor. We also studied these effects by performing finite element numerical simulation, where the dynamic photomechanical response of the LCE discs was modelled as a local strain tensor (Figure 2D).

Despite low speed and efficiency, the LCE micro-rotor has some advantages, such as the ability to be scaled down and to be powered remotely with light energy.

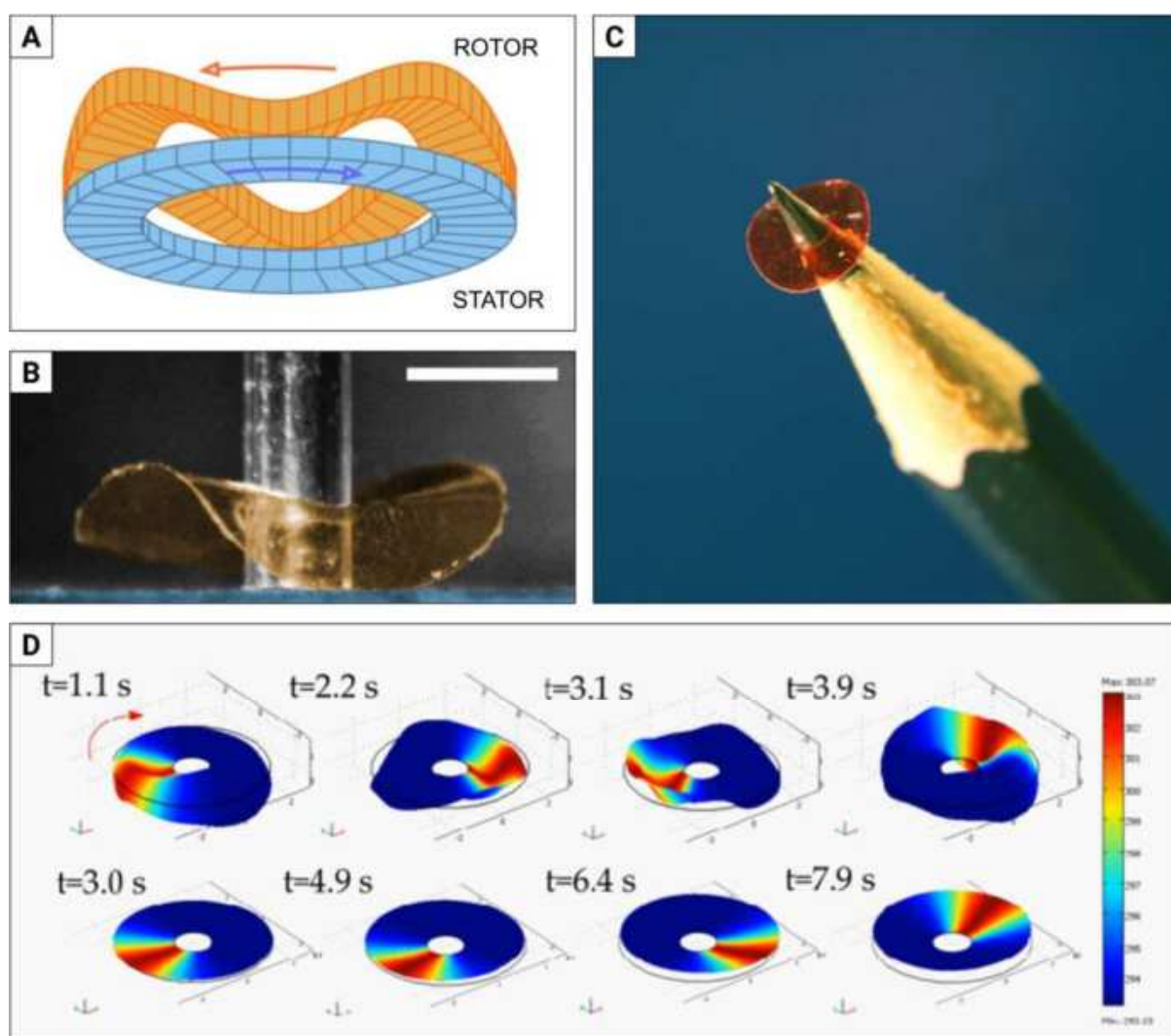


Figure 2. Rotary motor directly powered with light. (A) Schematic of the rotary piezoelectric (ultrasonic) motor. Travelling wave deformation generated in the rotor (orange) interacts via friction with the stator (blue) and sets the former in motion. (B) LCE rotary micro-motor, seen from the side. The LCE disc (rotor, orange) rotates with respect to the stator (rough solid surface, blue), around a steel axis. The white scale bar is 2 mm long. (C) The LCE rotor on a pencil tip for the scale demonstration (although, sadly, we must admit it cannot yet be used to sharpen pencils). (D) Numerical simulations of the LCE deformation upon illumination with a spatially modulated (rotating) laser beam. The top row is for azimuthal–radial, and the bottom row for azimuthal–azimuthal director orientation (see text for details). Adapted from [65].

2.2. Linear Stepping Inchworm Motor

Even basic understanding of the mechanics of soft materials reveals that it is much easier to build an actuator that generates a pulling force compared to the one that would push using light-responsive LCEs. To extend the portfolio of devices beyond simple LCE strips (often, rather bombastically, pitched as “motors”), we designed and built a linear stepping inchworm motor with two LCE accordion-like actuators [52]. Linear displacement is often a desired mode of operation and various linear motors are in use, either powered from rotary drives or directly from pressure, electromagnetic forces or shape change in different materials. The first inchworm motors had a rotor moved by a sequential action of piezo actuator(s) [70,71]. In a linear inchworm motor, two or more actuators are operated sequentially to push/pull (rather than rotate) a shaft along its axis.

When fabricating the actuators for our motor, we tested a new LCE-orientation technique that we had conceived a long time before—rubbing overwriting. Mechanical rubbing is commonly used for orienting liquid crystal molecules [72,73]. Our first approach to fabricating LCE films with patterned alignment with rubbing through masks was used to make crawling caterpillars [50], but it required at least two masks and their precise alignment. Here we used a different approach: we rubbed the poly-vinyl alcohol (PVA)-coated glass surface in one direction, then covered certain areas with a mask, and rubbed everything in the perpendicular direction. Accordion-like 50-micron-thick LCE actuators fabricated with this procedure can contract by up to 80% upon heating [50] and were used in the miniature linear motor. A laser beam reflected from a mirror mounted on a galvo scanner illuminates (and thus heats up) two of these actuators, that, in turn, set a small, heavy gripper into an orbital motion (Figure 3B). Each scanning cycle consists of grip–move–release sequence (Figure 3C) and, as a result, the shaft moves (in either direction, defined by the laser scanning direction) at speeds of up to 25 mm/s (see [74] for the video).

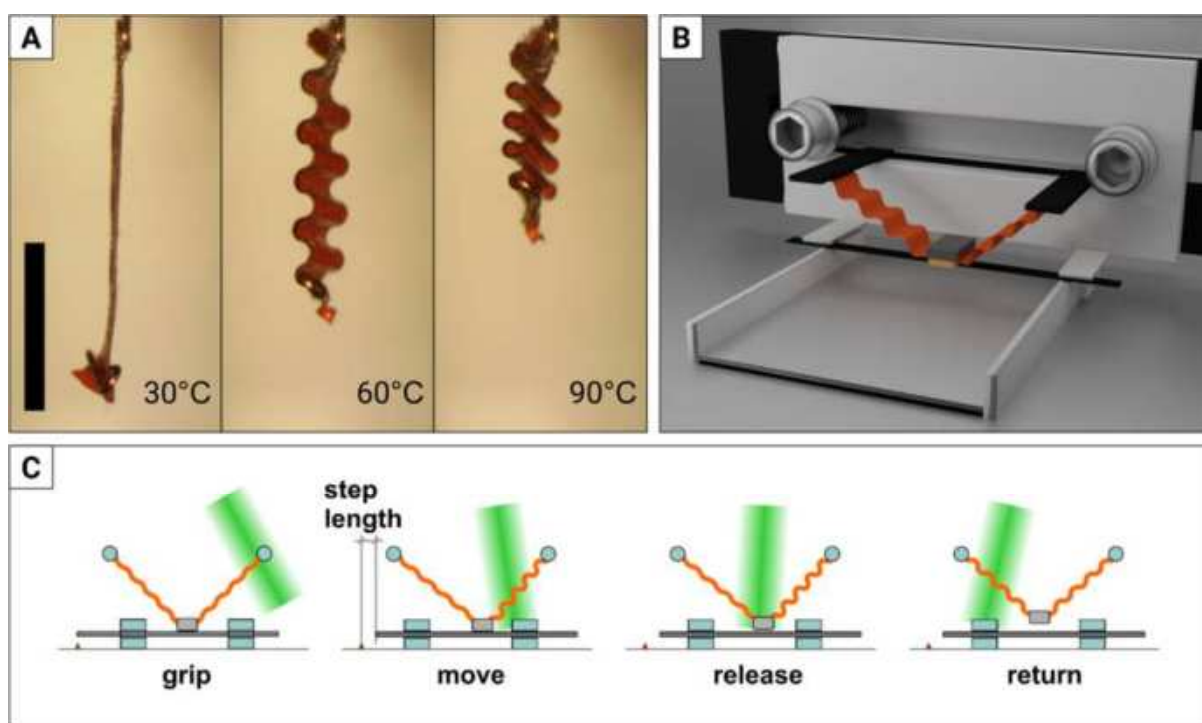


Figure 3. Light-powered linear inchworm motor. (A) Accordion-like actuator heated in an oven exhibits very large contraction, with up to 80% stroke. (B) CAD rendering of a linear stepping motor with two actuators (orange), gripper (dark grey/yellow) and a sliding shaft (black, under the gripper). (C) The sequential action of the actuators, powered by a scanned green laser beam, results in orbital motion of the gripper that, in turn, moves the shaft. The black scale bar in (A) is 5 mm long. Adapted from [52].

The motor design can be straightforwardly adapted to a rotary configuration by replacing the linear shaft with a circular one. If equipped with a position sensor, e.g., optical, the stepping motor can operate in a closed loop configuration, where the length of each step is not relevant, as the distance from the target position is continuously measured and adjusted, including with sub-single-step accuracy.

3. Bio-Inspired Millimeter-Scale Robots

For centuries, scientists and engineers have been fascinated by the movement of various animals at different scales and the possibility of copying this movement in man-made machines [75]. Nevertheless, after several decades and huge resources invested in

robotics, soft robotics and safe interactions of robots with humans in particular, the results are disappointing: most robots remain awkward automata, at best capable of performing basic repeatable tasks in well-controlled environments (perhaps the latest autonomous cars may soon change this rather gloomy picture). Robotics in the microscale is at an even worse stage—we are nowhere near scaling down robots to millimeters and below, not to mention their reliable fabrication and operation in real-life applications.

3.1. The Caterpillar

Conventional robots, made of multiple rigid parts connected by joints, typically have few degrees of freedom and poor adaptability to the environment. One attractive area of research is the realm of soft-bodied animals—segmented worms, mollusks, cephalopods and insects at some development stages (caterpillars)—where using elastic materials could allow their continuous movement to be mimicked, offering the bio-inspired robots the ability to move in confined spaces and to adjust to topologically complex environments [76]. To date, some attempts have been made to replicate them in real scale, with limited success [77].

Caterpillar locomotion consists of cycles of inching and crawling: the animal lifts and steps forward every pair of its legs—starting from the tail, towards the head. To detach the legs from the ground, it deforms parts of its body, generating a travelling wave of deformation along the body. Such deformations can be induced with spatially varying laser beam in the accordion-like actuators, such as those used in our stepping inchworm motor (compare Figure 3A). Our caterpillar robot [50] was fabricated with a 14.8×3.8 mm strip of 50 μm thick LCE film (Figure 4A). When illuminated locally with a scanned green laser beam steered by a galvo scanner (Figure 4B) the film deforms, becomes curved and lifts from the ground, thus generating a wave of deformation. When placed on a rough surface (e.g., sandpaper), this results in crawling locomotion, with a typical step length of 0.3 mm and a maximum speed of 30 mm/min (see Figure 4C for the snapshots and [78] for the video). This is about six times slower than the caterpillar *Cucullia verbasci*, commonly found in Europe and North Africa [79]. The robot is also able to squeeze through narrow slits, climb a sloped surface and push loads several times its own mass.

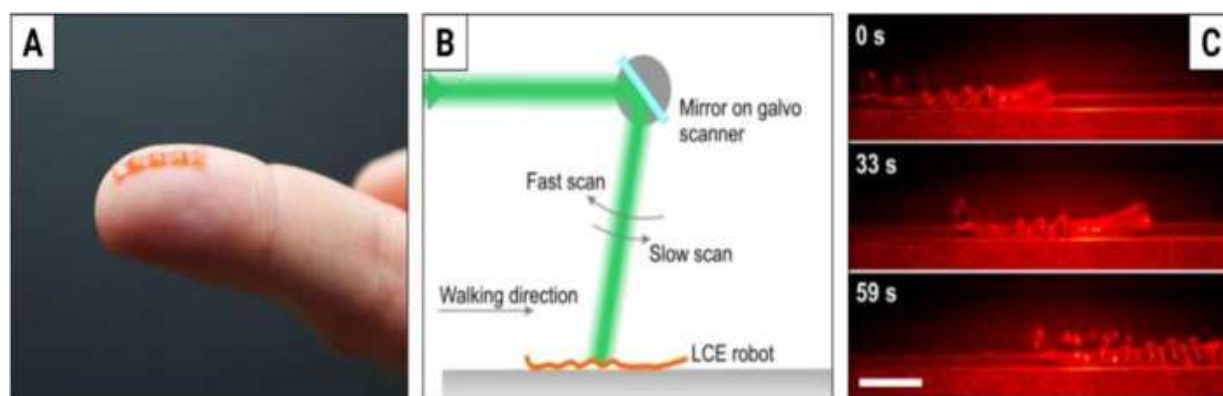


Figure 4. Natural scale crawling caterpillar robot. (A) The caterpillar robot on the fingertip of one of its creators. (B) Schematic of the experimental setup—the green laser beam is scanned along the robot’s body with a galvo mirror driven by an asymmetric sawtooth signal. The beam was scanned at 0.4 Hz and had 2.5 W of power. (C) Snapshots of the video with the light-driven caterpillar crawling on a level surface. The white scale is 5 mm long. The laser light is filtered out with an orange optical filter. Adapted from [50].

With a maximum speed of 30 mm/min, our caterpillar robot may be compared with a few similar demonstrations. In [80], a 29 mm-long robot made of LCE performs inchworm locomotion, powered through cables and reaching 1.91 mm/min. A composite tensegrity robot made of carbon nanotube-doped LCE was able to navigate a labyrinth with the

average speed of 6.87 cm/min [81]. Liquid crystal elastomer–carbon nanotube composite was also used in [82] in a crawling and jumping untethered robot, reaching 42 cm/min (half the body length) per minute.

3.2. The Snail

Gastropods—snails and slugs—have a single ventral foot, in which pedal waves propagate, propelling the animal (Figure 5A,B). To further increase the interaction between the foot and the surface, the former is covered with slippery mucus. The (apparently) low complexity of this design and its versatility made snail locomotion a promising target for implementations in robotics.

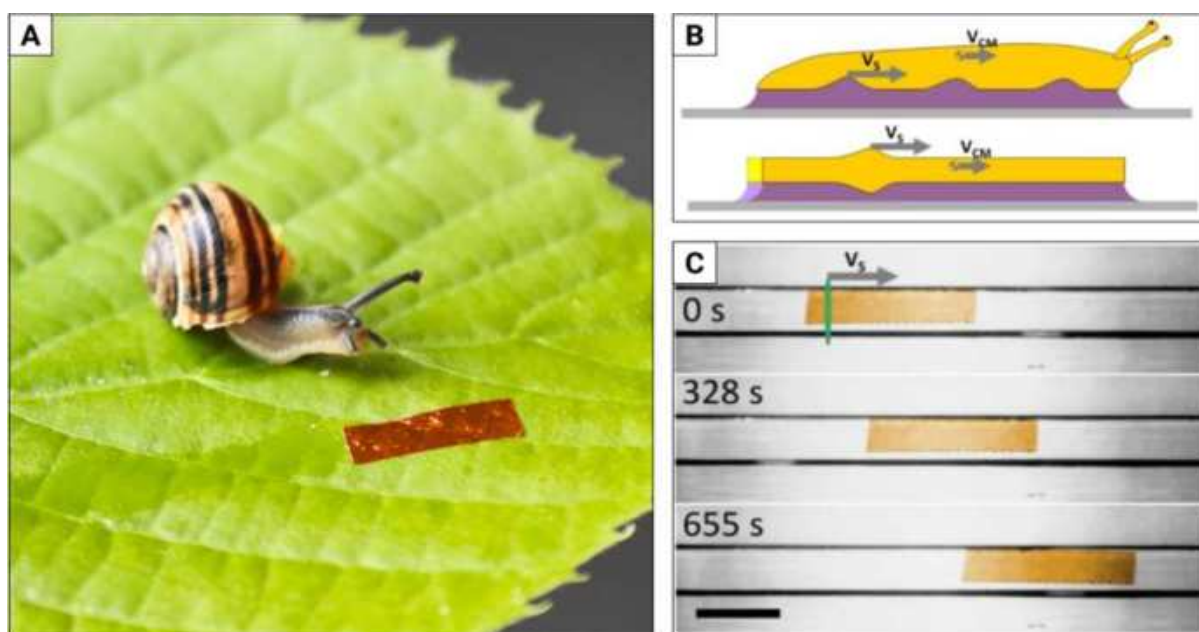


Figure 5. Light-powered snail mimicking the adhesive locomotion of terrestrial gastropods. (A) A garden banded snail *Cepea hortensis* meets the 10 mm long light-powered snail robot (having no shell, though). (B) In snails and slugs, pedal waves propagate along the ventral foot contact surface with a velocity V_s , propelling the animal with an average speed V_{CM} . In a similar way, the light-induced elastomer deformation moves along the robot's soft body (yellow) covered with an artificial mucus layer (purple). In both cases, the deformations are in fact much smaller. (C) Snapshots from a video with the snail robot crawling on a horizontal glass plate topped with glycerine as an artificial mucus. The average speed is 1 mm/min, the black scale bar is 5 mm long. The material contraction was of the order of 0.1 mm. Adapted from [5].

A 50 μm thick strip of LCE with planar nematic alignment film was placed on a glass substrate covered with a layer of glycerine (which proved to perform best as an artificial mucus) [5]. As with the caterpillar robot, local light-induced deformation was driven by a spatially scanned laser beam, but this time the elastomer contracted along its length, remaining in contact with the glass via the mucus layer. As the laser beam moved, the contraction propagated with a typical speed v_s on the order of several cm/s, resulting in an average robot speed V_{CM} of a few mm/s (see [83] for the video and Figure 5C for snapshots). The robot could move on various surfaces—in the same way as snails—from coarse sandpaper to PTFE (Teflon) [84,85], crawl over a glass tube obstacle, move horizontally upwards ($V_{CM} = 0.8$ mm/min), downwards ($V_{CM} = 1$ mm/min) and upside down ($V_{CM} = 3$ mm/min). Unlike snails that, interestingly, have no reverse gear [86], it could also move backward by reversing the direction of the laser beam scan. In this case, we are not aware of any similar demonstrations of robots performing the mucus-assisted locomotion in natural scale, even though it may be an interesting alternative for robots

operating in challenging environments, and it also provides good security margins due to large contact area with the surface.

3.3. The Water Strider

Not all our LCE-related projects have resulted in spectacular success. If they had, this would be a clear indication that the challenges were easy to overcome. In fact, a number of the projects have been abandoned, while others were put on hold, even for many years in some cases.

One example of the latter group is the light-powered, natural-scale water strider robot. There has been some substantial effort invested in understanding the dynamics of strider locomotion on the water surface, at some point leading to the so-called Denny paradox (baby striders seemingly move their legs “too slowly” to propel themselves, and yet they still manage to do so) [87], and later to its (apparent) resolution [88]. Striders offer a very good example of animal locomotion to be mimicked in the lab, as they do not need to struggle with friction, capillary or van der Waals forces while moving gracefully on the water’s surface, supported by surface tension (Figure 6A). Our first natural-scale (about 2.5 cm long) robo-strider was built (twisted) from 0.13 mm diameter copper wire, had the simplest muscle-actuator made of a bending strip and had to be guided by a thread, placed just above the water’s surface, to keep on track and remain within the green laser beam powering the LCE actuator (Figure 6B).

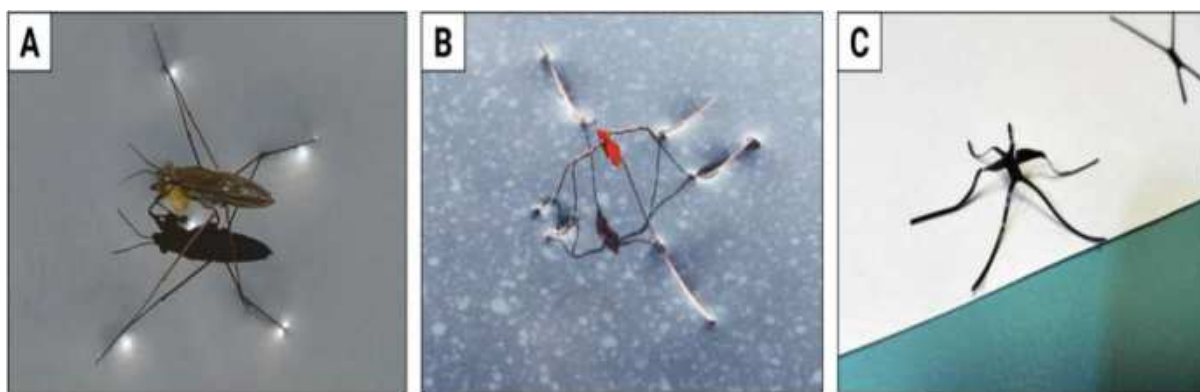


Figure 6. From the pond to the lab (and back)—a light-powered natural-scale water strider robot with LCE muscle. (A) Water striders are common inhabitants of reservoirs and rivers in many climate zones. (B) First prototype of the light-powered robo-strider with the orange LCE bending actuator powering a pair of legs. (C) The latest generation of the robot body (here without the actuator installed) was 2D printed with a 3D extrusion printer and then hot-shaped to take the final form. Water strider photo by Schnobby licensed under CC BY-SA 3.0.

Surprisingly enough, it did manage to move, with an average speed of around 180 mm/min—6000 times slower than some members of the Gerridae family. The project has been on hold for a number of years, but has recently re-emerged with a new approach to the strider body—this time with 3D printing (Figure 6C). We hope this will provide a much lighter frame, at the same time offering higher flexibility in the design and repeatability in manufacturing, should we decide to go for a swarm of light-driven water striders one day. As for increasing the speed, there is a class of so-called snap actuators [89], where a piece of material accumulates energy until it reaches a point of nearly instantaneous shape transition, releasing it to the mechanism to be driven (strider legs)—we hope this may be a way towards faster swimming speeds, perhaps one day comparable with wild-born animals.

Other approaches to making robo-striders included a jumping robot with nickel titanium shape memory alloy actuator [90] and a large scale (10 cm long) robot with as many as 12 legs and piezoelectric actuator, reaching the maximum speed of 180 cm/min [91].

As for now, we are not aware of any attempts to involve light-responsive materials in similar constructions.

3.4. The Ant

The second example of a project awaiting its turn is a millimeter-scale walking ant, powered and controlled by two colors of light, corresponding to the two degrees of freedom in the movement of its six legs. Two separate stimuli, applied in sequence, are the minimum for true non-reciprocal motion [4]. Insects (as opposed to arthropods, for instance) walk on six legs—this seems to be optimized for the number of points of contact with the ground, as they form two tripods that are lifted sequentially and shifted with respect to each other. To better understand this mode of locomotion, we have built several models (see Figure 7A for an example). The next step was to develop two LCE films, responding independently to two different colors of light (Figure 7B). Importantly, the illumination for the millimeter-scale walker must be provided by LEDs, not lasers, as the illuminated area must be large enough to let the walker make at least several steps, before walking into the dark [92]. Ultimately, the mechanical design must take into account many limitations of LCEs, e.g., illumination from one (or two) sides only and the available deformations of the actuators—one concept is presented in Figure 7C.

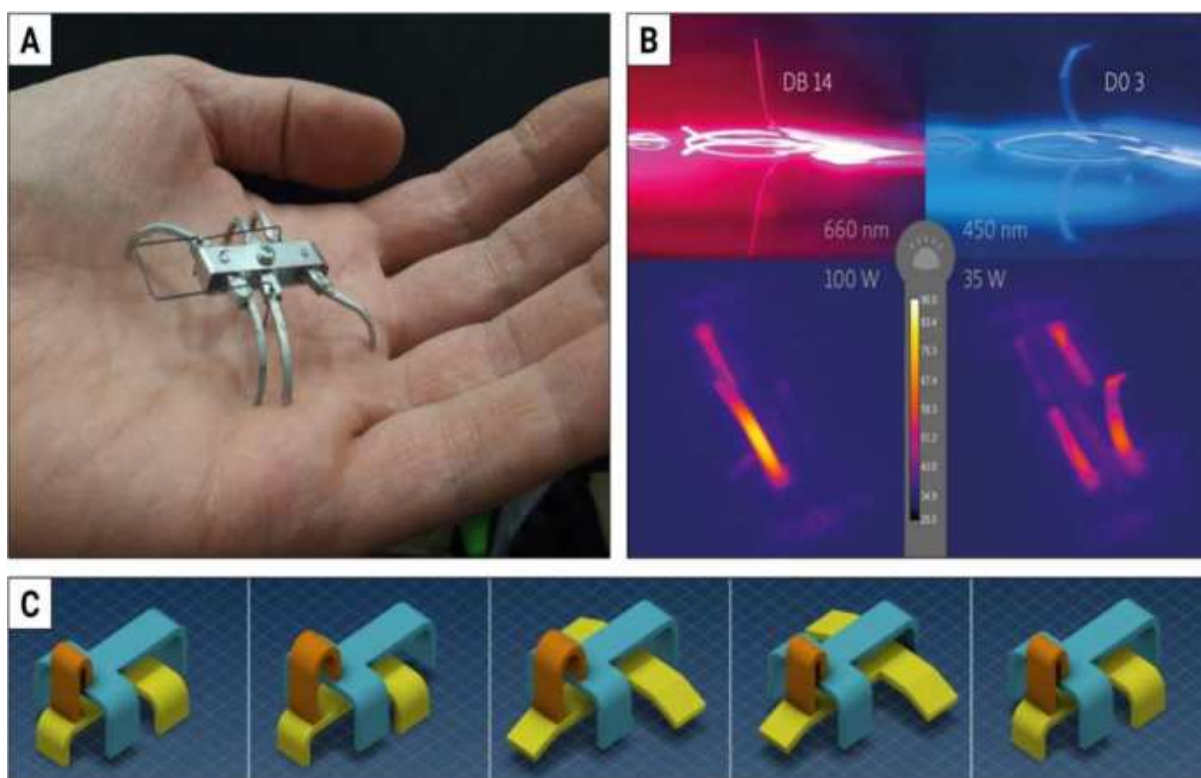


Figure 7. The ultimate walker: LCE ant with two degrees of freedom. (A) One of the models built to better understand six-legged locomotion. (B) Stripes of LCE with two different dyes, respond by bending to two high-power LEDs with 660 nm and 450 nm centered spectra, respectively. White light (top) and IR (bottom) images. (C) One of the concepts of the mechanical design of a six-legged walker with two tripods: one solid (blue) and one with lifting legs (yellow), shifted relative to each other with a bending strip (orange), performing a sequence of deformations that result in a step forward.

4. Micrometer-Scale Light-Power Tools

Perhaps our failures in mimicking nature, in particular when small scale and/or large quantities of mechanisms are in play, stem from the way we approach fabrication. In the lab, workshop or factory, we would start with materials (a plank of wood, a sheet of metal,

a piece of wire, a stretch of foil) and then cut, drill, mill or grind to make individual parts first, and then glue, weld, solder or rivet, to join them together. Obviously, this is not the way things are made in the natural world—there, they grow. This, then, is a question we asked ourselves: can we perhaps grow micro-mechanisms, instead of making them in the traditional way [93]?

To this end, we developed a method of fabricating micrometer-scale elastomer structures with a photo-mechanical response by sending UV light via an optical fiber immersed in an oriented liquid monomer, so that the polymerization occurs at the fiber tip—see Figure 8A–C for the snapshots of the fascinating growth process and [94] for the time-lapse video. In our experiments, a cone-shaped structure grows that bends when visible (green) light is delivered via the same fiber (Figure 8D and [94] for the video).

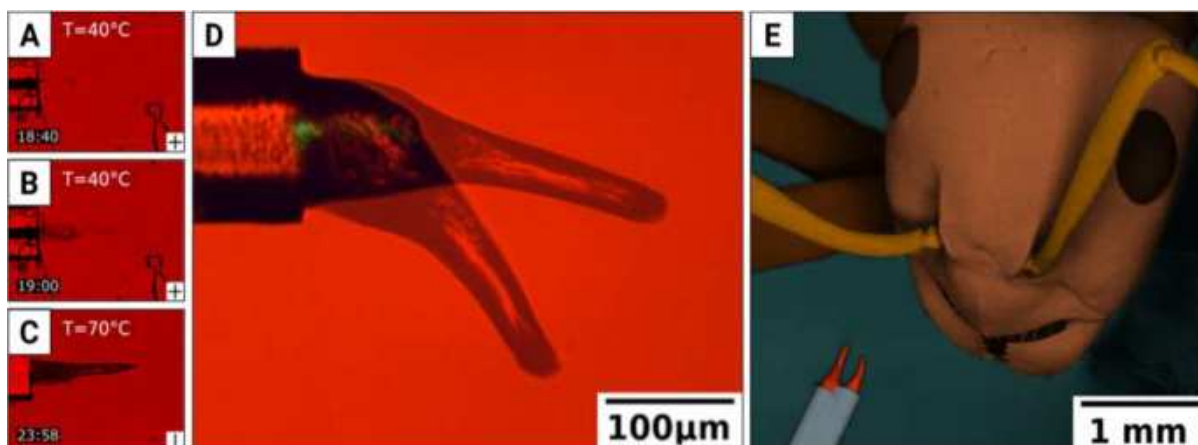


Figure 8. Fiber-grown microscale gripper. (A–C) Snapshots from the video show the bending actuator growth inside an LC-mixture-filled cell. Time in min: sec is shown in the bottom left corner of each frame. (D) Bending actuator in action, powered with green light delivered through the fiber (two photographs have been superimposed to show the magnitude of the deformation). (E) Two fibers with bending actuators make a light-powered gripper, here next to a *Formica polycytena* ant with its mandibles about ten times the size of our tool. Adapted from [93].

Gripping objects is fundamental for living organisms and in many machines. Mechanical grippers are typically powered by electric, pneumatic, hydraulic or piezoelectric servos and work well at larger scales, but their complexity and need for transmitting force from a distant servo to the gripper elements prevent their miniaturization and remote control. By joining two fibers with LCE-bending structures, we built a micrometer-scale gripper, powered and operated remotely with light energy delivered through the fibers (Figure 8E).

The gripper, nicknamed “optical pliers” (to avoid confusion with “optical tweezers” [95]), can deliver a gripping force of the order of 10^{-7} N that may be compared, for instance, to a single bending structure weight of approximately 3×10^{-9} N (see [94] for the video of the experiment where this force was determined). They do not require any displacement transmission—only energy is transmitted—which makes them very simple and potentially very reliable. In addition, they demonstrate that, with clever technologies, opto-mechanical micro-structures can be fabricated without resorting to any complex (and expensive) microfabrication technology, such as laser photolithography.

Will we one day be able to program and control the growth, so that we can make actuators, or even more complex tools and mechanisms, on fibers, at will? Where would the “genetic information” be stored? In the material, in the growth conditions (molecular orientation, temperature, flow) or in the light that initiates the polymerization—its wavelength(s), polarization, or spatial or temporal modulation? Perhaps in the structure of the optical fiber itself? The concept of “building by growth” involves many more questions than answers and opens up a new, fascinating avenue of research.

5. Conclusions and Outlook

Ten years of working and playing with soft, light-responsive materials have taught us many lessons. The most important one is perhaps the lesson of humility, when comparing our results in micromechanics and micro-robotics with the wonders of the natural world that we try to mimic.

At the moment we have several ongoing projects, including the following:

- Orienting LCE with the electric field during laser photolithography [96]. If successful, this technology will open up the ultimate realm of 5D photo-mechanical microstructures: the 3D-printed shape (with sub-micron resolution) with two angles of director orientation, programmable over the entire volume;
- Orienting LCE molecules with writing direction in laser photolithography. This method is somehow similar to orienting by squeezing through a small nozzle in 3D printing of LCEs [97], but on a much smaller scale. As it does not require any photo aligning layers or electrodes, it can be used with minute substrates, such as the end face of an optical fiber;
- Exploring the theme of “materials as machines” [98], we have developed a conveyor belt with LCE-sorting mechanisms, where small objects can be sorted into a number of buckets with LCE stripes (levers) that respond to their different colors and push them from the belt, without any sensors, data processing or separate actuators;
- Since our lab has its origins in photonics and optics, we are constantly playing with ideas of combining LCE structures with optical elements, e.g., optical fibers. We have tested several approaches to fiber switches, either with self-standing light-responsive actuators or with LCE micro-structures grown on the fiber tip.

Liquid crystal elastomers have been pitched as “promising materials” [99] that “open up new horizons in micro actuation and complex, remotely powered and controlled soft-robotics” [50]. Over the last decade, we have learned a lot about their many problems, not least related to repeatable fabrication, reliability and durability. If ever they may find their way into practical applications, the route will be a long one and not without hurdles and pitfalls. However, the journey into the world of light-responsive materials has given us a lot of joy and we hope that some of this is visible in our publications, including this one.

Author Contributions: M.R. was involved in most of the projects: he designed and made the caterpillar robot, with K.D. developed the snail robot, performed the earliest experiments with growing the (contracting) LCE micro-actuators on optical fibers, and worked on photo-orienting layers for RoboLEC with M.P. He is also involved, with Z.D., in all of the ongoing projects, where Z.D. focuses her efforts on LCE reorientation in laser photolithography; M.Z. led the optical pliers project, with contributions from K.D. and P.N.-J., who developed the early version of the tilting mirror mount with three contracting actuators grown on three optical fibers; P.G., accompanied by K.D., conducted many experiments designed to better understand the LCE response dynamics to various light stimuli. He also conceived the idea of robotic LCE film fabrication, which he later turned into RoboLEC with B.F.; P.W. was the group leader over the years and, to a greater or lesser extent, supervised all projects and made sure the lab worked smoothly from day to day. In the latter, he was supported by A.K., whose main project was the development of the two-color LCE actuators for the robotic ant. The manuscript was written by M.R., Z.D., K.D. and P.W. All authors have read and agreed to the published version of the manuscript.

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Publication III

A Millimeter-Scale Snail Robot Based on a Light-Powered Liquid Crystal Elastomer Continuous Actuator



A Millimeter-Scale Snail Robot Based on a Light-Powered Liquid Crystal Elastomer Continuous Actuator

Mikołaj Rogóż, Klaudia Dradrach, Chen Xuan, and Piotr Wasylczyk*

Crawling by means of the traveling deformation of a soft body is a widespread mode of locomotion in nature—animals across scales, from microscopic nematodes to earthworms to gastropods, use it to move around challenging terrestrial environments. Snails, in particular, use mucus—a slippery, aqueous secretion—to enhance the interaction between their ventral foot and the contact surface. In this study, a millimeter-scale soft crawling robot is demonstrated that uses a similar mechanism to move efficiently in a variety of configurations: on horizontal, vertical, as well as upside-down surfaces; on smooth and rough surfaces; and through obstacles comparable in size to its dimensions. The traveling deformation of the robot soft body is generated via a local light-induced phase transition in a liquid crystal elastomer and resembles the pedal waves of terrestrial gastropods. This work offers a new approach to micro-engineering with smart materials as well as a tool to better understand this mode of locomotion in nature.

Evolution has given rise to incredibly complex organisms capable of self-sustained locomotion, sensing and responding to the environment, down to the single-cell (micrometer) size. Robotics, after a considerably shorter time, is still struggling to approach the small (millimeter and smaller) scales, albeit with certain spectacular results: in the air,^[1] underwater,^[2] and on land.^[3] Soft robots, with deformable, continuous bodies harvesting energy from absorbed light, offer a promising path toward miniaturization, and indeed this path has already been followed successfully.^[4,5] Gastropods' adhesive locomotion has some unique properties: Using a thin layer of mucus,^[6,7] snails and slugs are able to navigate challenging environments, including glass, polytetrafluoroethylene (PTFE, Teflon), metal surfaces, sand, and (famously) razor blades, with only few super-hydrophobic coatings able to prevent them from crawling

up a vertical surface.^[8] The low complexity of a single continuous foot promises advantages in design and fabrication as well as resistance to adverse external conditions and wear, while constant contact with the surface provides a high margin of failure resistance (e.g., slip or detachment). The few attempts at robotic adhesive locomotion to date have been limited to externally powered, centimeter-scale demonstrators with mechanical drives.^[9]

Liquid crystal elastomers (LCEs)—solid, soft polymeric systems with well-defined molecular alignment of liquid crystals—can exhibit large, fast, and reversible shape changes upon external stimuli, in particular light absorption.^[10]

Due to the tight packing of rod-shaped molecules, arranged in cross-linked polymer chains, the order (anisotropic)–disorder (isotropic) transition reduces the effective chain length along the alignment direction (the director), at the same time increasing the spacing between the molecules in the perpendicular directions. As a result, the material undergoes macroscopic deformation, determined by the orientation of the director and the spatiotemporal distribution of the stimulus (Figure 1a). The mechanism can involve photochemical reactions,^[11,12] photothermal heating,^[13] or a combination of both. LCEs offer complex modes of actuation through molecular alignment engineering.^[14] Additional degrees of freedom are available, as the light-induced deformation can be proportional not only to the incident light intensity^[10] but also to its spectrum^[15,16] or polarization.^[17,18] Both “pure LCE” actuators^[19,20] as well as LCE-based composites^[21–23] have been used as light-powered actuators in various configurations (cf. ref. [24] for a recent review on LCE soft robots and ref. [25] on LCE actuators). In particular, periodic deformations of a cylindrical body (0.1 mm diameter, 1 mm length) generated by structured light illumination have been used on the micro-scale to propel an underwater swimmer, mimicking the metachrony of ciliate protozoa, with speed on the order of micrometers per second.^[26]

Combining the soft elastomer deformation induced by a spatially and temporally modulated laser beam with the adhesive properties of the synthetic mucus, we demonstrate a light-driven, millimeter-scale crawler that mimics terrestrial gastropods' mode of locomotion. It uses the photo-thermally induced response of a continuous liquid crystal elastomer actuator, similar to the caterpillar robot we presented before,^[4] but this time in a different crawling gait. The robot body contracts along its long axis while remaining flat and always in contact (via the mucus layer) with the surface over its entire area. By

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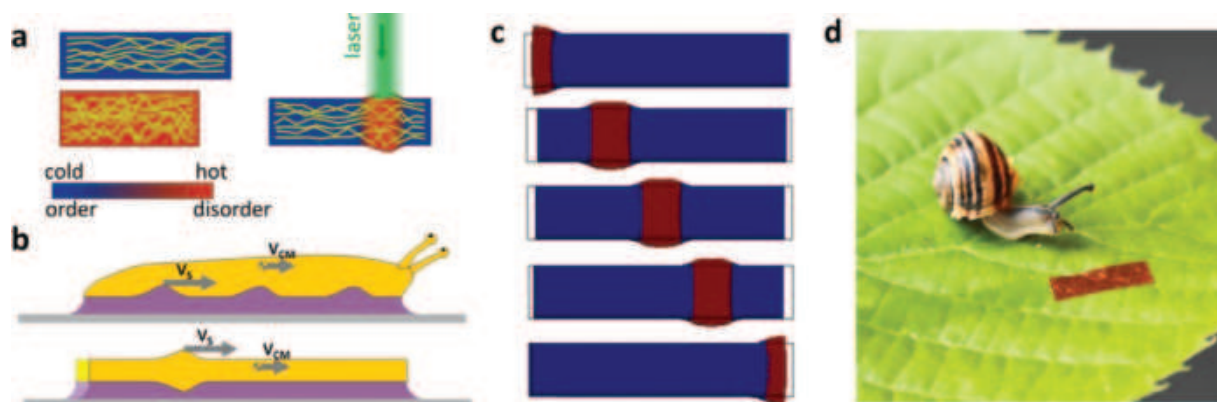


Figure 1. Light-induced traveling deformation in a liquid crystal elastomer generates pedal waves in a millimeter-scale soft robot. a) Cross-linked polymer chains (olive) are arranged in one direction in a strip of liquid crystal elastomer. When (and where) the material is heated up (e.g., by light absorption), the molecular order decreases, the polymer chains effectively shorten along the director, and macroscopic deformation results. b) In terrestrial gastropods, adhesive locomotion is driven by pedal waves propagating along the ventral foot's contact surface at velocity V_s , propelling the animal with the average speed V_{CM} . In a similar way, the light-induced elastomer deformation moves along the robot soft body covered with an artificial mucus layer. Deformations are not drawn to scale. c) Finite element numerical simulation of the photo-thermally induced deformation propagating along the robot body during one scanning cycle. d) Our light-powered snail robot next to a garden banded snail (*Cepaea hortensis*).

means of these small-scale (on the order of 0.1 mm) traveling body contractions, the soft robot can navigate various surfaces, up to the extremes of crawling upside down on a glass ceiling.

By scanning the laser beam along the robot body, a periodic time- and position-dependent heat distribution is created that results in the traveling deformation of the material. When set on a thin layer of artificial mucus (glycerin), the robot performs crawling locomotion with average velocity on the order of a few millimeters per minute (Figure 2a; see also Movie S2, Supporting Information). As the laser beam is scanned from the rear toward the front, the robot body contracts as a result of the local temperature increase due to light absorption (see inset in

Figure 2a and Movie 3, Supporting Information), and the contracted volume follows the laser illuminated area at speed V_s , typically on the order of a few centimeters per second. The interaction with the synthetic mucus layer, and via this layer with the substrate, results in thrust generation and net forward motion.

During one scanning cycle, the trailing edge (the tail) of the robot is stationary for most of the time, except within a short period when it moves forward quickly, at around 0.3 mm s^{-1} (Figure 2b). At the same time, the leading edge (the head) moves forward at around 0.02 mm s^{-1} , and also makes a large backward-forward stroke with zero net displacement for approximately one-third of the cycle. Each laser scanning cycle generates

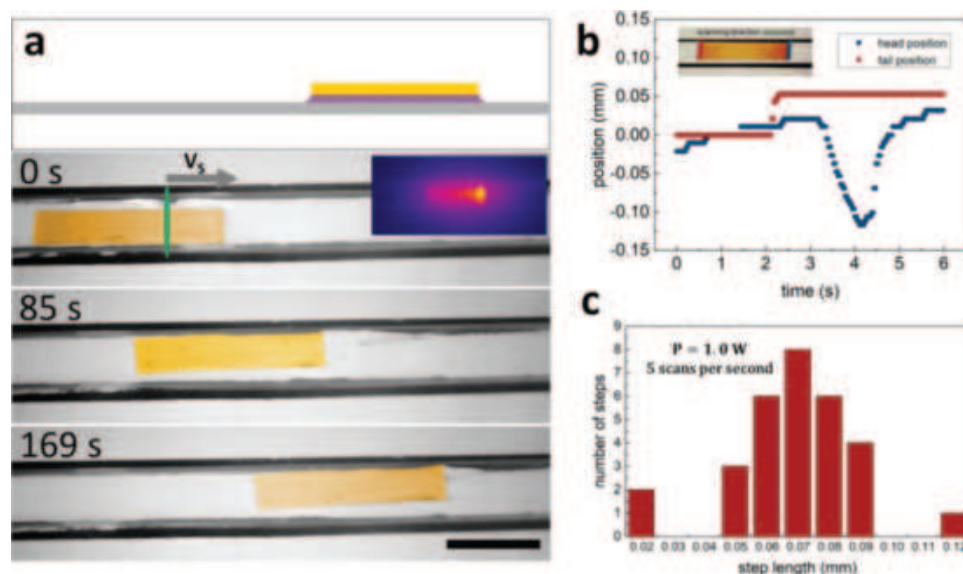


Figure 2. Adhesive locomotion on a flat surface. a) The soft robot crawling on a horizontal glass surface with glycerin mucus at an average speed $V_{CM} = 0.4 \text{ cm min}^{-1}$. The laser beam, scanning at speed V_s , is shown schematically in green. The color inset shows the measured temperature distribution in the robot. The scale bar is 5 mm long; the colors are artificial. b) Measured position of the robot head (blue dots) and tail (red dots) during one scanning cycle. c) Histogram of 30 measured step lengths during adhesive locomotion in the configuration from (a).

one forward step of around 0.1 mm net length, depending on the laser power and the scanning speed. Random processes involved in the crawling dynamics result in a measured step length histogram with normal distribution, as shown in Figure 2c.

Measurements of the robot's average velocity V_{CM} under various conditions give insight into the light-powered crawling mechanics based on photomechanical response in the LCE. In the steady state, with a continuous (CW) laser beam, under thermal equilibrium, the LCE actuator deformation is proportional to the laser power illuminating the material. With the scanned laser beam, in the regime where the polymer does not reach thermal (and mechanical) equilibrium in the illuminated area, the light-induced material deformation is proportional to the ratio of the incident laser power and the beam scanning speed P/V_S . At the same time, 2D analysis of a thin Newtonian liquid layer^[27] indicates that for sinusoidal pedal waves of

a given amplitude, the center of mass velocity V_{CM} is a linear function of the wave speed (i.e., the laser scanning speed) V_S . Assuming a linear relation between the pedal wave amplitude and P/V_S , our experimental data can be fitted with the resulting formula

$$\frac{V_{CM}}{V_S} = \frac{3 \left(D \frac{P}{V_S} \right)^2}{1 + 2 \left(D \frac{P}{V_S} \right)^2} \quad (1)$$

where D is a constant proportional to the polymer actuator response to the absorbed light energy and the average thickness of the mucus layer^[9] (Figure 3a). For low laser powers this relation fails; if little light energy is available within a given material

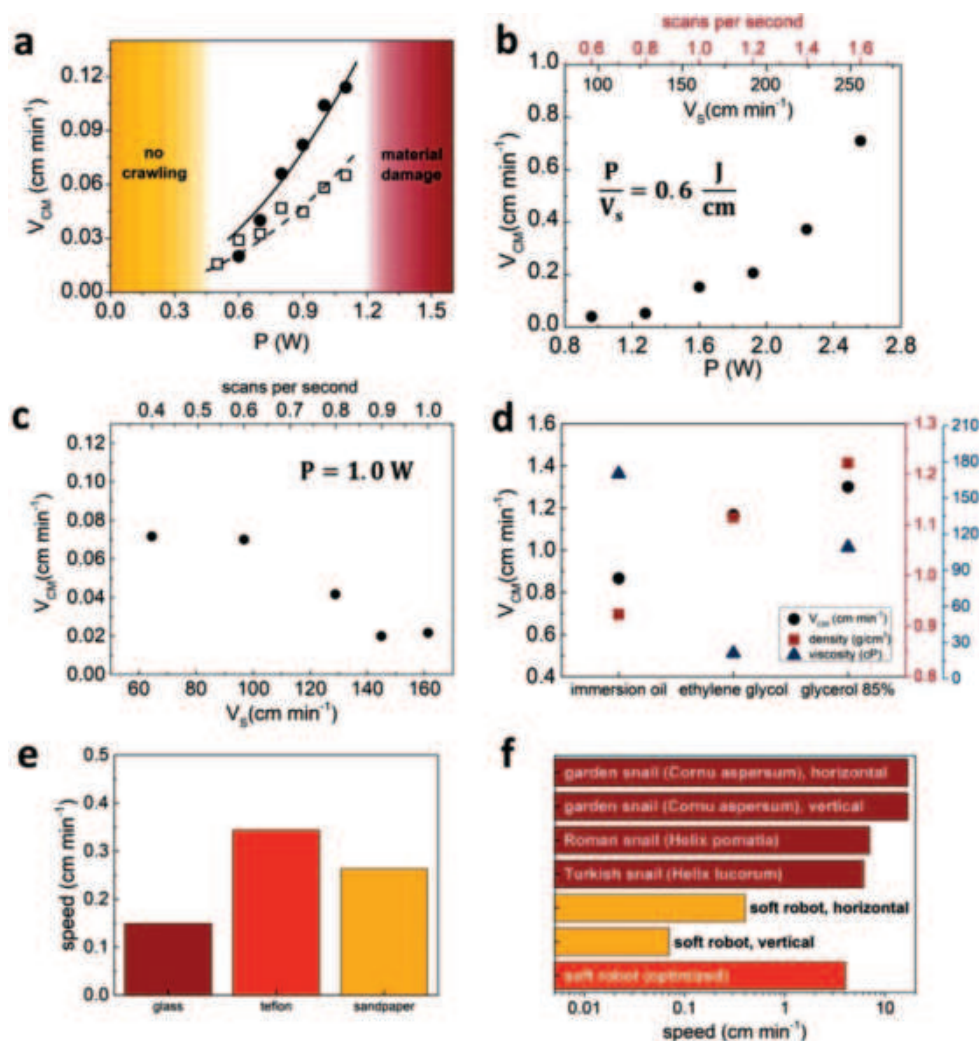


Figure 3. Mucus-assisted crawling performance. a) Measured average robot speed V_{CM} for different laser powers at constant pedal wave speed $V_S = 25$ cm min⁻¹ (filled circles, solid line) and $V_S = 12.5$ cm min⁻¹ (empty squares, dashed line). The fitted curves are described by Equation (1), assuming body deformation proportional to the ratio of the laser power (P) and the scanning speed (V_S). b) Average robot speed V_{CM} measured for different laser powers (P) with constant P/V_S . c) Measured average robot speed V_{CM} for laser power $P = 1$ W and various pedal wave velocities V_S and stepping frequencies. d) Average robot speed V_{CM} measured for different liquids used as mucus. e) Measured average robot speed on different horizontal substrates. f) Average crawling speed for several types of snail and the soft robot on horizontal and vertical surfaces. Optimized speed assumes 100% illumination duty cycle and five simultaneous pedal waves, together potentially resulting in a tenfold speed increase.

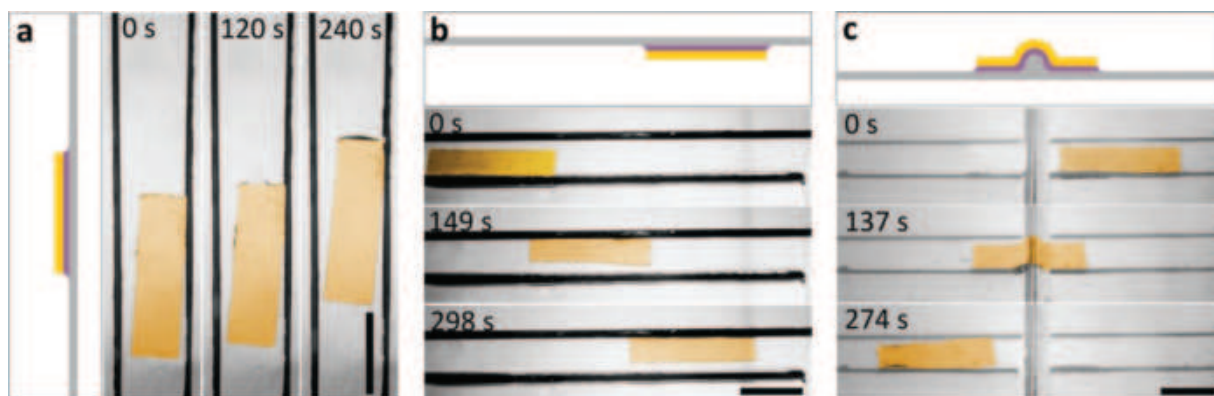


Figure 4. Adhesive locomotion in various configurations. a) Climbing up a vertical glass surface (average speed $V_{CM} = 0.08 \text{ cm min}^{-1}$). For climbing down in the same configuration (not shown), the measured speed was $V_{CM} = 0.1 \text{ cm min}^{-1}$. b) Crawling on an upside-down horizontal glass surface ($V_{CM} = 0.3 \text{ cm min}^{-1}$). c) Crossing an obstacle (0.8 mm diameter glass tube) on a horizontal glass surface ($V_{CM} = 0.3 \text{ cm min}^{-1}$). The scale bar is 5 mm long, the colors are artificial.

volume over a given period of time, due to heat exchange with the environment, the temperature increase is too small to affect the elastomer shape. Only for sufficiently fast light energy deposition can the polymer heat up, before it equilibrates (mainly through thermal conductivity) with the environment. Increasing the laser power while maintaining a constant P/V_S ratio (Figure 3b) shows that the highest average speed is achieved for high light intensity; given the same magnitude of the thermo-mechanical response, it is the scanning speed V_S and the number of scans (i.e., steps) per second that determine the average crawling speed. Although according to formula (1), for a constant P/V_S , V_{CM} should be a linear function of V_S , the average speed nevertheless increases much faster for the highest scanning speeds. This also suggests that at high scanning speeds with high laser power, the photo-mechanical response of the polymer is governed by a much lower time constant than heat exchange with the mucus and the substrate, and the approximation of a constant LCE actuator response is no longer valid; the response increases with increasing laser power, even for constant P/V_S . For even higher laser powers (and higher V_S), random deformations are observed, without a net displacement of the robot over time.

Varying the pedal wave speed (V_S) by changing the laser beam scanning frequency (Figure 3c) reveals an intrinsic tradeoff in light-driven adhesive locomotion as we have implemented it. While for low scanning frequencies (small V_S), the temperature increase and the material deformation due to light absorption in the material are high and result in longer individual steps, these steps are taken less often. As the scanning speed increases, up to a certain point, the decrease in step length is compensated by their increased frequency. Further decrease in the polymer deformation results in a significant (nearly fourfold) reduction of the average crawling speed. Figure 3d presents measured average robot velocities for three different liquids with different densities and viscosities used as mucus—glycerin, ethylene glycol, and microscope immersion oil—on a glass substrate. A control experiment was also performed with the robot set flat on a dry glass substrate; in this case, no crawling is observed, the LCE actuator locally deforms (bends) and detaches from the substrate, and for higher light intensities, it breaks and/or burns. We also verified that

adhesive locomotion can be performed with similar efficiency on a number of substrates, including glass, PTFE (Teflon), and coarse sandpaper (Figure 3e). Our robot's performance falls short of that of its natural relatives; even accounting for the lower duty cycle and only one pedal wave being present at any given time, the maximum velocity on a horizontal surface is around one order of magnitude lower (Figure 3f). To verify the robot's performance in various conditions, we set it up to walk up a vertical glass surface (Figure 4a and Movie S4, Supporting Information), on a glass "ceiling" (Figure 4b and Movie S5, Supporting Information) and over an obstacle made of a glass tube perpendicular to the crawling direction (Figure 4c and Movie S6, Supporting Information). In all experiments, a thin layer of glycerin was deposited on the substrate, and the measured average walking speeds are on the order of millimeters per minute. The adhesion forces between the robot body, the artificial mucus, and the substrate (glass) are strong enough to support the robot mass on both the horizontal surface and in the upside-down configuration, and at the same time allow for the light-induced body deformation that can propel it forward.

Interestingly, it seems that gastropods cannot reverse the direction of their pedal waves to move backward, and instead have to rely on sharp U-turns.^[28] In our robot, when the laser scanning direction is reversed (see Movies S2 and S4–S6, Supporting Information), the crawling direction is also reversed.

Our results indicate that mucus-assisted, light-powered crawling without body lift-off is a feasible mode of locomotion for small-scale soft elastomer robots. As the major part of the robot surface in contact with the substrate remains flat during adhesive crawling, the propulsion mechanism likely relies on (nonlinear) fluid dynamics in the mucus layer.^[7] At the same time, there is no fundamental difference in the propulsion efficiency between different liquids used as the artificial mucus, which suggests that its exact rheological properties are of secondary importance as long as it provides continuous adhesion and lubrication of the body–substrate area. Light-induced traveling deformation in LCEs provides an effective means to design soft robots with complex body geometries, enabling various modes of locomotion on millimeter scales and below. Despite the slow speeds, need for constant lubrication, and low energy

efficiency,^[29] the presented elastomer soft robot offers unique insights into micromechanics with smart materials and may also provide a platform for studying adhesive locomotion.

Experimental Section

During gastropod locomotion, muscle contractions known as pedal waves propagate along the central part of the animal foot at speed V_s , greater than the speed of the animal's center of mass V_{CM} (Figure 1b). To reproduce such pedal waves in a millimeter-scale soft robot, a reversible, light-induced phase transition in a 50 μm thick 3×10 mm strip of soft liquid crystalline polymer was used. The cross-linked LCE mixture contained 78 wt% of the LC monomer (Synthon), 20 wt% of the LC crosslinker (Synthon), 1 wt% of the photoinitiator (Irgacure 369, Sigma-Aldrich), and 1 wt% of the dye (Disperse Red 1, Sigma-Aldrich).^[4] All chemicals were used as received. To fabricate the LCE film, two microscope cover slides were spin-coated with polyvinyl alcohol and dried on a hot plate. After mechanical rubbing with a cloth, the slides were assembled into a cell with 50 μm diameter spacers. Molten LC mixture was infiltrated into the cell at 80 °C and the temperature was decreased to 40 °C. A 380 nm peak wavelength UV LED was used to polymerize the material for 60 min after it was supercooled in the freezer to −5 °C. The LCE film exhibited a reversible contraction in the direction parallel to the director whenever and wherever the nematic order of the polymer network was decreased by an increase in the temperature (Figure 1a).

A CW green 532 nm laser (Verdi V-5, Coherent) was used to power and control the robot. The laser beam was shaped with a cylindrical lens into an ellipse with 3 mm and 0.03 ± 0.02 mm full widths at half maximum and steered with a mirror mounted on a galvo scanner, driven with a sawtooth signal. The slow forward motion (99% of the cycle duration) generated the propagating deformation of the robot body while the fast backwards scan brought the beam back to the initial position with no visible actuation. To measure the polymer strip deformation and displacement, a high-resolution camera was used at 60 frames per second. The temperature distribution was measured with the Fluke Ti400 thermal imaging camera. To account for the sub-optimum duty cycle of the robot, it was assumed that laser light was absorbed by the elastomer during one half of each scan and that up to five pedal waves could be generated along the robot body, potentially resulting in a tenfold increase in speed.

The time-dependent response of the LCE actuator was simulated using a finite element model in COMSOL Multiphysics. The simulation results demonstrated how the local increase in temperature decreased the molecular order in the elastomer and thus generated the traveling deformation (Figure 1c and Movie S1, Supporting Information). The deformation dynamics is modeled by introducing a local photo-strain tensor $\epsilon_{ph}(\mathbf{x}) = \epsilon_{ph}[(1 + \nu)\mathbf{n} \times \mathbf{n} - \nu\mathbf{I}]$ into the stress (σ)–strain (ϵ) constitutive relation $\sigma(\mathbf{x}) = \mathbf{C}[\epsilon(\mathbf{x}) - \epsilon_{ph}(\mathbf{x})]$, where $\mathbf{C}$ is the stiffness tensor of the elastomer strip, ϵ_{ph} is the photo-thermally induced strain along the director $\mathbf{n}$, and the Poisson ratio of the strip $\nu = 0.5$.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

bioinspired robotics, liquid crystal elastomers, photoactuation, soft robots

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Publication IV

Traveling Wave Rotary Micromotor Based on a Photomechanical Response in Liquid Crystal Polymer Networks

Traveling Wave Rotary Micromotor Based on a Photomechanical Response in Liquid Crystal Polymer Networks

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Supporting Information

ABSTRACT: The photomechanical response of liquid crystal polymer networks (LCNs) can be used to directly convert light energy into different forms of mechanical energy. In this study, we demonstrate how a traveling deformation, induced in a liquid crystal polymer ring by a spatially modulated laser beam, can be used to drive the ring (the rotor) to rotate around a stationary element (the stator), thus forming a light-powered micromotor. The photomechanical response of the polymer film is modeled numerically, different LCN molecular configurations are studied, and the performance of a 5.5 mm diameter motor is characterized.



KEYWORDS: liquid crystal polymer networks, photomechanic actuation, light-powered mechanics, micromotor, traveling wave motor

INTRODUCTION

“Can light drive a motor?” was the opening question asked in the seminal paper of Ikeda et al.¹ In most power systems, light (solar) energy is converted either to directly useful heat (in solar thermal collectors) or to electric energy (in solar cells) to be used or stored. Converting light energy directly into mechanical energy is more challenging, although several approaches have been demonstrated at different scales, from the molecular to macroscopic.^{2–4} An attractive aspect here is the possibility of remotely powering mechanical devices with light (delivered either via free space propagation or through optical fibers), where using electrical conductors and/or currents is not feasible.

In 1975, De Gennes proposed the idea of anisotropic cross-linked polymer networks with the orientational order of nematicogenic monomers,⁵ thus initiating research efforts to fabricate, understand, and use ordered, responsive polymers, in particular liquid crystal networks (LCNs). LCNs are solid polymers with a well-defined alignment of molecules (i.e., with a well-defined director of the molecular blocks), similar to that observed in liquid crystals, with a moderate to densely cross-linked network architecture. With partially anisotropic properties of the LC phase, LCNs have unique anisotropic properties, including the elastic ones. They may exhibit very large, reversible deformations in response to the external stimuli: heating,⁶ light triggering photochemical reactions,^{7,8} light inducing photothermal heating,^{6,9,10} external electric field,^{11,12} or the presence of solvent.¹³ Cross-linked chains of molecules undergo the order–disorder (anisotropic–isotropic) transition and, as a result, the material deforms in a

way determined by the spatial distribution of the director and the spatio-temporal distribution of the stimulus, for example, light. Additional mechanical actuation modes may be available with light spectral^{14,15} or polarization^{16,17} degrees of freedom. Light-powered LCE elements^{18,19} have been used as micro-actuators in various configurations^{20,21} (cf.²² for a recent review on light-driven LC materials,²³ on LCN and LC elastomers (LCE) soft robots and²⁴ on LCN and LCE actuators).

A light-driven motor utilizing the contraction (expansion) of an azobenzene-containing LCE laminated with a polyethylene film upon UV (visible) light irradiation was reported by Yamada et al.¹ The motor presented by Geng et al.²⁵ used a looped stripe of a hydroxypropylcellulose film that deformed under humid air to drive a rotating element. The LCE rod, described in ref 26 can roll with either a light beam or a heated surface as the energy source. Recently, different modes of deformations in LCE tubes and helical ribbons have been demonstrated as drives for centimeter-scale light-powered rolling vehicles.²⁷

In this paper, we present a rotary micromotor, inspired by the ring-shape piezoelectric ultrasonic motors (USMs), often used in the auto-focusing mechanism of compound lenses, for example, in SLR cameras. USMs of this type have a

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piezoelectric ring actuator in which a traveling mechanical flexural wave is electrically generated and it drives a rotor via friction coupling^{28–30} (Figure 1). Our micromotor has a ring-

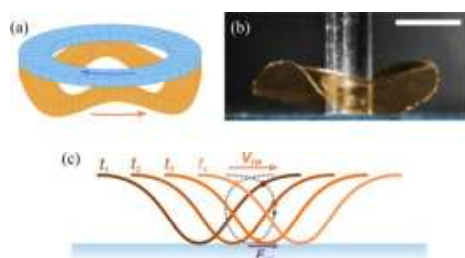


Figure 1. (a) Dynamic deformation induced in the stator (orange) interacts via friction forces with the rotor ring (blue). (b) Traveling deformation of the liquid crystalline polymer ring (orange), induced by a spatially modulated laser beam, interacts with the stationary substrate (blue), and thus, the ring rotates around the vertical axis. (c) Photothermally induced deformation traveling along the rotor circumference (time $t_1 < t_2 < t_3 < t_4$, V_{TW} —velocity of the traveling wave) results in each point on the surface following a regular orbit (grey dashed line). When the rotor comes in contact with a stationary substrate, the static friction force F_{SF} acts on the rotor and sets it in motion. The white scale bar is 2 mm long. Panel (a) is adapted from.³²

shaped rotor made of an LCN film with a radial or azimuthal director orientation of the director at each surface. An all-optical method employing photo-oriented azopolyimide layers for generating arbitrary two-dimensional director distribution in the LCN film was used for the first time. The principle of a photo-induced traveling deformation in an LCN element, combined with friction coupling that earlier allowed us to demonstrate a life-size crawling caterpillar soft robot,³¹ now also proved to be well suited for the rotary micromotor design. In the USM-inspired micromotor, the traveling deformation is generated in the ring rotor by the photo-mechanical response due to light absorbed from a spatially modulated (rotated) laser beam.

EXPERIMENTAL SECTION

Liquid Crystal Polymer Network Film. The liquid crystal cross-linked polymer mixture is based on commercially available compounds and contains 78 wt % of the LC monomer (Synthon), 20 wt % of the LC cross-linker (Synthon), 1 wt % of the photoinitiator (Irgacure 369, Sigma-Aldrich), and 1 wt % of the dye (Disperse Red 1, Sigma-Aldrich), as reported in ref 31. The orange dye provides efficient green laser light absorption. All the compounds were melted and stirred on a hot plate at 80 °C.

To prepare LCN films with a proper molecular orientation, a photopatterning technology was used. Glass slides were spin-coated with a newly synthesized photoalignment azopolyimide (PI) solution (~1%) in *N*-methyl-2-pyrrolidone, dried, and heated at 130 °C for 2.5 h to evaporate the solvent. Chemical structure and characterization of the PI are presented in the Supporting Information. To create radial or azimuthal distribution of the director in the LCN film, the polyimide orienting layers were exposed to linearly polarize UV light with wavelengths in the 345–380 nm band (Hamamatsu LC8 Xe-Hg UV lamp with a Glan-Taylor prism polarizer) with 15 mW/cm² intensity at the glass plane for 1 h. A wedge-shaped aperture was used in the UV beam, and the substrate was rotated continuously during the exposure. Two substrates were assembled to form a 25 μm cell with the azimuthal–azimuthal (A–A) or azimuthal–radial orientation (A–R, also known as circular twisted nematic,³³ Figure 2). The cell was filled with the molten LCN mixture on a hot plate at 80 °C and

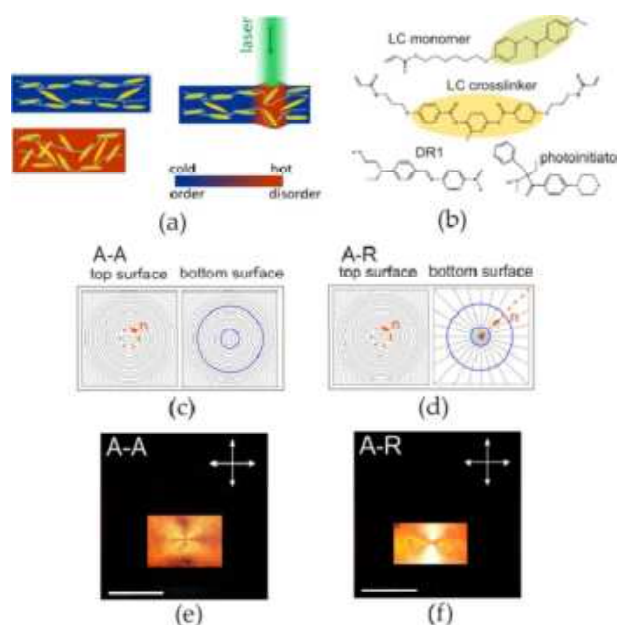


Figure 2. (a) Cross-linked polymer chains (yellow) arranged in one direction in an LC polymer network stripe deform locally because of increased temperature. With efficient light-absorbing dye, the heating can be induced by light absorption from a laser beam. (b) LC monomer, cross-linker, dye (Disperse Red 1) and photoinitiator used in the LCN. The coloring is consistent with panel (a). (c,d) Orientation of the molecular director n at the top and bottom surfaces of the cells used for preparing the LCN films: azimuthal (A) and radial (R). Two types of LCN films were prepared: A–A and A–R. Blue rings present how the micromotor rotor was cut from the film. (e,f) A–A and A–R cells filled with the LCN mixture, observed with a polarizing optical microscope (the arrows indicate the polarizer and analyzer orientation). The white scale bars in the photos are 10 mm long.

cooled slowly (2 °C/min) to 40 °C. The LCN layer, with the director distribution defined by the LCE interaction with the orienting layers, was then polymerized with 380 nm UV LEDs for 20 min at 40 °C. After being dipped in water at room temperature, the cell was wedged open, the LCN film was removed and the rotor ring (5.5 mm outer diameter, 1.3 mm inner diameter) was cut with a razor blade.

Differential scanning calorimetry (DSC) of the LCN film was used to determine the glass transition temperature (T_g) and the LC phase transitions (Figure S2 in the Supporting Information). The material has T_g of 11 and 7 °C upon heating and cooling, respectively, and the isotropization temperatures of 120 and 95 °C upon heating and cooling, respectively. The dynamic mechanical thermal analysis (DMTA) reveals the α relaxation corresponding to the glass–rubber transition (Figure S3 in the Supporting Information). The loss modulus peak and the inflection point of the storage modulus drop were observed at 17 °C, close to the values obtained from DSC.

Experimental Setup. A CW 532 nm laser (Verdi V-5, Coherent, 5.0 W maximum power, Gaussian beam full width at half maximum equal 1.9 mm at the LCN ring plane) was used to power the micromotor. The beam was steered with a rotating mirror to follow a 1.7 mm radius circular path (Figure 3a). The LCN was in contact with a sandpaper substrate (2000 grit) and could rotate freely around a stationary axis (a glass tube or steel rod). In our earlier works on light-driven microrobots based on photomechanically induced traveling wave,³¹ we tested several substrates, including glass, paper, and different sandpapers, with different tribological properties. For the current experiments, we have chosen the substrate that proved to perform best in terms of the friction coupling and thus the motor performance. The static friction coefficient between the rotor and the

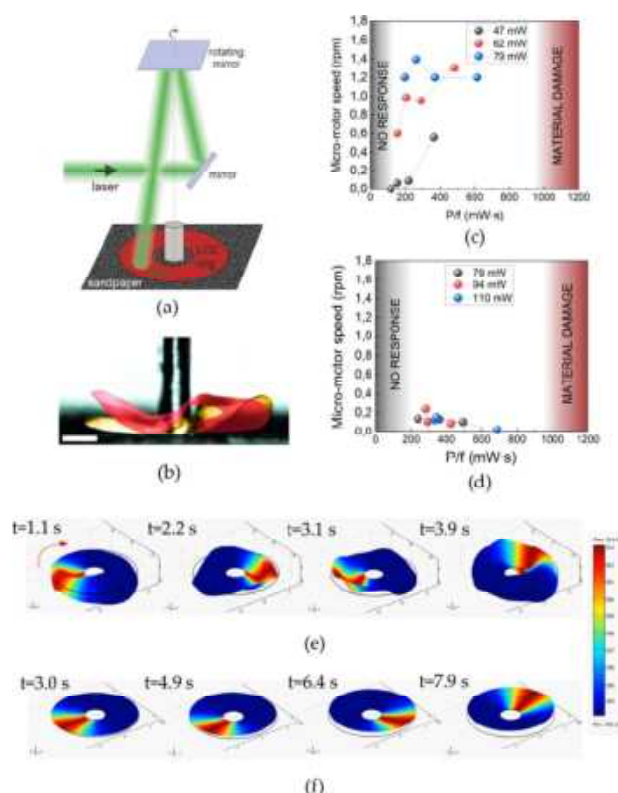


Figure 3. (a) Schematic of the experimental setup. The laser beam is reflected from a rotating mirror and sweeps around the LCN rotor ring. (b) Static LCN ring deformation due to the photomechanical response: yellow and red (computer-added colors) areas are images of the ring without and with the laser on, respectively. The white scale bar in the photo is 1 mm long. (c) LCN micromotor speed varies with the increasing P/f ratio, where P is the laser power and f is the mirror rotation frequency. Rotor with A–R orientation. (d) Same as (c) for the rotor with A–A orientation. (e) Snapshots of the numerical simulation of the traveling LCN ring deformations for the A–R orientation. (f) Same as (e) for the A–A orientation.

substrate $\mu = 1.19 \pm 0.04$ was measured by pulling a flat LCN film over the substrate and recording the minimum force (Lutron FG-5000A force gauge) needed to set it in motion under a given load. A video camera (DLT Cam PRO 5 MP, 30 frames/s) was used to record the ring deformation and the motor rotation. A paint mark on the LCN ring edge was used to track and analyze the motion of the micromotor rotor. An IR imaging camera (Fluke Ti400) was used to record the time-dependent temperature distribution in the rotor.

Numerical Simulations of the Photothermal Deformation.

Finite element numerical simulations of the rotor ring deformation dynamics were performed in COMSOL Multiphysics using the Solid Mechanics Module. In the model, laser illumination locally raises the temperature of the LCN ring and thus reduces the liquid crystal order, causing the ring to contract along the local director, and at the same time to swell in the perpendicular dimensions. The photomechanical deformation is modeled by introducing a local photo-strain tensor $\epsilon_{ph}(\mathbf{x}) = \epsilon_{ph} [(1 + \nu) \mathbf{n} \times \mathbf{n} - \nu \mathbf{I}]$ into the stress (σ)–strain (ϵ) constitutive relation: $\sigma(\mathbf{x}) = \mathbf{C} : [\epsilon(\mathbf{x}) - \epsilon_{ph}(\mathbf{x})]$, where $\mathbf{C}$ is the stiffness tensor of the ring, $\epsilon_{ph}(\mathbf{x})$ is the photo-induced strain along the director $\mathbf{n}$ and the Poisson ratio of the ring $\nu = 0.5$. The simulation results are presented in Figure 3e,f and in the Supporting Information Movies 3 and 4 for the two molecular orientations in the LCN rotor ring. The simulations indicate that the light-induced stress in the rotor ring indeed results in a traveling deformation, following the spatially modulated laser beam. The model we use is able to predict the qualitative dynamics of the circular rotor with different

molecular orientations—the exact shape of the deformation in the experiments is more complex, as there are other factors involved, not accounted for in the model: residual stress from the LCN film fabrication, (anisotropic) thermal expansion, and nonlinear relation between the deformation and the light intensity (becoming more important for the light intensities where the photomechanical response saturates).

RESULTS AND DISCUSSION

Light-Powered Deformation and the Traveling Wave Micromotor Rotary Motion. In our LCN micromotor, the rotor deformation is realized by photoheating³⁴—laser light absorption (25 μm film absorbs $\sim 90\%$ of the incident green light) which induces a reversible nematic–isotropic phase transition in the liquid crystal polymer network. As a result, an LCN stripe with homeotropic alignment locally contracts (expands) in the direction parallel (perpendicular) to the molecular alignment (director) and as a result the initially near-flat ring deforms into a complex three-dimensional shape^{35–37} (Figure 3b). More complex, patterned alignments may provide more sophisticated shape-reconfigurable surfaces from flat LCE or LCN sheets,³⁸ in particular the azimuthal–azimuthal (A–A) orientation in a flat film results in a conical deformation, whereas radial (R–R) orientation makes the film deform into a saddle.^{33,39}

With the laser power and the scanning speed adjusted properly, a traveling macroscopic deformation of the LCN is observed, which results in a rotary motion of the ring via friction coupling with the substrate (Supporting Information Movies 1 and 2). The LCN ring deformation resembles the traveling wave of an ultrasonic ring motor. The light-driven motor works in the regime where the laser-induced polymer film temperature increases only locally and, as the light beam moves away, it decreases with a time constant of the order of tens to hundreds of milliseconds, mainly due to conductive heat transfer to the surrounding air.^{31,34} As a result, after many scan cycles (rotations), the average temperature of the rotor remains close to the room temperature, and there is a significant difference in the temperature between the illuminated area (spot) and the surrounding film—Supporting Information Movie 5 shows the temperature distribution in the moving rotor, visualized with an IR camera.

Depending on the molecular alignment in the LCN ring, the actuation in response to the local temperature increase varies. For the azimuthal (A–A) alignment, the ring contracts circularly and swells along the radius and in thickness. The A–R film bends locally, within a sector of around 70° , toward the substrate (sandpaper) and the neighboring sectors bend up and lose contact with the substrate (Figure 3b). If the laser scanning speed is slow enough, the ring deformations follow the laser beam around the ring—Movies 1 and 2 in the Supporting Information.

The micromotor rotation speed depends on the light energy absorbed in the material within a given time that is proportional to P/f , where P is the laser power and f is the mirror rotation frequency (in turns/s).³⁴ For a constant laser power, the motor speed increases with the increasing beam scanning speed, up to a point where the time constant governing the photomechanical response in the LCN is too large to allow for enough heat to accumulate and produce a significant ring deformation (Figure 3c). For low laser powers, the film reaches the regime where no significant deformation is present, while at the other extreme of high laser powers,

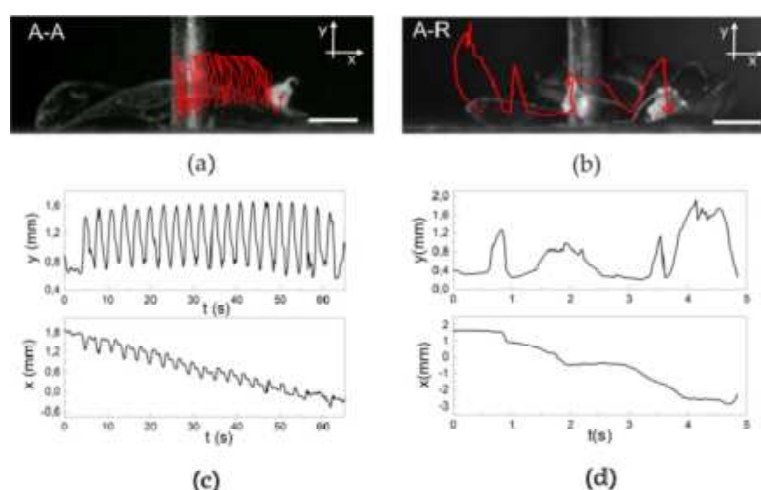


Figure 4. Measured trajectory of one point at the LCN ring edge for two film orientations: A–A (a,c) and A–R (b,d). The white scale bars are 1 mm long. (a,b) Measured (tracked) position of the point at the rotor circumference. (c,d) x and y coordinates of the points in (a,b) vs time.

material damage results. For the A–R alignment, there is no visible difference between the configurations with the A or R side facing up (toward the laser). The micromotor with the A–R LCN ring rotates around ten times faster than the one with the A–A LCN rotor (Figure 3d)—the conical rotor deformation induced in the latter case is not as efficient in the friction coupling to the stator (compare also the trajectory analysis below).

Movie 6 in the Supporting Information demonstrates how the rotor can be friction-coupled to an external axis (made of a 2.1 mg paper tube, compared to a rotor mass of 0.8 mg) to deliver a usable mechanical power output.

Tracking one point at the edge of the LCN rotor offers some insight into the rotor ring deformation and the micromotor kinematics. For the A–A oriented film, the point at the rotor circumference follows a spiral trajectory, reversing after each step, and at the same time, the steps are more regular and shorter. In the case of the A–R orientation, such a point follows a sine-like trajectory, with the motor segments moving in large steps after each laser beam sweep. (Figure 4). The A–A motor needs 172 s to perform a 90° rotation with the average angular frequency $\omega = 0.009$ rad/s while the A–R motor makes a full 360° rotation in 64 s with average $\omega = 0.098$ rad/s. The micromotor efficiency, defined as the ratio between the motor revolutions per minute to the laser beam rotations per minute, is between 0.4 and 0.6% for the A–A orientation and between 4.7 and 8.3% for the A–R orientation.

CONCLUSIONS

Our results show that dynamic photothermally induced deformations in liquid crystal elastomer films can drive rotary motion on a small (millimeter) scale. We harnessed the time-dependent mechanical response of a properly oriented LCN ring, combined with friction coupling to power a rotary micromotor resembling ultrasonic piezoelectric ring motors. In such an elastomer film rotor, the molecular alignment defines the deformation topology, while the photomechanical response time constant, the absorbed light energy and the laser beam scanning speed determine the motor speed. As the A–A orientation offers a more regular motion, it may be more suitable for applications where regulation is of primary importance, rather than the net rotation speed. The

irregularities in the rotor with the A–R orientation most likely originate from a combination of the nonlinear dependence of the photomechanical response to the light intensity, anisotropic thermal expansion of the rotor, and the residual stress in the polymer film.

Most of the polymer light-driven motors and drives presented to date rely on shifting the center of the mass position by photo-induced deformation in a contacting, bending, or twisting element(s)—as a result, the system finds a new, different equilibrium position and this generates motion. Our motor uses the friction coupling between two elements to generate rotary motion and as such is, in principle, independent from gravity.

Despite the low speed and poor efficiency, the LCN micromotor presents a new approach to small-scale mechanics and micro-scale drives and opens up a new class of light-powered, friction-coupled actuators, for example, based on the inchworm drive principle. With the current LCN technology, also using laser photolithography, it can be directly scaled down to sub-millimeter dimensions^{40,41} and may find applications in areas where devices must be powered and controlled remotely and/or without electrical currents being involved.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acsami.9b20309>.

Chemical structure and characterization of the PI, DSC, and DMTA characterization of the LCN films (PDF)

Rotating micro-motor with A–R orientation (MP4)

Rotating micro-motor with A–A orientation (MP4)

Numerical simulation results for the photomechanical response of an LCN rotor ring with A–R orientation (MP4)

Numerical simulation results for the photomechanical response of an LCN rotor ring with A–A orientation (MP4)

Temperature distribution in the rotating micro-motor (MP4)

Ring micro-motor turning a tube on the axis (MP4)

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Author Contributions

K.D. and M.R. contributed equally. K.D., M.R., and P.G. fabricated the micro-motor and performed the experiments. They also built the experimental setup with contributions from P.W. who conceived the idea of the LCN traveling wave motor. R.W. and W.P. prepared the photo-aligning cells. J.K. and E.S.-B. synthesized the photo-alignment azo(polyimide)s. C.X. performed the numerical simulations. K.D. and P.W. wrote the manuscript with contributions from all authors. All the authors analyzed and discussed the results.

Notes

The authors declare no competing financial interest.

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Publication V

Light-Driven Linear Inchworm Motor Based on Liquid Crystal Elastomer Actuators Fabricated with Rubbing Overwriting

Light-Driven Linear Inchworm Motor Based on Liquid Crystal Elastomer Actuators Fabricated with Rubbing Overwriting

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Abstract: Linear displacement is used for positioning and scanning, e.g., in robotics at different scales or in scientific instrumentation. Most linear motors are either powered by rotary drives or are driven directly by pressure, electromagnetic forces or a shape change in a medium, such as piezoelectrics or shape-memory materials. Here, we present a centimeter-scale light-powered linear inchworm motor, driven by two liquid crystal elastomer (LCE) accordion-like actuators. The rubbing overwriting technique was used to fabricate the LCE actuators, made of elastomer film with patterned alignment. In the linear motor, a scanned green laser beam induces a sequence of travelling deformations in a pair of actuators that move a gripper, which couples to a shaft via friction moving it with an average speed in the order of millimeters per second. The prototype linear motor demonstrates how LCE light-driven actuators with a limited stroke can be used to drive more complex mechanisms, where large displacements can be achieved, defined only by the technical constraints (the shaft length in our case), and not by the limited strain of the material. Inchworm motors driven by LCE actuators may be scaled down to sub-millimeter size and can be used in applications where remote control and power supply with light, either delivered in free space beams or via fibers, is an advantage.

Keywords: liquid crystal elastomer; linear motor; actuator; smart material; rubbing



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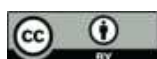
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1. Introduction

Nature does not seem to use rotary motion often. We are aware of only three examples of rotary motion used in the animal kingdom: bacterial flagella are powered by a rotating motor built into the cell envelope, driven by the flow of protons or sodium ions; dung beetles use rolling to transport loads; and some caterpillars from the *Crambidae* family roll away when in danger.

Our civilization, on the contrary, relies chiefly on rotary motion for energy conversion, transport and manufacturing. At the same time, linear motion is vital for many applications, e.g., in handling, positioning and metrology. There are a number of solutions for realizing linear displacement with rotary motors: via rack and pinion, wheel and belt or lead screw. It is also possible to generate linear displacement directly with either a pressurized medium acting on a piston, as in hydraulic and pneumatic actuators, or electromagnetic forces, as in voice coils and more complex linear electric motors. Another approach is based on direct material deformation, most notably in piezoelectric actuators, or in more exotic examples, based on shape memory alloys, or other shape memory material, such as liquid crystalline epoxy resins and composites [1,2], dielectric elastomers or liquid crystal networks [3,4]. Each of these solutions has certain advantages and limitations and several characteristics are taken into account when considering specific applications, among them: speed; stroke; maximum load that can be handled; maximum force that can be generated; accuracy; and locking force at rest with no power supply. In some applications, such as positioners for nuclear magnetic resonance (NMR) scanners, the materials involved in the actuator

constructions can be critical (e.g., non-metallic or non-magnetic components only), while in others, e.g., in environments sensitive to electromagnetic interference, remote control without electric currents may be required.

Liquid crystal elastomers (LCEs) are elastic polymers with an ability to reversibly deform under external stimuli, e.g., temperature. Their rod-shaped molecules can be aligned in cross-linked polymer chains and, upon this arrangement, the order (anisotropic)–disorder (isotropic) transition reduces the effective length of these chains along the alignment direction (the director). This is accompanied by an increase in the spacing between the molecules in perpendicular directions. Consequently, the material can exhibit large, fast and reversible shape changes; thus created deformation geometry is determined by the molecular alignment within the material [5] and can generate forces well exceeding the weight of the deforming element [6,7]. The mechanisms responsible for the photomechanical response can be either photochemical reactions, often associated with the *cis*–*trans* isomerization, or photothermal heating, when the temperature of the material (locally) increases due to light absorption, which can be improved by adding a suitable dye or other absorber. In the case of such heating, generated by light absorption, the fast photomechanical response can be used to remotely power and control mechanisms [8–10]. Selective heating by temporally and/or spatially modulated laser beam(s) has been demonstrated in devices performing various tasks, including rotary micro-motors [11], also in micro-scale [12–14].

Inchworm motors were initially developed as rotary mechanisms with a rotor moved by a sequential action of piezo actuators [15,16]. In a linear inchworm motor, a number of actuators are operated in a sequence, consisting of gripping, moving and releasing the shaft. Inchworm motors in various forms, most commonly with piezoelectric actuators and millimeter- to centimeter-scale are now widely used, mainly in precision fabrication and positioning. Their most prominent advantages are: infinite motion (i.e., not limited by the step length); high speed; sub-micrometer precision and accuracy; high locking force also with no power supply. At the same time, if driven with piezo actuators, inchworm motors require high voltages, typically in the order of tens to hundreds of volts, and dedicated controllers for reliable operation.

Here, we present a light-driven linear motor, using the inchworm principle, with two accordion-like LCE actuators. The actuators use crosslinked liquid crystal elastomer film, polymerized with UV light, with red dye added to increase green light absorption [5]. Photothermal response in the 50 micron LCE film with patterned molecular alignment, fabricated by the rubbing overwriting technique, is used to convert light energy into the movement of a carbon fiber shaft. A spatially modulated laser beam creates a traveling deformation along the actuators that displace a gripper, which is friction-coupled to the shaft. Bi-directional operation is possible with reversed spatiotemporal modulation (scanning) of the laser beam.

2. Materials and Methods

2.1. Light Responsive Actuators

To fabricate light-responsive actuators made of LCE films with patterned molecular alignment, we introduced the technique of mechanical rubbing overwriting. There are several techniques used for inducing molecular orientation in liquid crystals, as well as in the fabrication of liquid crystal polymers or networks, and some of them allow for patterned orientation: laser written gratings [17,18]; photo-alignment [19,20]; imprint lithography [21]; and micro-rubbing [22]. Mechanical rubbing is widely used for liquid crystal orientation: after coating a glass surface with a thin layer of soft material, typically a polymer, a velvet-like cloth is rubbed in one direction. This induces a preferential direction of the molecular orientation in the liquid phase, once such prepared glass is used to make a thin cell, infiltrated with the liquid crystal. The mechanisms involved include micro-grooves being formed and/or local heating of the coating material above the melting point and modification of the surface, as the solidification front moves along, so that the

LC molecules anchor to such treated surface, oriented along the rubbing direction [23]. Rubbing is often used to fabricate LCE films with simple orientations, uniform across the whole film area such as nematic [24–26] or twisted nematic [27,28]. We demonstrated how rubbing with masks that leave only some fragments of the surface exposed could be used to prepare LCE films with patterned orientations. Films with different areas having different molecular alignment and, as a result, different photomechanical responses (deformations) to a stimulus—temperature or light—were fabricated and used to demonstrate a light-driven crawling caterpillar micro-robot [29]. This procedure involved the mechanical rubbing of glass slides spin-coated with a layer of poly (vinyl alcohol) with a home-built rotating drum rubbing machine and two masks (hand- or laser-cut out of paper), one for each orientation of the rubbing direction (Figure 1a).

In [30], it was reported that a homogeneous substrate rubbed in two different directions results in the alignment of liquid crystals along the axis oriented between these two directions. The dependence of the alignment axis angle on the number of strokes in the first and second (perpendicular) rubbing directions was presented, together with a theoretical model. Authors of [31] described a method involving the unidirectional rubbing of a polyimide coating, which was subsequently partly covered with a mask formed by photolithography. The surface was then rubbed perpendicular to the first rubbing direction and the mask was removed. The rubbed coating thus produced induced position-dependent orientation of molecules in a liquid crystal light modulator.

Inspired by these results, we have introduced a new approach to producing patterned rubbing—which we call rubbing overwriting—where we combine unidirectional and multidirectional rubbing by applying a mask. In the first step, the entire surface is exposed and rubbed in one direction, and in the second step the mask covers certain areas and the exposed surface is rubbed in the perpendicular direction (Figure 1b). In this new procedure, the tedious step of mask alignment is eliminated.

A 50 μm -thick cell was made with two glass slides oriented in such a way that the areas were rubbed in a certain direction on one slide face the areas rubbed in perpendicular direction on the other slide. The cell was filled with the monomer mixture containing 83 mol% of monomer 4-methoxybenzoic acid 4-(6-acryloyloxyhexyloxy) phenyl ester (Synthon Chemicals), 15 mol% of crosslinker 1,4-Bis-[4-(3-acryloyloxypropyloxy)benzoyloxy]-2-methylbenzene (Synthon Chemicals), 1 mol% of photoinitiator 2-benzyl-2-(dimethylamino)-4'-morpholinobutyrophenone (Irgacure 369, Sigma-Aldrich) and 1 mol% of red dye N-ethyl-N-(2-hydroxyethyl)-4-(4-nitrophenylazo)aniline (Disperse Red 1, Sigma-Aldrich). The cell was filled on a hot plate at 80 °C by capillary forces, then heated to 140 °C for 2 min and cooled on an aluminum plate at 22 °C. After 15 min, the mixture was polymerized with a 379 nm UV LED. White light polarizing microscope observations confirmed that the film had rectangular areas with twisted nematic director orientation, where the adjacent segments had their orientation rotated by 90° (Figure 1d) and a stripe cut perpendicular to these segments deformed into the accordion-like shape when heated up (Figure 1e), similar to the results of previously reported experiments, where photo orientation was used for patterned alignment [32].

The homogenous film, using the same LCE composition and heated uniformly in an oven, had the maximum contraction of 35% at 130 °C. The linear actuator stroke could be increased (amplified) using bending instead of contraction and the accordion-like deformation was demonstrated to provide up to an 80% reduction in the initial length of the actuator [29].

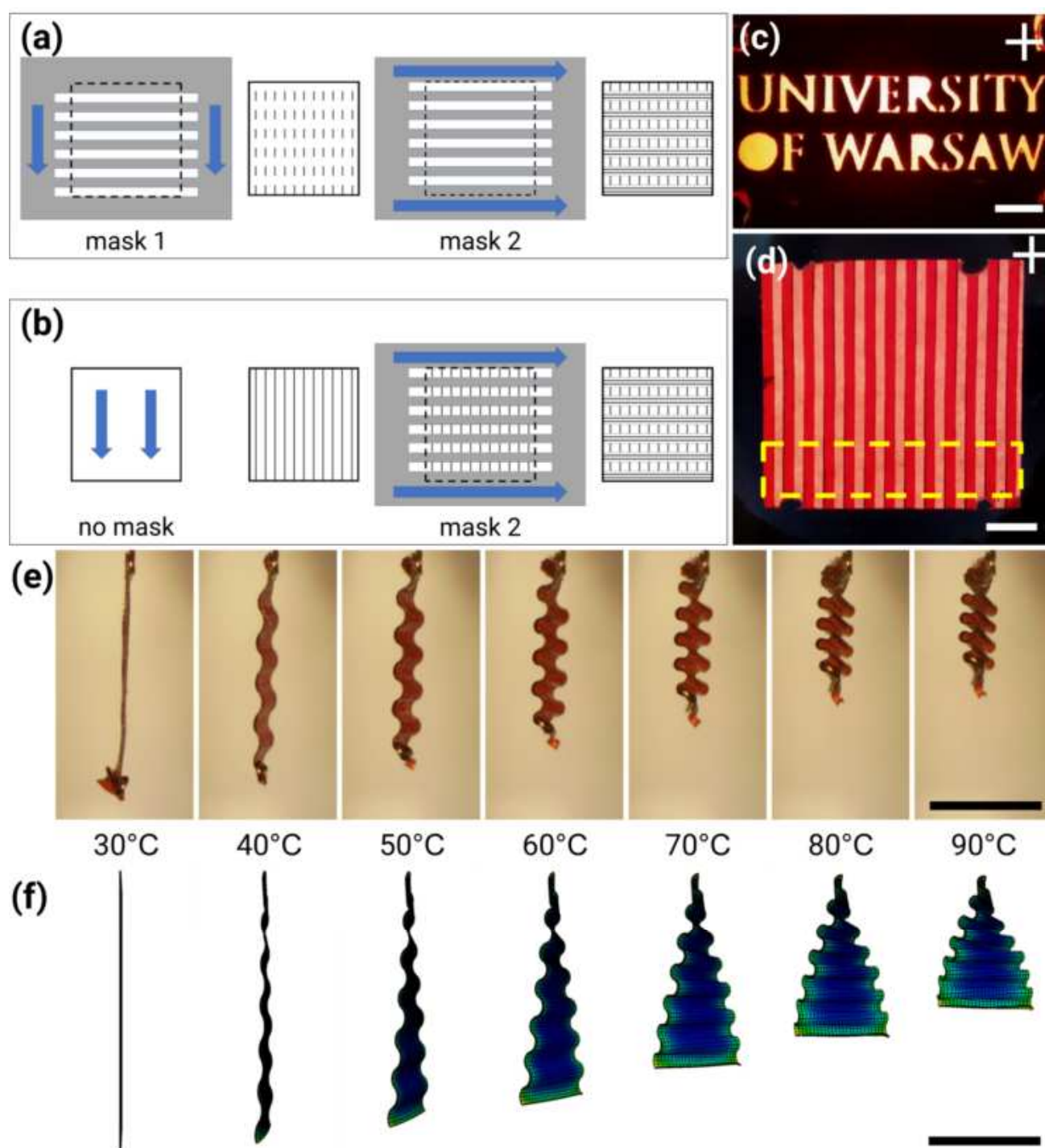


Figure 1. Two approaches to the fabrication of LCE films with patterned orientation. In the previously used method, two masks (grey) were used, one for each rubbing direction (indicated by blue arrows and grey lines) (a). In the rubbing overwriting method, the entire surface is rubbed in one direction first and one mask is used for the regions with a perpendicular rubbing direction (b). (c,d) LCE films prepared with rubbing overwriting technique using: (c) a mask with the University of Warsaw logo cut out and (d) a mask with 1 mm wide slits cut out. The yellow box shows how the accordion-like actuator was cut from the film. White light photographs were taken between crossed polarizers, with the white cross showing the orientation of the polarizers. (e) Accordion-like deformation in a stripe of LCE film cut perpendicular to the pattern lines, heated from 30 °C to 90 °C. (f) Finite element simulation of the LCE stripe deformation. Temperature values relate to conditions in (e,f). All scale bars are 5 mm long.

2.2. Linear Inchworm Motor

Light-induced travelling deformation in an LCE ring was used to demonstrate a rotary micro-motor (5.5 mm in diameter), similar in design to the travelling wave piezoelectric motors used in, e.g., focusing mechanisms of camera lenses [6]. Building on these results, we have designed a light-driven linear motor with infinite stroke, i.e., not limited by the actuator's deformation, but, in principle, only by the shaft length. By sequential action of the two accordion-like LCE actuators, powered by a scanned laser beam, a small mass (gripper) moves in an orbital trajectory. At the lower part of this trajectory it comes into friction coupling with a shaft and pulls it a step (typically around 1 mm long) in one direction. Repeated cycles of this motion result in large linear displacement of the shaft and the sequence order (defined by the laser beam scanning direction) defines the shaft movement direction (Figure 2a).

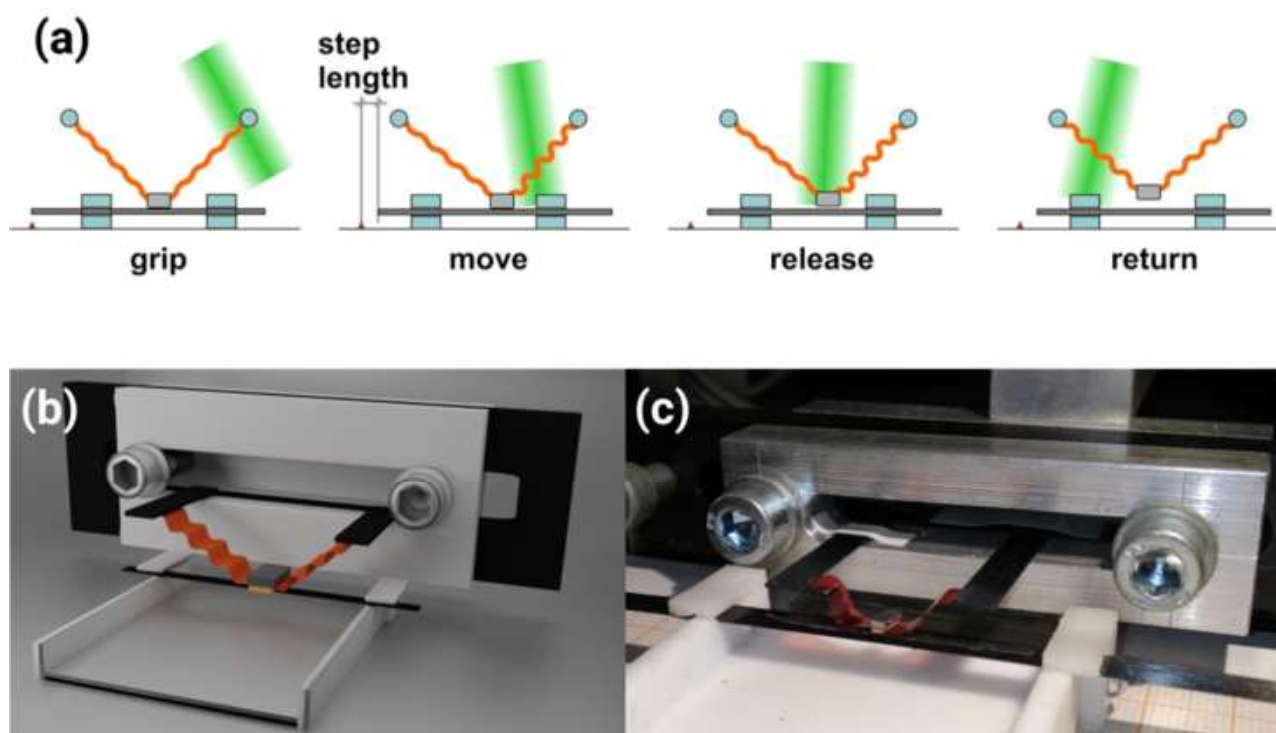


Figure 2. (a) The light-driven linear inchworm motor principle of operation. During the slow scan of the laser beam (green), the deformation traveling along two light-responsive elastomer accordion-like actuators (orange) results in the gripper (gray) coupling to the shaft by friction, pulling the shaft by one step, releasing the gripper by pulling it up and returning it to the initial position with the relaxation of the elastomer. Three-dimensional model of the linear motor (b) and a photograph of the device (c) (the M6 screw heads, 10 mm in diameter, give a scale reference).

Each of the LCE actuators was 13 mm long, 5 mm-wide stripe, cut perpendicular to the segmented rubbing (Figure 2c). One end of the actuator was attached (glued) to a stationary support and the other to a gripper, made of a lead block (106 mg weight) with a piece of 2000 grit sandpaper glued to the bottom surface. A strip cut from the same sandpaper was also glued to the top side of the carbon fiber shaft (0.2 × 5 mm cross section, 120 mm length, 144 mg total weight). The shaft was sliding in two polytetrafluoroethylene (Teflon) slits (Figure 2c). The motor was powered by a laser beam (shaped with a cylindrical lens into an ellipse with full widths at half maximum of 4.5 mm and 0.4 mm), reflected from a mirror mounted on a galvo scanner, scanned along the actuators. The galvo scanner was controlled by an asymmetric sawtooth signal from a waveform generator. Each slow scan of the laser beam across the two LCE actuators (scanning speed on the actuator surface was between 21 mm/s for 0.4 Hz stepping frequency and 47 mm/s for 0.9 Hz stepping frequency) resulted in one motor step, consisting of: gripping the shaft; moving (pulling)

the shaft by the gripper; detaching the gripper from the shaft; and returning the gripper to the initial position, as presented in Figure 2a. The slow scanning direction determined the direction of the net shaft movement. The fast backward scan of the laser beam (scanning speed on the actuator surface above 3 m/s) did not induce any visible response in the LCE actuators [33]. The motor operation was recorded with a CMOS camera (640×480 pixels, 90 fps, DLT-Cam PRO 5 MP, Delta Optical, Nowe Osiny, Poland) mounted on a stereo microscope. To visualize the thermal response of the LCE actuators after light absorption, an infrared camera was also used (320×240 pixels, 60 fps, FLIR A35sc, FLIR, Wilsonville, OR, USA).

The photomechanical response of the LCE actuator was simulated using the finite element method (FEM) with coupled temperature-displacement implicit analysis (SIMULIA, Abaqus/CAE). The actuator was modeled as a $13 \text{ mm} \times 5 \text{ mm} \times 50 \text{ }\mu\text{m}$ cuboid and the LCE as an elastic material with a 500 MPa Young's modulus and 0.4 Poisson's ratio. The contraction of the polymer network in response to a thermal stimulus was taken into account as a negative temperature expansion coefficient along the director orientation (α_{33}) and a positive coefficient in two other orthogonal directions (α_{11} , α_{22}). The total sample volume was made constant by setting $\alpha_{11} = \alpha_{22} = -0.5 \cdot \alpha_{33}$. Based on the experimental results, the contraction was modeled to take place within the 30–120 °C temperature range and was up to 30%. Mass density, specific heat and thermal conductivity were 960 kg m^{-3} , $1200 \text{ J kg}^{-1} \text{ K}^{-1}$ and $0.49 \text{ J m}^{-1} \text{ K}^{-1}$, respectively. The simulation domain was divided into 1 mm-wide sections along the longest dimension. In each section, the director orientation gradually changed by 90° from the top to the bottom of the actuator; in the neighboring sections, the director arrangement was flipped by 90° [29].

The LCE stripe exchanged heat with the environment by radiation (the emissivity was 0.67) and convection (with a moderate heat transfer coefficient of $0.49 \text{ W m}^{-2} \text{ K}^{-1}$). The temperature was first increased to 120 °C, inducing the wave-like deformation of the actuator, and then decreased to 24.5 °C, resulting in its return to the initial flat state. The experimental results (Figure 1e and Video S1, Supplementary Materials) were compared with the simulation results (Figure 1f and Video S2).

3. Results and Discussion

Figure 3 and Video S3 demonstrate the bi-directional operation of the light-driven linear motor. Using image tracking software, the shaft position versus time was measured. The analysis of a single step revealed that the shaft moved forward and then retracted slightly during each cycle of the laser beam scanning (Figure 3a). First, at rest, the gripper was in contact with the shaft, due to gravity and the elasticity of the actuators pushing it towards the shaft. As the first actuator was activated with light, the shaft was pulled by the friction force between the gripper and the shaft. Just before the gripper detached, being pulled away (in the upward direction) from the shaft, it moved in the opposite direction (Figure 3e,f, frame 100.42 s): the net length of one motor step depends on the difference between these two displacements. In the first few steps in Video S3, the warm-up phase is visible, when the average temperature of the LCE actuators gradually increased with each laser scan, until it reached a steady-state operation. Figure 3f and Video S4, recorded in the infrared, reveal the temperature distribution in the LCE actuators during the motor operation. The actuators were locally heated up by the absorbed laser light, reaching approximately 120 °C, and the photomechanical response, resulting in contraction, followed. This sequence of light absorption → temperature increase → deformation → relaxation depends on the interplay of the properties of the photo-responsive material (absorption coefficient, thermal conductivity) and its coupling to the environment, leading to heat dissipation via conductivity, emission and convection, and has been previously analyzed [33].

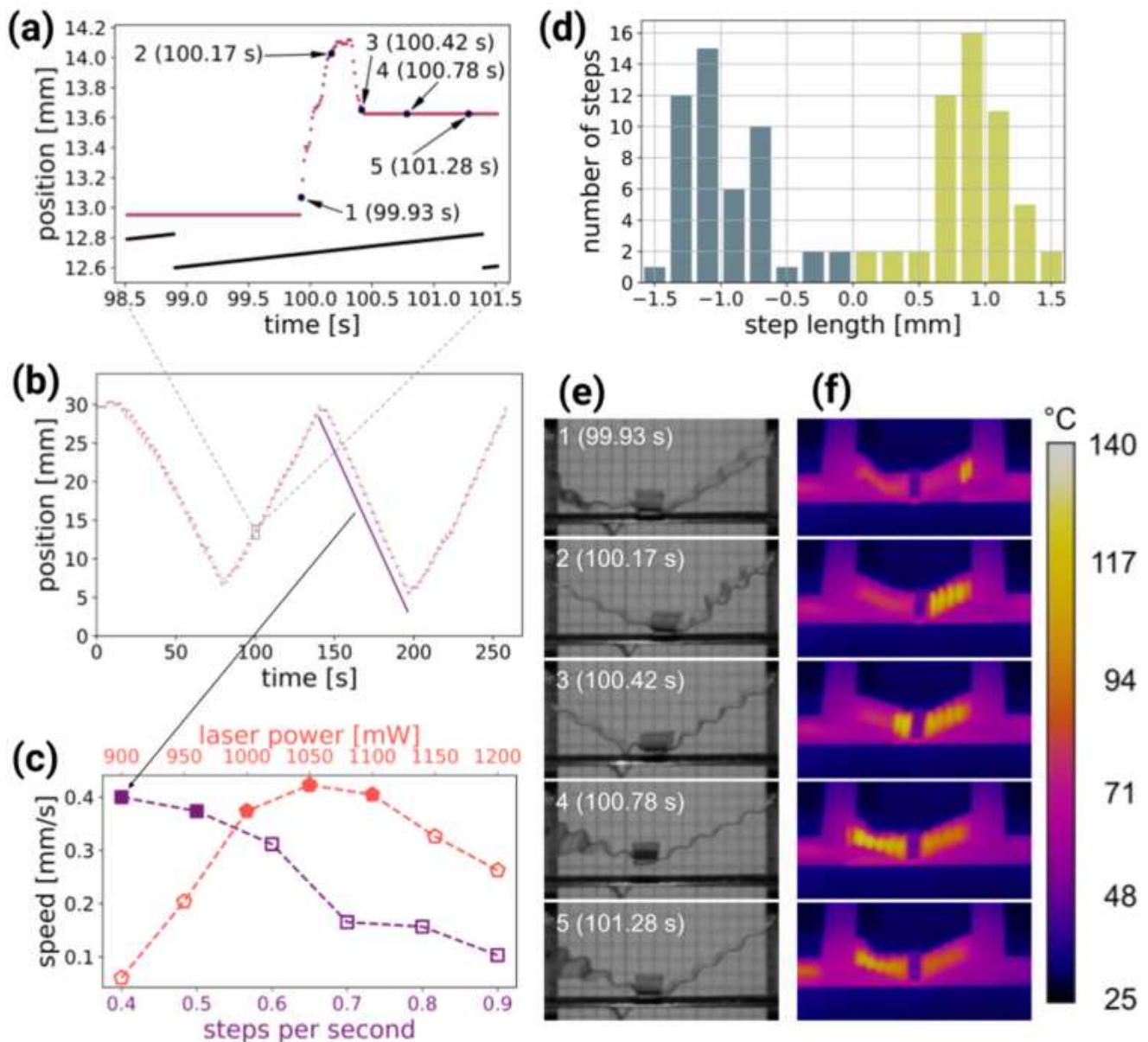


Figure 3. Performance of the light-driven linear inchworm motor: (a) horizontal position of the shaft during one laser scanning cycle (red points) and the laser beam scanning galvo signal (black line). The numbers correspond to the positions in the photographs in (e). (b) The shaft position versus time for continuous operation of the motor, with the scanning direction changed three times. The average shaft speed is measured by fitting straight lines (purple line) to the position over many steps. (c) The motor average speed for different scan frequencies and laser powers. Filled symbols represent the parameters where repeatable, linear motor operation was achieved, while open symbols represent parameters where stable operation was not possible (see text for details). (d) Step length histogram of the measurement presented in (c). The motor operation during one cycle recorded with visible (e) and infrared (f) camera. A triangular marker is visible, attached at the bottom of the shaft, that was used for position tracking. Scan parameters in (a–e): 0.4 steps per second, 1 W laser power.

To determine the average motor speed, linear functions were fitted to the points corresponding to the unidirectional motor motion (Figure 3b), and the average of the absolute values of the slopes was calculated. The average speed was measured against two parameters: the laser power and the laser beam scanning frequency. The first set of measurements was performed with a constant laser beam scanning frequency (0.5 scans s^{-1}) and varying laser power (Figure 3c, red pentagons), and the second with a fixed laser power (1000 mW) and varying scan frequency (Figure 3c, purple squares).

Laser power that enabled motor operation with a scanning frequency of 0.5 scans per second ranged from 900 to 1200 mW. For laser power below this range, the gripper did not release the shaft, and for power above this range, the LCE actuators were damaged. With a constant scanning frequency, the highest average shaft speed (0.42 mm s^{-1}) was measured with a laser power of 1050 mW. When using lower powers, the backward shift in each step became longer and the net step length decreased, which resulted in lower average speed. When using higher powers, the LCE actuators absorbed too much energy to be able to relax between scans, which resulted in the shortening of the shaft movement phase within the cycle.

In the second series of experiments, the scanning frequency was changed from 0.4 to 0.9 scans s^{-1} with 0.1 scans s^{-1} step, with a constant laser power of 1000 mW. Overall, the lower the scanning (stepping) frequency, the higher the average shaft speed, with the maximum of 0.40 mm s^{-1} for 0.4 scans s^{-1} . For this frequency, the step length histogram is plotted in Figure 3d. It reveals an average step length of $(0.94 \pm 0.32) \text{ mm}$, with $(0.91 \pm 0.32) \text{ mm}$ and $(0.97 \pm 0.33) \text{ mm}$ for the right and left motor operation directions, respectively.

When using parameters that resulted in a lower shaft speed (below approximately 0.35 mm s^{-1} , open symbols in Figure 3c), the motor operation became less stable: the step length varied from scan to scan, and the average shaft speeds depended on the scan direction. In this “non-linear” mode of operation, the motor can still be used, but for accurate positioning, some kind of position sensor and a feedback loop would have to be added.

For some applications, the holding force on the shaft with the unpowered motor is an important parameter. In our linear motor, this force was measured to be 1.3 mN, independent of the direction.

4. Conclusions

We used the rubbing overwriting technique for fabricating patterned liquid crystal elastomer films. The method involved multidirectional mechanical rubbing with a mask to obtain materials with complex molecular order. Using the actuators made by this technique, we demonstrated that the idea of a linear inchworm motor can be implemented in a light-powered, centimeter-scale device, using LCE actuators with a thermally mediated photomechanical response. With properly chosen driving parameters—laser power and stepping frequency—reliable operation of the linear motor was achieved.

Although, in the current prototype, the actuator response to the light stimulus is highly nonlinear and the shaft positioning accuracy is limited, the linear motor can still be used in applications where a large stroke is more important than the positioning accuracy or repeatability. For more demanding applications, a contactless position sensor (encoder), e.g., optical or magnetic, can be added, and the motor can operate in the closed loop configuration, where the exact length of each step (or even residual backward steps) is not critical and a wider range of driving parameters can be used. Scanned laser beam illumination can be replaced with a different modulated illumination, e.g., two light sources, activating the entire LCE actuator, operating in sequence.

For the increased precision that may be required in certain applications, the motor can be operated in a hybrid mode, where the stepping action is used to approximately reach the destination position and a proportional response of the actuators is used to fine-tune the final shaft position to within one step length.

The idea of a light-powered inchworm motor can be directly implemented in an infinite-angle-of-rotation rotary motor (positioner) by replacing the straight shaft with a circular one.

With direct laser writing, the LCE actuators can be scaled down to the sub-millimeter scale, where patterned rubbing can—be realized by either photo-alignment [34] or multiple alignment [18].

Supplementary Materials: The following are available online at <https://www.mdpi.com/article/10.3390/ma14216688/s1>, Video S1: Thermo-mechanical response of the accordion-like LCE actuator heated in an oven, Video S2: Results of the FEM simulations of the thermo-mechanical response of the LCE actuator, Video S3: Light-driven linear inchworm motor operation, Video S4: Light-driven linear inchworm motor operation filmed in the infrared to visualize the temperature distributions in the actuators.

Author Contributions: Conceptualization, P.W.; Methodology, M.R., J.H. and P.W.; Investigation, M.R. and J.H.; Writing—Original Draft Preparation, M.R., J.H. and P.W. All authors have read and agreed to the published version of the manuscript.

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Conflicts of Interest: The authors declare no conflict of interest.

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Conclusions

This dissertation aimed to advance the understanding and thus the use of stimuli-responsive materials for constructing untethered, functional devices capable of generating controlled motion at small scales. The research focused on three central themes: programmability, locomotion, and miniaturization. Programmability refers to the ability to encode complex deformation patterns into the material structure and behavior, allowing deformation to follow predetermined shapes and activation sequences. Locomotion is explored through the development of soft, bio-inspired robots and actuator-driven mechanisms that convert light-induced deformation into directional movement. Miniaturization is addressed by developing compact systems that operate without onboard electronics or tethered components.

A series of five publications presents original light-driven systems that employ LCEs as actuators, along with the development of a dedicated workstation (RoboLEC) for fabricating LCE materials for these and other applications. The systems include two bio-inspired soft robots and two types of motors, with particular focus on achieving spatiotemporal control of deformation using laser illumination. Light absorption-induced heating, combined with custom optical setups, enabled programmable actuation modes without the need for built-in electronics or other control systems, thus realizing the idea of “material as a machine”.

RoboLEC enables automated fabrication of LCE films with controlled molecular alignment. Although samples prepared with RoboLEC were not used in any of the projects presented in this dissertation, the workstation represents the most recent achievement described here. To be adopted for routine fabrication, the system requires further improvements, particularly to simplify the calibration process used for positioning glass substrates in sandwich-like cells. One of the test samples with periodically varying molecular alignment fabricated using RoboLEC demonstrates a pattern similar to that used in the linear motor introduced in this work. In that motor, however, the desired alignment was achieved using the novel rubbing overwriting technique, also introduced in this dissertation. The linear motor is capable of bi-directional, continuous shaft movement, by lifting and shifting a load many times heavier than the LCE actuators integrated into the design. The second motor system presented in this work is a rotary motor based on a photo-aligned LCE film shaped into a ring with a +1 topological defect. These two motors, linear and rotary, complement each other by achieving the two most desired types of motion in mechanical systems: translational and rotational. Both devices are powered by a scanning laser beam, and the direction of motion is controlled by the direction of the scan.

The use of light as a stimulus also enabled the creation of soft robots that mimic the locomotion strategies of living organisms. One of the examples presented is a snail-inspired robot powered by a traveling wave of deformation. This design demonstrates that synthetic mucus can provide sufficient adhesion for the robot to crawl upside down and navigate obstacles. The second robot replicates the movement of a water strider. Although its speed is much lower than that of its natural counterpart, the experiment confirmed the fundamental operating principle: time-asymmetric bending and relaxation of the LCE-actuated legs is sufficient to generate propulsion on the water surface.

This is also an appropriate place to reflect on unachieved goals and outline future directions. One regret is that the results of the underwater swimming robot optimization experiments have not yet been published. I was deeply engaged in this project, which led to the surprising discovery of a self-oscillating locomotion mode, where the robot moves by oscillating at sound-range frequencies under continuous laser illumination. Although the work is ongoing, a preprint manuscript is already available on arXiv [94]. Other advanced-stage projects I am involved in focus on DLW. One of them, conducted in collaboration with Zofia Dziekan, explores the possibility of controlling molecular alignment via printing direction during the DLW process. If successful, this would open a path to spatially programmable alignment within microstructured materials. Despite similar approaches being recently reported [95, 96], I believe our method addresses important gaps left in the existing literature. The project I currently enjoy the most involves quantitative measurements of the maximum activation frequency of LCE microactuators.

What makes this project particularly rewarding is that it is being carried out by two high school students, Olga Górna and Marcelina Wawrzyńczyk. I am proud of their progress and believe this work has the potential to become a rare example of a high-quality research paper authored by students before they even begin their formal academic education.

As I finish this work, I would like to share with the reader that I had two goals in writing this dissertation. The first was, of course, to obtain a PhD degree. The second was to introduce the members of the research group I hope to lead in the future to the world of liquid crystalline elastomers, including the methods, challenges, and opportunities in this area. As I write these concluding words, I believe I am on the path to achieving both.

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Author's contribution

This dissertation is based on five publications, with the author identified as the main contributor in **Publications II–V (Publication IV** equally contributed with a co-author). My role in each publication is outlined below using the CRediT (Contributor Roles Taxonomy) system, along with a brief description of my specific contributions. These have been confirmed by the leader of projects leading to each publication and by the co-authors most involved in the work.

Publications II–V were co-funded by the Diamentowy Grant awarded to the author. It is also worth noting that **Publication III** was published several months before the official start of my PhD studies, as the execution of the same grant. However, this work has not been included in any other thesis and has not been used to obtain any other academic degree. The publication was reported as a scientific achievement in my annual report submitted to the Doctoral School of Exact and Natural Sciences at the University of Warsaw.

Publication I

CRediT: Methodology, Investigation

I was responsible for integrating the photoalignment into the device presented in the article. I also prepared reference samples to serve as benchmarks for those produced using the developed setup.

Publication II

CRediT: Conceptualization, Methodology, Investigation, Writing - Original Draft, Writing - Review & Editing, Visualization

I was the main contributor to most of the projects presented in the article and prepared a significant part of the manuscript and figures. Since the article summarizes the research group's progress in the field of LCEs and includes previously published results, the focus here is on the original contributions, specifically the robot that mimics water striders. I conducted the majority of the experimental work on this project, most notably designing and constructing the robot.

Publication III

CRediT: conceptualization, methodology, investigation, Writing - Original Draft, Writing - Review & Editing, Visualization

I carried out the main work in preparing the LCEs used in the experiment, designing and building the experimental setup, and preparing the visual materials (figures and videos) for publication. I also contributed to writing the manuscript.

Publication IV

CRediT: conceptualization, methodology, investigation, review & editing, visualization

I participated in preparing the LCE samples using cells provided by co-authors from the Military University of Technology. I also played a leading role in building the experimental setup and prepared the videos included in the supplementary materials.

Publication V

CRediT: conceptualization, methodology, investigation, writing original draft, review & editing, visualization

With the exception of the simulations, all experimental results presented in this publication were measured exclusively by me. I prepared all LCE samples used in the experiments, developed the final design of the motor and all its prototypes, and proposed the inclusion of a load in the system to increase the blocking stress. I also developed software for position–time tracking and prepared all visual materials (excluding those related to simulations). In addition, I wrote the majority of the manuscript.

Statement on the Use of Artificial Intelligence Tools

This thesis was written with the assistance of ChatGPT-4o, which was used as a native-speaker editor to improve the language and correct grammar. I used the tool with caution, as it is not capable of fully understanding the context in many cases. I adopted the following workflow: writing the text in English independently and using ChatGPT only for language correction, while avoiding Polish-to-English translation.

Co-authors statements

Oświadczenie

Wyrażam zgodę na wykorzystanie niżej wymienionych artykułów w rozprawie Mikołaja Rogóza pt. „Zasilane i sterowane światłem elementy i układy z inteligentnych materiałów polimerowych” (ang. „Light-driven components and systems made of smart polymer materials”). Mój wkład w powstanie tych prac jest następujący:

1. Mikołaj Rogóż, Zofia Dziekan, **Klaudia Dradrach**, Michał Zmysłony, Paweł Nałęcz-Jawecki, Przemysław Grabowski, Bartosz Fabjanowicz, Magdalena Podgórska, Anna Kudzia, Piotr Wasylczyk, „From Light-Powered Motors, to Micro-Grippers, to Crawling Caterpillars, Snails and Beyond—Light-Responsive Oriented Polymers in Action”, *Materials* 2022, 15, 8214, doi:10.3390/ma15228214

Wkład: konceptualizacja, opracowanie metodologii, prowadzenie badań, przygotowanie manuskryptu, recenzja i redakcja tekstu

2. **Klaudia Dradrach**, Mikołaj Rogóż, Przemysław Grabowski, Chen Xuan, Rafał Węglowski, Jolanta Konieczkowska, Ewa Schab-Balcerzak, Wiktor Piecek, Piotr Wasylczyk, „Traveling Wave Rotary Micromotor Based on a Photomechanical Response in Liquid Crystal Polymer Networks”, *ACS Applied Materials & Interfaces* 2020, 12, 8681–8686, doi:10.1021/acsami.9b20309

Wkład: konceptualizacja, opracowanie metodologii, prowadzenie badań, przygotowanie manuskryptu, recenzja i redakcja tekstu

3. Mikołaj Rogóż, **Klaudia Dradrach**, Chen Xuan, Piotr Wasylczyk, „A Millimeter-Scale Snail Robot Based on a Light-Powered Liquid Crystal Elastomer Continuous Actuator”, *Macromolecular Rapid Communications* 2019, 40, 1900279, doi:10.1002/marc.201900279

Wkład: prowadzenie badań, wizualizacja wyników, recenzja i redakcja tekstu

Wyżej wymienione artykuły powstały w ramach mojej pracy w Pracowni Nanostruktur Fotonicznych na Wydziale Fizyki Uniwersytetu Warszawskiego pod kierownictwem prof. Piotra Wasylczyka. Mikołaj Rogóż jest autorem wiodącym w tych publikacjach, z wyjątkiem artykułu 2, w którym oboje jesteśmy autorami wiodącymi. We wszystkich artykułach uczestniczył w konceptualizacji, opracowywaniu metodologii, realizacji prac eksperymentalnych, wizualizacji wyników, recenzji i redakcji tekstu oraz w przygotowaniu odpowiedzi dla recenzentów. Współtworzył manuskrypty artykułów 1 i 3.

Z poważaniem,

dr inż. Klaudia Dradrach



Warszawa, 01.09.2025

Oświadczenie

Jako współautor artykułu:

Mikołaj Rogóż, **Zofia Dziekan**, Klaudia Dradrach, Michał Zmysłony, Paweł Nałęcz-Jawecki, Przemysław Grabowski, Bartosz Fabjanowicz, Magdalena Podgórska, Anna Kudzia, Piotr Wasylczyk, „From Light-Powered Motors, to Micro-Grippers, to Crawling Caterpillars, Snails and Beyond—Light-Responsive Oriented Polymers in Action”, *Materials* 2022, 15, 8214, doi:10.3390/ma15228214

wyrażam zgodę na wykorzystanie wyżej wymienionej pracy jako część rozprawy doktorskiej Mikołaja Rogóza pt. „Zasilane i sterowane światłem elementy i układy z inteligentnych materiałów polimerowych” (ang. „Light-driven components and systems made of smart polymer materials”).

Przygotowywałam ww. artykuł w ramach pracy naukowej w grupie prof. Piotra Wasylczyka na Wydziale Fizyki Uniwersytetu Warszawskiego. Moją wkład obejmował napisanie części manuskryptu, brałam też udział w jego redagowaniu. Uważam, że artykuł powstał przy wiodącej roli Mikołaja w procesie badawczym i, mimo że jest artykułem przeglądowym, są opisane w nim niepublikowane wcześniej wyniki badań przeprowadzone przez Mikołaja dotyczące zasilanego laserem robota imitującego narтника. Potwierdzam, że Mikołaj jest również autorem istotnej części tekstu artykułu i uczestniczył w jego końcowej redakcji.

Z poważaniem,

Zofia Dziekan

Zofia Dziekan

Oświadczenie

Wyrażam zgodę na wykorzystanie niżej wymienionych artykułów w rozprawie Mikołaja Rogóza pt. „Zasilane i sterowane światłem elementy i układy z inteligentnych materiałów polimerowych” (ang. „Light-driven components and systems made of smart polymer materials”). Mój wkład w powstanie tych prac jest następujący:

1. Mikołaj Rogóż, Zofia Dziekan, Klaudia Dradrach, Michał Zmyślony, Paweł Nałęcz-Jawecki, **Przemysław Grabowski**, Bartosz Fabjanowicz, Magdalena Podgórska, Anna Kudzia, Piotr Wasylczyk, „From Light-Powered Motors, to Micro-Grippers, to Crawling Caterpillars, Snails and Beyond—Light-Responsive Oriented Polymers in Action”, *Materials* 2022, 15, 8214, doi:10.3390/ma15228214

Wkład: prowadzenie badań, wizualizacja wyników, recenzja i redakcja tekstu

2. Klaudia Dradrach, Mikołaj Rogóż, **Przemysław Grabowski**, Chen Xuan, Rafał Węglowski, Jolanta Konieczkowska, Ewa Schab-Balcerzak, Wiktor Piecek, Piotr Wasylczyk, „Traveling Wave Rotary Micromotor Based on a Photomechanical Response in Liquid Crystal Polymer Networks”, *ACS Applied Materials & Interfaces* 2020, 12, 8681–8686, doi:10.1021/acsami.9b20309

Wkład: prowadzenie badań, recenzja i redakcja tekstu

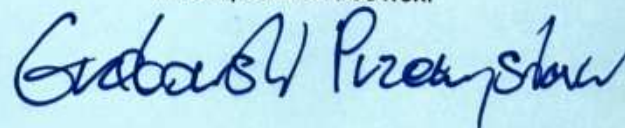
3. **Przemysław Grabowski**, Bartosz Fabjanowicz, Magdalena Podgórska, Mikołaj Rogóż, Piotr Wasylczyk, „Automated photo-aligned liquid crystal elastomer film fabrication with a low-tech, home-built robotic workstation”, *Scientific Reports* 2022, 12, 17598, doi:10.1038/s41598-022-22556-8

Wkład: konceptualizacja, opracowanie metodologii, prowadzenie badań, wizualizacja wyników, recenzja i redakcja tekstu, nadzór projektu

Wyżej wymienione artykuły powstały w ramach mojej pracy w Pracowni Nanostruktur Fotonicznych na Wydziale Fizyki Uniwersytetu Warszawskiego pod kierownictwem prof. Piotra Wasylczyka. Mikołaj Rogóż jest autorem wiodącym w artykułach 1 i 2, przy czym w artykule 2 jego wkład jest równorzędny z dr inż. Klaudią Dradrach. W artykułach 1 i 2 brał udział w konceptualizacji, opracowaniu metodologii, realizacji prac eksperymentalnych, przygotowywaniu wizualizacji wyników, recenzji i redakcji tekstu oraz w opracowaniu odpowiedzi dla recenzentów. Jest współautorem manuskryptu artykułu 1. Artykuł 3 opisuje konstrukcję stacji (ROBOLEC) automatycznie przygotowującej folie z ciekłokrystalicznych elastomerów. W tym projekcie Mikołaj odpowiadał za końcową integrację metody fotoporzadkowania próbek w ROBOLC-u oraz ręczne przygotowanie próbek porównawczych.

Z poważaniem,

Przemysław Grabowski





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prof. dr hab. Piotr J. Wasylczyk
Pracownia Nanostruktur Fotonicznych

Warszawa, 01.09.2025

Oświadczenie

Wyrażam zgodę na wykorzystanie niżej wymienionych artykułów w rozprawie doktorskiej Mikołaja Rogóza pt. „Zasilane i sterowane światłem elementy i układy z inteligentnych materiałów polimerowych” (ang. „Light-driven components and systems made of smart polymer materials”). Mój wkład w powstanie tych prac jest następujący:

1. Mikołaj Rogóż, Zofia Dziekan, Klaudia Dradrach, Michał Zmyślony, Paweł Nałęcz-Jawecki, Przemysław Grabowski, Bartosz Fabjanowicz, Magdalena Podgórska, Anna Kudzia, **Piotr Wasylczyk**, „From Light-Powered Motors, to Micro-Grippers, to Crawling Caterpillars, Snails and Beyond—Light-Responsive Oriented Polymers in Action”, *Materials* 2022, 15, 8214, doi:10.3390/ma15228214

Wkład: konceptualizacja, wizualizacja wyników, przygotowanie manuskryptu, recenzja i redakcja tekstu, nadzór projektu

2. Klaudia Dradrach, Mikołaj Rogóż, Przemysław Grabowski, Chen Xuan, Rafał Węglowski, Jolanta Konieczkowska, Ewa Schab-Balcerzak, Wiktor Piecek, **Piotr Wasylczyk**, „Traveling Wave Rotary Micromotor Based on a Photomechanical Response in Liquid Crystal Polymer Networks”, *ACS Applied Materials & Interfaces* 2020, 12, 8681–8686, doi:10.1021/acsami.9b20309

Wkład: konceptualizacja, recenzja i redakcja tekstu, nadzór projektu

3. Mikołaj Rogóż, Jakub Haberko, **Piotr Wasylczyk**, „Light-Driven Linear Inchworm Motor Based on Liquid Crystal Elastomer Actuators Fabricated with Rubbing Overwriting”, *Materials* 2021, 14, 6688, doi:10.3390/ma14216688

Wkład: konceptualizacja, przygotowanie manuskryptu, recenzja i redakcja tekstu, nadzór projektu

4. Mikołaj Rogóż, Klaudia Dradrach, Chen Xuan, **Piotr Wasylczyk**, „A Millimeter-Scale Snail Robot Based on a Light-Powered Liquid Crystal Elastomer Continuous Actuator”, *Macromolecular Rapid Communications* 2019, 40, 1900279, doi:10.1002/marc.201900279

Wkład: konceptualizacja, wizualizacja wyników, przygotowanie manuskryptu, recenzja i redakcja tekstu, nadzór projektu

5. Przemysław Grabowski, Bartosz Fabjanowicz, Magdalena Podgórska, Mikołaj Rogóż, **Piotr Wasylczyk**, „Automated photo-aligned liquid crystal elastomer film fabrication with a low-tech, home-built robotic workstation”, Scientific Reports 2022, 12, 17598, doi:10.1038/s41598-022-22556-8

Wkład: przygotowanie manuskryptu, recenzja i redakcja tekstu, nadzór projektu

We wszystkich wyżej wymienionych pracach pełniłem rolę koordynacyjną w ramach kierowania działalnością Pracowni Nanostruktur Fotonicznych na Wydziale Fizyki Uniwersytetu Warszawskiego. Dyskutowałem wyniki z osobami zaangażowanymi w projekty badawcze oraz doradzałem w ich realizacji. Prace były częściowo finansowane z grantu NCN OPUS nr 2018/29/B/ST7/00192 pt. „Mechanizmy wykonawcze w mikro-skali na bazie foto-responsywnych polimerów”, w którym pełniłem rolę kierownika. Mikołaj Rogóż jest autorem wiodącym w artykułach 1–4, przy czym w artykule 2 jego wkład jest równorzędny z dr inż. Klaudią Dradrach. We wszystkich wymienionych artykułach uczestniczył w opracowywaniu metodologii i prowadzeniu prac eksperymentalnych. W artykułach 1–4 uczestniczył w konceptualizacji, przygotowywaniu wizualizacji wyników, redakcji i recenzji tekstów publikacji oraz w przygotowaniu odpowiedzi dla recenzentów, a także współtworzył manuskrypty artykułów 1, 3 i 4.

Z poważaniem,
Piotr Wasylczyk

