



## Programmed morphing of liquid crystal networks



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### ABSTRACT

A promising candidate for the development of stimuli-responsive morphing materials are based on liquid crystal polymer networks. These anisotropic materials will contract along the alignment director and expand perpendicular to it when subjected to an anisotropy-reducing stimulus, such as heat. As the liquid crystals can be aligned prior to polymerization using various alignment techniques, it is possible to create networks with programmed, complex director profiles in three dimensions. This review shows the various designs that can be implemented and the complex morphing behavior that can be achieved in liquid crystal polymer networks.

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### 1. Introduction

Stimuli-responsive polymer materials, which respond to stimuli from the environment by changing their properties, are one of the focal points in materials science [1]. An important class of materials are those which reversibly change their shape upon exposure to a stimulus [2]. In particular, the ability to program a certain morphology into the material itself, which can be accessed at any time by applying the stimulus, would lead to materials that can be tailored to achieve a target actuation behavior without the need to assemble separate mechanical components. Applications for such materials could range from moving elements in microfluidic systems [3], such as shutters and flow controllers, to medical systems [4] and robotics [2c,5], where precisely controlled, complex movements are desired.

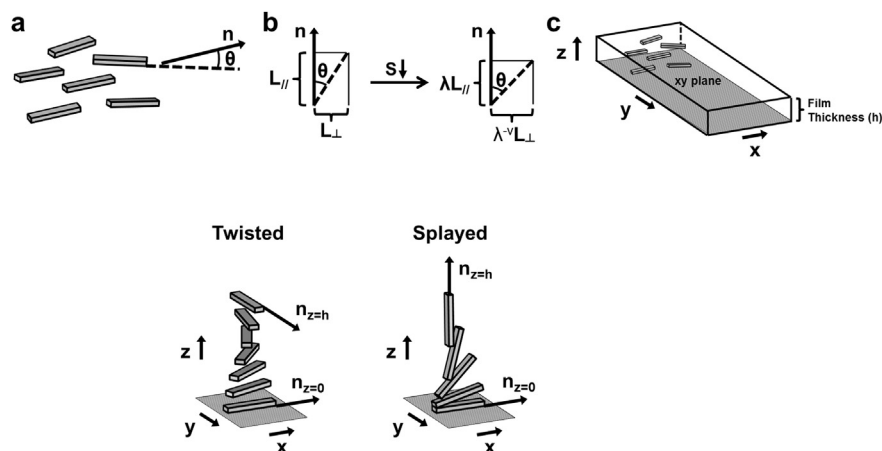
Promising candidates for the development of such materials are based on nematic liquid crystals. Liquid crystals possess a phase that has both anisotropic and fluid-like properties [6]. The material consist of rod-like molecules with orientational alignment that can be described by the alignment director  $n$  and the order parameter  $S$ , where  $S = \frac{1}{2}3\cos^2\theta - 1$ , with  $\theta$  ( $0 < \theta < \pi/2$ ) being the angular deviation from  $n$  (Fig. 1a) [7]. Liquid crystals typically have a multi-domain microstructure, but the alignment of the molecules can be controlled using various alignment techniques, such as electric fields, magnetic fields, or command surfaces, which allows

excellent control over the molecular alignment even at macroscopic length scales.

When liquid crystals are equipped with polymerizable end groups, and a crosslinker is present, they can be polymerized into molecularly well-ordered polymer networks, commonly referred to as liquid crystal networks. UV initiated radical polymerization is usually the chosen polymerization technique, which can be performed on liquid crystals containing acrylate moieties. The polymer network formed retains the molecular alignment of the monomers [6,8]. By combining such reactive mesogens with alignment techniques, excellent control can be obtained over the molecular arrangement within the material, though the restrictions imposed by the alignment techniques often require the liquid crystal polymer networks to be prepared as thin films. In addition, the alignment techniques tend to work only on monomers and not polymers. Depending on its glass transition temperature, the network can either be an elastomer or a glass at room temperature, which have different mechanical properties. However, in both cases the films undergo a shape deformation upon changing the temperature. An increase in temperature lowers  $S$  and therefore increases the average  $\theta$ . This manifests as a contraction  $\lambda$  (with  $\lambda < 1$ ) along the nematic director ( $L_{||}$ ) and an expansion  $\lambda^{-\nu}$  perpendicular to it ( $L_{\perp}$ ), where  $\nu$  is the Poisson ratio that relates the responses of  $L_{||}$  and  $L_{\perp}$  (Fig. 1b) [9]. In the presence of molecules that respond to other stimuli, the order parameter of the polymer network becomes susceptible to other triggers and the dimensions may change, for instance, upon contact with water or chemicals or by being exposed to light of a specific wavelength. The ability to change shape upon actuation by an external trigger makes these

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**Fig. 1.** a) Schematic representation of the orientational alignment of rod-like molecules in the nematic liquid crystalline phase. b) Deformation of a liquid crystal polymer network upon decreasing the order parameter  $S$ . As the average  $\theta$  increases, contraction ( $\lambda < 1$ ) occurs parallel to the alignment director ( $L_{//} \rightarrow \lambda L_{//}$ ), and expansion occurs perpendicular to it ( $L_{\perp} \rightarrow \lambda^{-1} L_{\perp}$ ). c) A liquid crystal polymer film depicted as a coordinate system, with the  $xy$  plane parallel to the surface of the film and the  $z$  axis from  $z = 0$  to  $z = h$ , where  $h$  is the film thickness. The liquid crystals are shown in a planar alignment, with the alignment director parallel to the  $xy$  plane.

materials of interest for their use as actuator. The most popular liquid crystal actuators are those which contain mesogens based on azobenzene. This mesogen will undergo a trans-to-cis conformational change upon irradiation at 365 nm, which causes a deformation similar to heating. This allows for the preparation of UV-responsive films, which are desirable due to light being a well-controlled and versatile stimulus [10].

While the linear contraction and expansion behavior of liquid crystal actuator is interesting [11], their true potential lies in the generation of bending into three-dimensional shapes [12]. A common strategy to achieve bending is by using a bilayer consisting of a responsive liquid crystal network and a non-responsive polymer layer. Alternatively, bending can be achieved by exposing the material to an inhomogeneous stimulus. For example, when a UV-responsive material containing a UV absorbing dye is exposed to irradiation on one side, an intensity gradient will develop through the material. This causes a stronger deformation on one side, and bending takes place [13]. However, it would be more desirable to have access to single-layer, monolithic materials that undergo complex, 3-dimensional shape deformations without depending on the inhomogeneity of the stimulus, as this would greatly increase their versatility and the number of possible applications. To accomplish this, the morphology change has to be programmed into the molecular structure of the material itself, which is possible with liquid crystal networks. In the case of a uniaxially aligned film,  $n$  is constant in all directions. When a homogeneous stimulus is used to actuate such a film, contraction and expansion do not lead to a bending deformation. However, by applying certain alignment techniques, films having a director variation in one or more directions can be prepared and more complex deformations can be achieved. This allows the development of programmable, complex shape deformation. The variation in director is often referred to as the director profile. To describe the director profile, a film can be pictured as a coordinate system, in which the  $z$ -axis run from  $z = 0$  to  $z = h$ , where  $h$  is the thickness of the film, and the  $x$ - $y$  plane is positioned parallel to the surface of the film (Fig. 1c).

To create liquid crystal polymer films with a non-uniform director profile in the  $xy$  plane, an alignment technique that can facilitate the preparation of such alignments in the reactive mesogens is required. Polyimide layers are favored for the alignment of liquid crystals, but while advanced techniques for polyimide buffing are capable of generating non-uniform director profiles [14], the most versatile technique for the generation of

complex director profiles is photoalignment [15]. To prepare photoaligned alignment layers, a photoresponsive material is irradiated with polarized UV light to cause a permanent or reversible chemical transformation. The molecules are more likely to absorb the light, and undergo the transformation, if the transition dipole moment is parallel to the electric vector of the polarized UV light. This creates anisotropy in the alignment layer, which can then be transferred to the reactive mesogens that come into contact with it. There are different photoalignment materials available. A popular class of materials are based on azobenzene. These material undergo a reversible trans to cis rearrangement when they absorb UV light, which causes the molecules to reorient. As the absorption is more efficient for molecules in which the transition dipole matches the polarization direction of the light, the alignment of the molecules is driven towards the direction perpendicular to the polarization direction, and anisotropy is created. Another class of alignment materials are the photopolymerizable polymers (LPP's), which rely on a [2 + 2] cycloaddition reaction between the photosensitive side groups of the polymer, which results in crosslinking of these groups [16].

Liquid crystal network actuators and morphing systems have been the topic of several reviews. These reviews often focus on systems that respond to one specific stimulus [10a,10g,17], or the applications of liquid crystal networks [4,9c,18]. In contrast, this review provides an overview of the methods that lead to complex morphology changes in liquid crystal polymer networks by controlling the director profile of the mesogenic units in three dimensions and without relying on an inhomogeneous stimulus or a bilayer system.

## 2. Alignment director variation through the thickness of the polymer film

### 2.1. Bending

A straightforward way to obtain bending deformations in liquid crystal networks is by utilizing the twisted and splayed nematic alignment profiles (Fig. 2). When a twisted or splayed alignment is present, there is a  $90^\circ$  change of the director along the  $z$ -axis. This causes expansion and contraction in the  $xy$  plane at  $z = 0$  to be different compared to the plane at  $z = h$ , and this mismatch in deformation causes bending. Therefore such films are suitable for the preparation of actuators that behave as cantilevers. The

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