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Research paper

Modeling of surface waves in photo-responsive viscoelastic liquid crystal thin films under a moving light source

Kanishk Mehta^a, Akhil R. Peeketi^a, Jeroen A.H.P. Sol^c, Michael G. Debijs^c, Patrick R. Onck^{*,b}, Ratna K. Annabattula^{*,a}

^a Stimuli-Responsive Systems Laboratory, Department of Mechanical Engineering, Indian Institute of Technology Madras, Chennai, 600036, India

^b Micromechanics of Materials, Zernike Institute for Advanced Materials, University of Groningen, Nijenborgh 4, 9747 AG, Groningen, The Netherlands

^c Laboratory of Stimuli-Responsive Functional Materials and Devices (SFD), Department of Chemical Engineering and Chemistry, Eindhoven University of Technology (TU/e), 5600 MB, Eindhoven, The Netherlands

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ABSTRACT

In this paper we present a computational model to simulate surface waves on photo-responsive liquid crystal thin films under dynamic illumination. The influence of light attenuation is included to model the dynamic photo-mechanical response as a result of the forward (*trans* to *cis*) and the backward (*cis* to *trans*) isomerization of the embedded azobenzene molecules. The viscoelastic nature of the liquid crystal films is also estimated and incorporated in this study. The temporal response of the material under static spot illumination is analyzed first to understand the underlying mechanisms behind the observed response. The relation between the fraction of transformed molecules and the macroscopic light induced strains and stresses through the thickness is investigated to establish the significance of light attenuation. Subsequently, the model liquid crystal film is exposed to a rectangular illumination waveform to simulate a moving light source and thereby generate surface waves. Results are presented that relate the wave attributes, (i.e., the amplitude, frequency and phase lag) to the time scales associated with the moving light source, the molecular *trans-cis* isomerization reaction and the viscoelastic response of the material. A design guideline for producing surface waves with specific features (amplitude, phase) is presented.

1. Introduction

Surface topology is key to a wide range of functions including wetting and de-wetting (Allen et al., 2013), hydrodynamics (Herminghaus, 2000), haptics (Mavroidis et al., 2000; Visschers et al., 2018) and biofouling (Granick et al., 2003). In many additional applications including camouflage and drag reduction (Gutttag and Boyce, 2015), self-cleaning surfaces (Yuan et al., 2007), autonomously adjusting lenses (Dong et al., 2006), scattering, diffraction and reflection in modulated optics (de Jong et al., 2011; Ibn-Elhaj and Schadt, 2001; Schiff et al., 2006), there exists an additional need to dynamically control the surface topography. There are only a few systems that are capable of producing dynamic, reversible surface textures. Among these, stimuli-responsive polymers are gaining distinct prominence owing to their potential application in a broad range of industries (Stuart et al., 2010), such as drug delivery (Ganta et al., 2008), biomedical devices (de las Heras Alarcón et al., 2005), microfluidics (Dong and Jiang, 2007; Sin et al., 2011) and textiles (Hu et al.,

2012). Smart polymer systems exist that are responsive to a wide range of environmental stimuli such as temperature, light, humidity, electric fields, chemical gradients, and magnetic fields (Meng and Hu, 2010; Chen and Chang, 2014; Rath et al., 2018). Of these external stimuli, light-based activation offers non-contact actuation and facilitates fast local switching through spatial control of illumination. Azobenzene-modified liquid crystal (LC) systems are one of the most promising candidates to realize such light-controlled actuation (Gelebart et al., 2017; Liu et al., 2012; Sol et al., 2019).

Azobenzene molecules show isomerisation between a thermally stable *trans* isomer and a metastable *cis* isomer. Under ultraviolet light illumination, *trans*-azobenzene transitions to the *cis* isomer, while the *cis*-azobenzene reverts back to the *trans* isomer by exposure to visible light or thermodynamically to eventually achieve an equilibrium between the forward and the backward reactions. The *trans-cis* isomerisation of azobenzene results in light-induced molecular shape changes which can be further amplified by cross-linking the azobenzene molecules in the polymeric skeleton of ordered liquid crystal networks.

* Corresponding authors.

E-mail addresses: p.r.onck@rug.nl (P.R. Onck), ratna@iitm.ac.in (R.K. Annabattula).

Irradiating such azo-modified liquid crystal systems causes contraction along the average molecular director and expansion in the plane perpendicular to the molecular director (Liu and Onck, 2018). These light-induced local anisotropic deformations can result in a wide range of shape changes by synthesizing films with non-uniform director orientations, varying geometry and boundary conditions (Liu et al., 2012, 2015; Liu and Onck, 2017, 2018, 2019; Pilz da Cunha et al., 2019; Sol et al., 2019; Wie et al., 2015; Yu et al., 2003). The cause of the deformations is the strain gradient that develops in the material due to the variation of light intensity through the thickness and/or due to the varying director orientation within the film. An Azo-LC film with uniform director orientation and cantilever boundary condition bends upon illumination. On the other hand, when the film is bonded to a stiff substrate, out-of-plane surface deformations are observed in the film under exposure to light (Liu et al., 2012, 2015; Liu and Onck, 2018). Thus, the boundary conditions play an important role in engineering the shape changes besides the geometry of the film and the distribution of directors. Additionally, under moving or time varying illumination, dynamic surface patterns can be produced.

Light-induced surface deformations can be utilized in friction control (Hendriks et al., 2017; Koçer et al., 2017; Liu et al., 2015), microfluidics (Mijatovic et al., 2005) and wettability control (Onda et al., 1996). Computational models to predict the light-induced surface deformations help in identifying the underlying mechanisms as well as play a pivotal role in designing reliable and optimized systems. Liu et al., 2015 and Liu and Onck (2018, 2017, 2019) presented a computational photo-mechanical model to investigate the steady-state response of Azo-LC surfaces assuming the film to be a linear elastic material. Since the light-absorbance of *trans* and *cis* azo molecules is different, there exists a two-way coupling between the intensity of light and the fraction of *trans/cis* molecules at a particular depth in the LC film. The steady state computational model presented in Liu and Onck (2018) includes the above-discussed variation in intensity with the fraction of azobenzene molecules and the model has been successfully applied to study the reversible switching of linearly-patterned films, randomly ordered polydomain and fingerprint coatings (Liu et al., 2015; Liu and Onck, 2018), as well as the effect of two-wavelength exposure (Liu and Onck, 2017). In these studies, the emphasis was on steady-state on/off switching of illumination, assuming the duration of the presence/absence of illumination to be much larger than the molecular relaxation time of the azobenzene molecules and the viscoelastic relaxation time of the polymer network. However, when a moving light source is used, the interaction between the internal (material) time scales and the external (light source) time scales becomes an essential ingredient to describe the response of the system. In this paper, we extend the steady state model of Liu and Onck (2018) to consider transient *trans-cis* isomerization and further include the viscoelastic constitutive behaviour of the film. The proposed model is used to investigate the influence of the three time scales (light source, viscoelastic response and *trans-cis* isomerization) on the temporal surface deformations of Azo-LC films.

The paper is organized as follows. In Section 2, we present a transient photo-mechanical model that considers the time dependent light attenuation and the viscoelastic constitutive response of the liquid crystal films. The developed computational model is validated through a comparison with results available in the literature in Section 3. The influence of the viscoelastic nature of the film under a static light source is presented in Section 4, followed by a detailed study on the surface waves under a moving light source. Section 5 lists the conclusions of the work and future outlook.

2. Computational photo-mechanical model

2.1. Photo-mechanical model

The computational model for light-induced surface deformations

presented in Liu and Onck (2018) is extended in the current paper to account for the viscoelastic nature of the Azo-LC films. The Cartesian components of the photo-strain tensor for the Azo-LC films in rate form can be written as

$$\dot{\epsilon}_{ij}^{\text{ph}} = P_{ij} \dot{n}_c, \quad (1)$$

where, i, j correspond to the axes of the local reference frame, P_{ij} is the photo-responsivity tensor and n_c is the volume fraction of the *cis* molecules. Azo-LC polymers contract along the director (i.e., in the x -direction) and expand in the direction perpendicular to the director (i.e., in the y - and z -direction). This anisotropic response results in a photo-responsivity tensor (P_{ij}) with P_{xx} , P_{yy} and P_{zz} as the non-zero components and $P_{yy} = P_{zz}$. The effect of the Azobenzene concentration, the density and length of the cross-linkers and the working temperature are considered to be captured by the photo-responsivity tensor P_{ij} , which is assumed to be constant during light exposure. The sum of the elastic ($\dot{\epsilon}_{ij}^{\text{e}}$), inelastic ($\dot{\epsilon}_{ij}^{\text{in}}$) and the photo-responsive ($\dot{\epsilon}_{ij}^{\text{ph}}$) strain rate gives the total strain rate tensor,

$$\dot{\epsilon}_{ij} = \dot{\epsilon}_{ij}^{\text{e}} + \dot{\epsilon}_{ij}^{\text{ph}} + \dot{\epsilon}_{ij}^{\text{in}}. \quad (2)$$

The stress tensor can be obtained through the basic hereditary integral formulation for linear viscoelasticity as (Roylance, 2001):

$$\sigma_{ij}(t) = \int_0^t C_{ijkl}(t-t') \dot{\epsilon}_{kl}^{\text{e}}(t') dt'. \quad (3)$$

Here C_{ijkl} is the elastic stiffness tensor, which is a function of time t . Note that Einstein's summation convention for repeated indices is followed. The modulus relaxation function can be defined in terms of a series of exponentials known as the Prony series (Smith, 2009):

$$C_{ijkl}(t) = C_{ijkl}^0 \left[1 - \sum_{p=1}^{n_p} C_{ijkl}^p (1 - e^{-t/t_{ijkl}^p}) \right] \quad (\text{no sum on the indices}), \quad (4)$$

where C_{ijkl}^0 represents the initial modulus, and C_{ijkl}^p , t_{ijkl}^p are the Prony series parameters. On solving Eq. (3) using integration by parts, the inelastic strain is obtained as:

$$\epsilon_{kl}^{\text{in}}(t) = \sum_{p=1}^{n_p} \frac{C_{ijkl}^p}{t_{ijkl}^p} \int_0^t e^{-(t'-t)/t_{ijkl}^p} \epsilon_{kl}^{\text{e}}(t') dt' \quad (\text{no sum on the indices}). \quad (5)$$

The instantaneous shear modulus can be related to the elastic modulus for an isotropic material as

$$E(t) = 2G(t)(1 + \nu). \quad (6)$$

The equations specifying the rate of *cis* molecules ($\dot{n}_c$) to be substituted in Eq. (1) is a result of the transient nature of the light attenuation and *trans-cis* isomerization and will be presented in the following section.

2.2. Transient light attenuation model

Upon illuminating the surface of an Azo-LC film with UV-light, the rate of isomerisation will be higher for the top surface as compared to the deeper layers due to the attenuation (drop in light intensity) with depth. The isomerisation of *trans* to *cis* affects the attenuation since the light absorbance of *cis* is much smaller than that of *trans*. This difference in absorbance results in an increased intensity at deeper layers as the time of illumination increases. The coupled *trans-cis* isomerisation and light-attenuation model can be formulated as (Corbett and Warner, 2008)

$$\dot{n}_c = (1 + \beta \zeta \Phi(z, t)) - [1 + (\alpha + \beta) \zeta \Phi(z, t)] n_c(z, t), \quad (7)$$

$$\frac{\partial \Phi}{\partial z} = \left[\left(\frac{1}{d_t} - \frac{1}{d_c} \right) n_c(z, t) + \frac{1}{d_c} \right] \zeta \Phi(z, t), \quad (8)$$

where, $\Phi(z, t) = I(z, t)/I_0$, is the normalized local light intensity, d_c and d_t are the attenuation lengths of the *cis* and *trans* isomers, respectively¹; α and β are parameters that describe the capacity of light to trigger the forward and backward reactions, respectively²; τ is the average lifetime of the *cis* conformer and depends on the temperature; n_c and n_t are the mass fractions of the *cis* and *trans* isomers, respectively, adding up to one:

$$n_t + n_c = 1, \quad (9)$$

and ζ is defined as the polarization coefficient, quantified as

$$\zeta = \frac{1}{3}[1 - SP_2(\cos \phi)], \quad (10)$$

where ϕ is the angle between the director and the direction of light propagation, $P_2(x) = (3x^2 - 1)/2$ is the second Legendre polynomial, S is an order parameter describing the orientational order of the LC molecules with respect to the director. There also exists a relation between the attenuation lengths d_c , d_t and the ratio of the quantum efficiencies (η) of the forward and backward reaction given by

$$\frac{d_t}{d_c} = \frac{\beta \eta_c}{\alpha \eta_t}. \quad (11)$$

The coupled Eqs. (7) and (8) need to be solved together with the boundary and initial conditions. For surface topological deformations, the film is bonded to the substrate. The initial condition for the transient simulations is that the mass fraction of the *trans* isomer is one throughout the domain, i.e., $n_t(z, 0) = 1$. The 'photo' boundary condition is that the top surface of the film that is exposed to light will have $\Phi(h, t) = 1$ for the time under illumination, with h as the thickness of the film. For the case of a moving light source, the set of nodes with $\Phi = 1$ get updated based on the displacement of the light source at each step. When the film is not illuminated, the thermal back reaction takes place. The equation for the back reaction can be obtained by substituting $\alpha = 0$ and $\beta = 0$ in Eq. (7), so that

$$\tau \dot{n}_t = 1 - n_t. \quad (12)$$

In this work, light sources moving along a straight line are considered to generate surface waves as will be discussed in Section 4. Eventually, the prospect of producing complex temporal surface topographies is demonstrated through the generation of a sinusoidal pattern.

3. Model validation

The photo-mechanical parameters employed in this work were taken from Liu and Onck (2018), shown in Table 1, unless specified otherwise.

The photo-mechanical model discussed above is validated with the numerical/experimental results available in the literature. Solving Eqs. (7) and (8) (for each time t , the values of Φ and n_c are iteratively solved until a converged solution is achieved) provides the evolution of the mass fractions of the *cis* and *trans* isomers, n_c and n_t . The numerical solution of the evolution of Φ and n_c with time was illustrated by Knežević et al. (2013). The model developed here includes the added effect of polarization of light through the polarization coefficient (ζ) as described in Eq. (10). For $\zeta = 0.433$ the results obtained here are similar to Knežević et al. (2013) as shown in Figs. 1a and 1b. Note that even a small change in β , relative to the value of α , drastically affects the intensity and the fraction of the transformed molecules as shown by the dashed and solid lines in Fig. 1.

The viscoelastic and light-attenuation models described in the previous section are implemented in the commercial finite element

software ABAQUS (Smith, 2009). The light attenuation model and the resulting photo-induced strain are incorporated through the user subroutine UTEMP. The discretization of the film is done using eight-noded brick (C3D8) elements, and a dynamic implicit analysis is carried out. First, a local spot as shown in Fig. 2 is illuminated in three materials with different elastic properties (isotropic $E = 500$ MPa, isotropic $E = 1000$ MPa and transversely isotropic $E_{xx} = 1000$ MPa and $E_{yy} = E_{zz} = 500$ MPa (Liu and Onck, 2018)) with a planar director alignment (along the x-axis in Fig. 2). Note that even when the elastic properties of the film are considered to be isotropic, the induced photo-strain is still transversely isotropic due to the anisotropic nature of the photo-responsivity tensor P_{ij} . The photo-mechanical parameters are taken as shown in Table 1, with variations only in the elastic properties to highlight the differences in strain and stress developed in the material. On attaining steady state, the illumination is switched off to observe the relaxation in each of the three materials. Fig. 3 shows the evolution of the normalized displacement (u_z/h) and von-Mises stress as a function of time. The figure describes two regions with the left portion describing the duration for which the light is 'on' and the right side representing the duration when the light is 'off'. The response described in Fig. 3 corresponds to point P in Fig. 2. The strain estimated by the transversely isotropic elastic model is around 6.4% at steady-state as shown in Fig. 3a and is close to the results available in the literature (Liu et al., 2015). For an isotropic material, the magnitude of the normalized vertical displacement is independent of the elastic modulus as the photo-induced strain in the z-direction is a function of the mass-fraction of *cis* molecules Eq. (1) only (for $D \gg h$) (Liu and Onck, 2018). On switching off the illumination, the material relaxation (strain and stress) to its original state develops more gradually compared to the growth rate. The response of a purely elastic material to the backward reaction after switching off the light is independent of the stress/strain/modulus at a point and is fully driven by the molecular back reaction described by Eq. (12). As each material has an identical steady-state value of n_c , the time taken for stress/strain relaxation is the same for all the three elastic materials. The von-Mises stress is proportional to the elastic modulus and hence the stress is higher for $E = 1000$ MPa compared to $E = 500$ MPa as illustrated in Fig. 3b. Since the transversely isotropic material has its directional moduli as 1000 MPa and 500 MPa, the von-Mises stress response lies between the von-Mises stress responses of the two isotropic materials.

As seen from Fig. 3a, a material with isotropic elastic properties (strain of 7%) predicts about 10% more strain than one with transversely isotropic elastic properties (strain of 6.4%). Since the difference between the isotropic and transversely isotropic cases is minimal, an isotropic viscoelastic model is employed in this work. The inclusion of anisotropy in the developed viscoelastic photo-mechanical model is reserved for future work. The initial elastic modulus is taken as $E = 1000$ MPa for the viscoelastic simulations.

An Azo-based LC film is synthesized (the experimental procedure is described in the supplementary information) on which a stress relaxation experiment is carried out to record the change in modulus with time. Viscoelastic-A in Fig. 4 shows the measured values of the shear modulus as a function of time under a constant strain of 0.3% at 35°C. Liquid crystal films show diverse relaxation responses depending on the temperature, cross-linking density and other parameters. Hence, three additional viscoelastic responses named, Viscoelastic-B, Viscoelastic-C and Viscoelastic-D as shown in Fig. 4 are considered to develop a comprehensive understanding of the effect of viscoelasticity on light-induced surface deformations. The properties of the four different materials are given in Table 2. The molecular reaction time scale is taken as $\tau = 600$ seconds as reported in the literature (Gelebart et al., 2017) for all further simulations unless mentioned otherwise.

Table 2 shows the Prony parameters utilized to model the shear modulus response of each of the viscoelastic materials namely

¹ The values of d_c and d_t depend on the concentration of azo molecule in the film and are sensitive to the wavelength of light.

² The values of α and β scale with input intensity, are temperature dependent, and sensitive to the wavelength of the light.

Fig. 1. The time evolution of Φ and n_c through the film thickness (z/d_t) including the effect of polarization of light obtained by solving Eqs. (7) and (8). The solid lines represent the case when $\alpha = 36$, $\beta = 0$ and $\eta = 3$, while the dashed lines correspond to $\alpha = 36$, $\beta = 1$ and $\eta = 3$ identical to Knežević et al. (2013).

Table 1
Photo-mechanical parameters.

Photo-mechanical parameters				Mechanical Properties			
α	30	d_t	1 μm	P_{xx}	-0.2	E_0	1000 MPa
β	3	d_c	3.33 μm	P_{yy}	0.12	ν	0.3
η	3	ζ	0.433	P_{zz}	0.12	G_0	384.6 MPa

Viscoelastic-A, Viscoelastic-B, Viscoelastic-C and Viscoelastic-D. The parameters are obtained by fitting prony series to the relaxation functions so as to encompass a range of viscoelastic responses.

4. Results and discussion

4.1. Static illumination

It is essential to understand the chemical conversions and their effect on the light attenuation to study the material response under unpolarised illumination. The mass fraction of *cis* molecules (n_c) is highest at the top surface and decreases through the thickness since the light intensity is maximum at $z = h$ and reduces as it travels through the material thereby converting fewer *trans* molecules to *cis* as seen in Fig. 5a. Moreover, n_c at the top surface reaches the steady-state much earlier than the inner regions which is an outcome of the light-attenuation model. The n_c at a particular depth reaches a steady-state only after n_c at all regions above that depth have saturated, as shown in Fig. 1 and schematically depicted in Fig. 5b with $I_2 > I_1$ (see also Liu and Onck (2018)). On switching off the light, the thermal back reaction of *cis* to *trans* takes place resulting in a reduction of the mass fraction of *cis* molecules as can be seen in Fig. 5. The backward reaction shows an exponential decay as it follows first-order kinetics Eq. (12).

The logarithmic strain at a particular depth increases with a rise in n_c at that depth and peaks when n_c at that depth reaches a steady-state (i.e., it saturates) as seen in Figs. 5a and 6a. The logarithmic strain is maximal at the surface and decreases with depth because the photo-strain described by Eq. (1) is proportional to n_c which is highest at the surface and reduces with depth (see Fig. 1). Once n_c at a depth attains a steady-state, the viscoelastic material begins converging towards an

Fig. 2. Side-view and top-view of a photo-responsive LC film fixed to a substrate. The film is exposed to a circular beam of light at the center under static illumination. The ratio of the film length and thickness is $L/h = 25$ ($L = 250 \mu\text{m}$, $h = 10 \mu\text{m}$) and the ratio of the diameter of the light beam to the film's thickness is $D/h = 4$ ($D = 40 \mu\text{m}$, $h = 10 \mu\text{m}$). The plots discussed in the following figures are for the point P shown in the figure above unless mentioned otherwise.

elastic material through local relaxation. The local relaxation in Viscoelastic-A is a result of the extra strain produced due to the strain rate dependent mechanical response (a function of the rate of *trans* to *cis* conversion) as opposed to an elastic material. This reasoning can be substantiated by the observation that the difference in maximal strain between a viscoelastic and an elastic material reduces with depth because regions at greater depths (lower z/h) have a smaller rate of increase of n_c (slope in Fig. 5a) and hence a lower strain rate dependence. Similarly, the relaxation of the material on switching off the illumination is also different for a viscoelastic material due to its strain rate dependence. Hence, a viscoelastic material initially relaxes much faster

Fig. 3. The transient response of a photo-responsive material under spot illumination and the corresponding material relaxation after switching-off the light at time $t = 5\tau$ (with $\tau = 1$ s) obtained using a purely elastic model. The results are shown at point P (see Fig. 2). a) The engineering strain as a function of time b) The von-Mises stress at a point on the top surface. The employed photo-mechanical parameters are shown in Table 1.

than an elastic material but eventually reaches the steady state strain-free state later due to a strain overshoot (see Fig. 6a). The impact of the light-attenuation model on the surface displacement shows that after the logarithmic strain at the top surface ($z/h = 1$) reaches its maximum at $t = 0.25\tau$, the surface displacement continues to increase as strain is still developing in the lower section of the material (see Fig. 6b). If the difference in photon absorbance between the *cis* and *trans* molecules is not considered, the film surface will simply stop growing as soon as the logarithmic strain in the top surface has saturated, thereby under-predicting the observable deformation.

Similar to the logarithmic strain, the local stress at a particular depth also peaks when n_c at that depth reaches a steady-state as shown in Figure S1. The von-Mises stress (Figure S1) in the viscoelastic material is lower than that in an elastic material due to the relaxation in elastic modulus with time (Fig. 4), resulting in stress relaxation. On switching off the light, an elastic material gradually relaxes whereas a viscoelastic material rapidly relaxes with the axial stresses crossing each others path (Figure S1). The cross-over of the axial stresses results in a minimum in the von-Mises stress response. The relaxation is rapid in a viscoelastic material as the stress is a function of the temporal integral of the fraction of *cis* molecules (by combining Eq. (1) to Eq. (3), and Eq. (12)). On the other hand, stress in an elastic material is directly proportional to the fraction of *cis* molecules (Liu and Onck, 2018).

The relative response of the four different viscoelastic materials is shown in Fig. 7. As the viscous character increases, the maximum strain

decreases and tends towards the maximum strain of a purely elastic material. This is due to the fact that the higher viscosity reduces the strain rate sensitivity. In addition, a material with a lower viscosity experiences less resistance and thus grows and relaxes at a faster rate. Clearly, when given enough time, the displacement overshoot due to the strain-rate is lost and the material relaxes to the displacement produced in a purely elastic material. Similarly, the stress under illumination relaxes faster for a material with lower viscosity (see Fig. 7b). Furthermore, the deformation in the material for a wide range of the molecular reaction time scale (τ) illustrate that the viscoelastic nature of the film dominates for small values of τ and ceases to affect for large values of τ for a given viscoelastic response as demonstrated in Fig. 8.

4.2. Moving light source

The ability to create dynamic patterns on a LC film is investigated by considering a rectangular waveform of a light source moving along a straight line as described in Fig. 9. Attributes of the obtained surface waves such as the amplitude, frequency and the phase lag are related to the molecular reaction time scale (τ), the viscoelastic response and the time period of the light source. The time period of the light source (τ_l) can be defined in terms of the wavelength (L) and wave velocity (v) as

$$\tau_l = \frac{L}{v} \quad (13)$$

Table 2

Prony series parameters Eq. (4) for the viscoelastic responses.

Material response	Viscoelastic - A		Viscoelastic - B		Viscoelastic - C		Viscoelastic - D	
Prony Parameters	G_i	t_i	G_i	t_i	G_i	t_i	G_i	t_i
Unit	No unit	s	No unit	s	No unit	s	No unit	s
$i = 1$	0.33944	28.617	0.19871	49.067	0.14421	62.809	0.0414	29.964
$i = 2$	0.36773	276.5	0.1471	300.38	0.16079	1919.8	0.0436	3312.3
$i = 3$	0.22142	2072	0.2592	2368.8	0	-	0	-

Fig. 4. Shear relaxation data ($G(t)$ - instantaneous shear modulus, G_0 - initial shear modulus) for an Azo-based liquid crystal film measured under a constant strain of 0.3% at 35°C is represented by Viscoelastic-A. Three additional viscoelastic responses, namely, Viscoelastic-B, Viscoelastic-C and viscoelastic-D are studied to sweep a larger range of LC materials (see Table 2 for their properties).

and the relative time scale of the light source is defined as,

$$\frac{\tau_l}{\tau} = \frac{L}{v\tau}, \quad (14)$$

where τ is the molecular reaction time constant.

Figs. 10a and 10b show the out of plane (u_z) displacement of point P (Fig. 9) as a function of time under the influence of a moving light source with two different time periods ($\tau_l = 1500$ s and $\tau_l = 4500$ s). The results are presented for an elastic, Viscoelastic-A and Viscoelastic-D material response to analyze the effect of viscoelasticity on the surface

wave attributes. The response under spot illumination discussed in the previous section can be extended to explain the response of the LC film under dynamic illumination. For instance, the evaluated amplitude of the surface waves is higher for a film with Viscoelastic-A properties as compared to Viscoelastic-D properties irrespective of the moving light velocity time scale (τ_l). As mentioned previously, this is because of the higher strain-rate sensitivity of a Viscoelastic-A material than a Viscoelastic-D material. Consequently, a material with lower viscous characteristics will achieve higher amplitudes of the surface waves. In addition, the amplitude of the surface waves is higher for larger values of the time period of the light source (τ_l) as the material stays under illumination for a longer time to achieve maximum deformation (see Fig. 10b). However, the amplitude saturates beyond a certain value of the time period of the light source as the *trans-cis* isomerizations and the mechanical response attain their steady-state.

The frequency of the surface waves is observed to be independent of the viscoelastic nature of the LC film (Figs. 10a and 10b). Instead, it is directly proportional to the frequency of the moving light source (Eq. (14)) as shown in Figs. 10a and 10b. Thus, for any given LC film, the frequency of the surface waves can be altered simply by tweaking the time period of the external light source. Hence, a significant amount of control is feasible in terms of achievable frequencies of the surface waves. However, there exists a lower limit of the relative time scale for the light source (τ_l/τ) and thereby an upper limit to the achievable frequency of the surface wave. At that limit, rather small amplitudes of surface waves are superimposed on a typical displacement path (Fig. 7a) as shown in Fig. 10c. This is characteristic of the fact that the time any point is exposed to illumination is lower than the time required for the photo-chemical reaction and the corresponding mechanical response to achieve a steady state (i.e., reaching its maximal strain). The displacement path on which the surface waves are superimposed can be deduced from the photo-mechanical parameters and the viscoelastic nature of the material based on the static illumination model as discussed in Section 4.1.

The phase difference of the surface waves accounts for the time lag

Fig. 5. a) The volume fraction of *cis* molecules as a function of reduced time at three different depths (through the thickness) - at the top surface ($z/h = 1$), mid-way ($z/h = 0.5$) and close to the bottom surface ($z/h = 0.2$). b) Schematic demonstrating the effects of *trans-cis* conversion and the consequences of the light attenuation model. Note that $I_2 > I_1$.

(a)

(b)

Fig. 6. The logarithmic strain (a) and the normalized displacement (b) at the mid-point of the spot under illumination as a function of normalized time at three different depths (through the thickness) - at the top surface ($z/h = 1$), mid-way ($z/h = 0.5$) and close to the bottom surface ($z/h = 0.2$) for an elastic material and the Viscoelastic-A material (Fig. 4).

(a)

(b)

Fig. 7. The temporal response in terms of the (a) normalized displacement and the (b) von-Mises stress at a point on the top surface (P) for an elastic and four different viscoelastic materials (Fig. 4) having a planar director distribution. The employed photo-mechanical parameters are mentioned in Table 1.

between the surface wave reaching a 'significant' amplitude and the time corresponding to the mid-point of the rectangular illumination waveform. Section 4.1 described the viscoelastic material behaviour under static illumination illustrating the overshoot followed by relaxation to a displacement value observed in an elastic material. To generalize the surface wave phase characterization for any viscoelastic response, the 'significant' amplitude is attained at a time in between the times at which the displacement approaches a value of $A/\sqrt{2}$, where A is the amplitude. On computing phase differences for different

combinations of the three time scales, the phase lag is found to have little dependence on the viscoelastic time scale as observed in Figs. 10a and 10b. However, there exists a phase dependency on the light source velocity time scale for a given molecular reaction time scale, i.e., Viscoelastic-A in Fig. 10a has a different phase lag from the corresponding waves in Fig. 10b. Also, the dip observed in Fig. 10b around $t = 3000$ s for Viscoelastic-D even before the point P experiences illumination is the effect of the neighbouring points encountering light prior to point P . The reasoning is consistent with the plot illustrating the displacement

Fig. 8. The temporal response in terms of the normalized displacement at a point on the top surface (P) for an elastic and four different viscoelastic materials having a planar director distribution. The molecular isomerization reaction time scale (τ) is 60 s (a) and 6000 s (b).

along the moving light source as will be discussed in Fig. 12b. For a given time period of the light source (τ_l), the phase lag of the surface waves largely depends on the molecular reaction time scale (τ), as clearly observable in Fig. 10d. As the molecular reaction time scale increases, the phase difference rises. Such a behaviour can be ascribed to the fact that photo-responsive materials, especially the Azo-LC films,

require a certain amount of time after the illumination time to grow and relax, the characterization of which is critical for the design of any moving light source application.

Fig. 11 shows the amplitude and phase lag as a function of the relative time scale of the light source velocity for a given material response (Viscoelastic-A and Viscoelastic-D in Fig. 4). The plot is

Fig. 9. A photo-responsive surface under exposure to a rectangular waveform of light moving along a straight line with velocity v . The ratio of length to thickness of the simulated domain is $2L/h = 25$ ($L = 125 \mu\text{m}$, $h = 10 \mu\text{m}$). The illumination pattern experienced by a point P as a function of time is shown, with τ_l representing the time period of the light source. The response of materials with different properties for a range of time periods of the light source are discussed in the main text. The plots obtained are for the point P in the figure unless mentioned otherwise.

Fig. 10. The normalized displacement of the point P (Fig. 9) as a function of time for a range of values of the time period of the light source (τ_l), molecular reaction time scale (τ) and viscoelastic responses (Fig. 4). All other parameters employed in these results are mentioned in Table 1. a), b) Viscoelastic-A has a larger amplitude than an elastic and Viscoelastic-D material for a given set of time scales. Moreover the frequency is chiefly dependent on the time period of the light source analogous to the case of forced vibrations. a) frequency = $6.67 \times 10^{-4} \text{ Hz}$, b) frequency = $2.22 \times 10^{-4} \text{ Hz}$. c) Scenario when the time period of the light source ($\tau_l = 150 \text{ s}$) is much smaller than the molecular reaction time scale ($\tau = 600 \text{ s}$). d) The phase dependency on the molecular reaction time scale (Gelebart et al., 2017) for a given time period of the light source ($\tau_l = 1500 \text{ s}$) and viscoelastic response (Viscoelastic-A - Fig. 4).

particularly important from the design perspective as it assists in choosing appropriate time scales to produce a certain wave amplitude and phase lag. Based on the understanding that the viscoelastic nature has negligible effect on the phase lag (also seen in Fig. 11), only the amplitude of the surface waves can be varied for a given relative time scale of the light source velocity by altering the viscoelastic response of the material. Fig. 12 shows a snapshot of surface deformation along the length of the film at a given instance of time. The independence of the phase difference of the surface waves on the material properties can be seen in Fig. 12a, wherein the Viscoelastic-A and the elastic material

have a similar phase difference with only a slight difference in the amplitude for a given relative time scale of the periodic light source (τ_l/τ). It can be observed in Fig. 12b that the surface waves show a variation of phase lag as a function of the relative times scales of the periodic light source (τ_l/τ). Additionally, the amplitude is highest for $\tau = 100 \text{ s}$ in Fig. 12b and it reduces with decrease in the relative time scale of the periodic light source (τ_l/τ), as discussed earlier. Fig. 13 showcases the formation of a sinusoidal pattern on a surface under a moving light source inching towards the goal of producing complex surface patterns with ease in control. The speed of the light source

Fig. 11. The amplitude and phase lag in the surface waves as a function of the relative time scale of the light source velocity for a material response as described by Viscoelastic-A and Viscoelastic-D (Fig. 4). The values obtained from the simulations (symbols) as well as the fitted curve (dashed line) is shown. Straight lines are obtained for both amplitude and phase lag on the logarithmic time scale.

along the x-axis is kept constant with time while the speed along the y-axis is varied to ensure that the light source traces a sinusoidal path. Thus the velocity of the light source is lowest close to y_{max} and y_{min} and the velocity is highest around y_{mean} . The out-of-the-plane displacement trail is elucidated in Fig. 14. The velocity of the light source is highest in 4 resulting in a dominant trail and a smaller displacement as compared to 2 or 6 wherein a point experiences illumination for a longer time thereby producing a larger displacement.

5. Summary and conclusions

Photo-responsive liquid crystal films are modelled by introducing viscoelastic effects in the constitutive relation of a transient photo-mechanical light attenuation model. The fact that *cis* isomers have a much lower absorbance than the *trans* isomer is accounted for in the two-way photo-chemically coupled light attenuation equations. The material is assumed to have an isotropic viscoelastic nature, while the anisotropic nature of the photo-induced strain is accounted for in the model. Since liquid crystal materials show a wide range of viscoelastic relaxation responses, four different relaxation behaviours are considered in this work. The inclusion of anisotropy in the viscoelastic response and the birefringence effects exhibited by LC thin films offer prospects for future development of the model presented in this paper.

The transient response of the deformation under local spot illumination has been analyzed. The results of the mass fraction of *cis* molecules, logarithmic strain, surface topography and the stresses developed inside the material have been presented as a function of illumination time. Even after the photo-induced strain at the surface has already attained saturation, the material continues to deform until the photo-strain through the thickness of the film reaches saturation. Hence, the time taken for a LC thin-film to attain steady state increases with increase in film thickness. In addition, the viscoelastic nature of the film is demonstrated by comparing the stress and strain response with that of an elastic material. The amplitude of the strain rate driven overshoot during growth and relaxation of the viscoelastic materials decreases with depth, implying that viscous effects will be less prominent farther from the surface due to reduced light intensity. Simulations for a wide range of molecular reaction time scales (τ) relative to the overall illumination time scale (t) show that the significance of viscoelasticity exists only when the molecular *cis/trans* reaction time scale is comparable to the viscous response time (see Fig. 8).

The ability to create dynamic patterns on photo-responsive surfaces has been demonstrated by the generation of surface waves using a moving light source (at constant speed) having a rectangular waveform. The attributes of the surface waves, such as the amplitude, frequency

Fig. 12. a) Snapshots representing the surface waves obtained in an elastic and Viscoelastic-A material for a given relative time scale of the periodic light source (τ_1/τ), b) snapshots representing the surface waves for a range of relative time scale of the periodic light source (τ_1/τ) in Viscoelastic-A material.

Fig. 13. The images (1 to 8) show the material (Viscoelastic-A) response to a light source moving along a sinusoidal curve. A similar sinusoidal pattern is formed on the surface with the points of maximum deformation moving with the light source and the previously illuminated material relaxing back to the strain-free state thereby forming a tail. With more complex movements of the light source, many more dynamic surface patterns can be generated. The dashed line shows the path of the light.

Fig. 14. Snapshots representing out-of-plane normalized displacement along the sinusoidal path (l is the total length of the sinusoidal curve) developed in the material (Viscoelastic-A) corresponding to image 2, 4 and 6 in Fig. 13.

and phase lag are related to the time period of the light source (τ_l), the molecular reaction time scale (τ) and the viscoelastic nature of the material. The amplitude is found to be dependent on the viscoelastic nature and the relative time scale of the light source velocity (τ_l/τ). The amplitude is proportional to the relative time scale (τ_l/τ) whereas the phase lag of the surface wave is inversely proportional to τ_l/τ , eventually tending to zero for very large values of τ_l/τ . Subsequently, a sinusoidal pattern was generated by controlling the directional speeds of the light source to demonstrate the potential to create dynamic surface topographies. More advanced simulations including multiple

illumination waveforms in different directions with varying time periods ($\tau_l = f(t)$) can now be performed to produce complex cyclical or ever-changing surface patterns. Such a flexibility can create novel possibilities for applications that may require controlled wettability, friction control, particle droplet transport and complex microfluidic networks.

Declaration of competing Interest

The authors declare that they have no conflict of interest.

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Supplementary material

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