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Phys. Rev. E **96**, 032701 — Published 18 September 2017

DOI: 10.1103/PhysRevE.96.032701

The electroclinic effect in chiral smectic-A liquid crystal elastomers

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Chiral smectic-A liquid crystal elastomers are rubbery materials composed of a lamellar arrangement of liquid crystalline mesogens. It has been shown experimentally that these materials shear when subjected to an electric field due to the electrically induced tilt of the director. Experiments have also shown that shearing a chiral smectic-A elastomer gives rise to a polarization. Roughly, the shear force tilts the directors which, in turn, induce electric dipoles. This paper builds on previous works and models the electro-mechanical response of smectic-A elastomers using free energy contributions that are associated with the lamellar structure, the relative tilt between the director and the layer normal, and the coupling between the director and the electric field. To illustrate the merit of the proposed model, two cases are considered - a deformation induced polarization and an electrically induced deformation. The predictions according to these two models qualitatively agree with experimental findings. Finally, a cylinder composed of helical smectic layers is also considered. It is shown that the electro-mechanical response varies as a function of the helix angle.

I. INTRODUCTION

Smectic-A liquid crystal elastomers are rubbery materials composed of mesogens that are uniformly oriented and organized in a layered structure. In their pioneering work, Garoff and Meyer [1] andGaroff and Meyer [2] found that low molecular weight smectic-A* liquid crystals comprising chiral molecules exhibit an electroclinic effect, i.e. there is a direct coupling between the molecular tilt and an applied electric field. In smectic elastomers comprising chiral mesogens, an electric field acting along the layer plane induces a shear deformation. This phenomenon directly emanates from the chirality of the mesogens. Specifically, the electric field couples to the transverse dipole of the chiral mesogens and induces a tilt between the mesogens and the layer normal. As a consequence, the elastomer shears and contracts along the direction of the layer normal. Typically, such elastomers are incompressible and subsequently this deformation is accompanied by an expansion along the transverse directions. Another consequence of the chirality of the mesogens was demonstrated by Lehmann *et al.* [3], where it was found that flipping the direction of the electric field reverses the direction of the mechanical tilt.

Several experimental works were carried out to study the response of a Smectic-A elastomer subjected to an electric field. Spillmann, Ratna, and Naciri [4] mounted a thin sheet of elastomer in a tension clamp with the smectic layers either perpendicular or parallel to the tension clamps and applied an electric field normal to the sheet. An electric field $E = 16.7\,\frac{\mathrm{MV}}{\mathrm{m}}$ was applied to a film whose layers are parallel to the clamps and a contraction of $\sim 1\%$ was measured. Next, the sample was placed such that the layers are perpendicular to the clamps. Upon application of the electric field, an expansion of $\sim 8.2\%$ was measured. Hiraoka *et al.* [5] and Hiraoka *et al.* [6] prepared a smectic elastomer film with an initial thickness $t_0 = 600\,\mu\mathrm{m}$ and applied an electric field $E = 1\,\frac{\mathrm{MV}}{\mathrm{m}}$. They measured an electrically induced tilt angle of $\sim 4^\circ$, a contraction along the layer normal of $\sim 0.2\,\%$ and a shear angle of $\sim 0.08^\circ$. A recent experiment by Spillmann *et al.* [7] demonstrated that smectic elastomers can also twist in response to an electric field.

Previous works have also investigated the purely mechanical behavior of smectic elastomers. The experimental work of Nishikawa, Finkelmann, and Brand [8], Nishikawa and Finkelmann [9] and Spillmann, Ratna, and Naciri [4] demonstrated that the mechanical modulus of Smectic-A elastomers parallel to the layers normal is significantly higher than that along the layer plane. This anisotropy emanates from the lamellar micro-structure of the elastomer. Interestingly, the experiments of Nishikawa and Finkelmann [9] revealed that the elastomer becomes significantly softer above a threshold of $\sim 3\%$ strain along the layers normal.

On the theoretical side, Adams and Warner [10] developed an energy-based model for Smectic-A elastomers which assumes that the layer normal and the director are locked and may only rotate in response to an external force. This assumption prevents a tilt and a Smectic-C like ordering. The work of Stenull and Lubensky [11] argued that the director is not rigidly coupled to the layer normal and as a consequence, a shear deformation can rotate the smectic layers and the director with respect to the layer normal. The later works of Adams *et al.* [12] and Stenull *et al.* [13] provided another model that accounts for the stiffness of the smectic layers and the resistance to the tilt by adding appropriate contributions to the free energy.

From an electric viewpoint, the work of Corbett and Warner [14] theoretically examined the electromechanical behavior of a nematic elastomer subjected to electric fields. In this work, it was shown that the mechanical and electrical anisotropies can cause a thin film expand in the direction of the electric field rather than contract. As a result of the Poisson's effect, the film shrinks along the two directions in the plane of the electrodes. Selinger *et al.* [15] found that under high elec-

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tric fields the state of uniform induced tilt can become unstable and lead to chiral modulations. In their work, they modelled and simulated this phenomenon.

The aim of this work is to model the coupled behavior of Smectic-A elastomers under finite deformations resulting from the electroclinic effect. To this end, we characterize the chirality of the mesogens by a chirality vector which couples to the tilt of the director. The proposed model, presented in Sec. II, is based on the minimization of the free energy of the smectic elastomer. To demonstrate the behavior predicted by the model, two cases are examined in Sec. III - an electrically induced deformation and a deformation induced polarization. In the first case, an electric field is applied to the Smectic-A elastomer and the resulting deformation is determined through the minimization of the free energy. Subjected to this loading, the tilt angle is larger than the shear angle. This trend is in agreement with the experimental findings of Hiraoka *et al.* [6]. We also show that the orientation of the chirality of the mesogens can either enhance or diminish the polarization. In the second case, we follow the experimental work of Kramer and Finkelmann [16] and consider the response of a Smectic-A elastomer subjected to a simple shear deformation. The proposed model predicts that the prescribed deformation rotates the director with respect to the layer normal and therefore induces a tilt of the mesogens. Subsequently, the inherent chirality gives rise to spontaneous dipoles which, in turn, induce a polarization in the elastomer. In accordance with experimental findings, the proposed model predicts that the shear angle is larger than the tilt angle. A cylindrical set-up of a Smectic-A elastomer with helical layers is considered in Sec. IV. The electro-mechanical response of an elastomer with various pitches and helix angles is computed. The main conclusions are summarized in Sec. V.

II. A COUPLED ELECTRO-MECHANICAL MODEL

Consider a Smectic-A elastomer that deforms from a reference to a current configuration due to a coupled electro-mechanical loading. The material points are denoted $\mathbf{x}$ and current positions as $\mathbf{y}(\mathbf{x})$, so that the deformation gradient is defined as $\mathbf{F} = \nabla \mathbf{y}(\mathbf{x})$, where ∇ denotes the gradient operation with respect to the referential coordinate system. The smectic order is characterized by two order parameters - the director and the chiral vector. These are denoted $\hat{\mathbf{n}}_0$ and $\mathbf{q}_0$ in the reference configuration and $\hat{\mathbf{n}}$ and $\mathbf{q}$ in the current configuration. Note that bold letters with hats on top denote unit vectors. The chirality vector is perpendicular to the director such that $\mathbf{q} \cdot \hat{\mathbf{n}} = 0$. The layer normal at the current configuration is given as

$$\hat{\mathbf{k}} = \sqrt{1-q^2}\,\hat{\mathbf{n}} - \hat{\mathbf{n}} \times \mathbf{q}, \tag{1}$$

where q is the magnitude of the chirality vector $\mathbf{q}$. Note that $\mathbf{q} = \mathbf{0}$ gives $\hat{\mathbf{k}} = \hat{\mathbf{n}}$ and characterizes smectic-A while $\mathbf{q} \neq \mathbf{0}$ gives rise to smectic-C. This smectic order gives rise to a spontaneous elongation along the director given by the step-length tensor $\boldsymbol{\ell} = r^{-1/3}\left(\mathbf{I} + (r-1)\,\hat{\mathbf{n}} \otimes \hat{\mathbf{n}}\right)$, where r is a parameter related to the strength of the ordering [12]. It also gives rise to a spontaneous polarization $\mathbf{P}^s = C\,\mathbf{q}$, where C is a parameter.

We assume that the free energy-density function of an elastomer subjected to a uniform electric field by surface electrodes can be viewed as the sum of five contributions,

$$W = W_{tr} + W_l + W_t + W_e + W_m. \tag{2}$$

The first contribution is the classical elastic free energy of an anisotropic elastomer [17]

$$W_{tr} = \frac{\mu}{2}\,\mathrm{Tr}\left(\mathbf{F}\,\boldsymbol{\ell}_0\mathbf{F}^T\,\boldsymbol{\ell}^{-1}\right), \tag{3}$$

where μ is the shear modulus and $\boldsymbol{\ell}_0 = r^{-1/3}\left(\mathbf{I} + (r-1)\,\hat{\mathbf{n}}_0 \otimes \hat{\mathbf{n}}_0\right)$ is the step-length tensor before the deformation. Here, $\mathbf{I}$ is the identity tensor.

The second contribution to the free energy in Eq. (2) concerns the change in the layer thickness,

$$W_l = \frac{B}{2}\left(\frac{1}{\left|\mathbf{F}^{-T}\hat{\mathbf{k}}_0\right|\sqrt{1-q^2}} - 1\right)^2, \tag{4}$$

where B is a measure of the stiffness of the layers. This is identical to the free energy contribution in the works of Adams and Warner [10] and Adams *et al.* [12], but written in terms of the magnitude of the physical chirality vector q. Above $\mathbf{F}^T$ and $\mathbf{F}^{-1}$ denote the transpose and inverse of $\mathbf{F}$, respectively. This formulation is advantageous because it is indifferent to the sign of the director $\hat{\mathbf{n}}$ and therefore satisfies the objectivity requirements associated with the energy-density function.

The third term in Eq. (2) concerns the change in chirality, or equivalently the tilt (or rotation) of the director with respect to the layer normal,

$$W_t = \frac{A}{2}\,q^2, \tag{5}$$

where A is the resistance of the mesogens to tilt. This is again the same as Adams and Warner [10] and Adams *et al.* [12], but written in terms of the magnitude of the chirality vector. Note that Eq. (5) assumes that the ground state is smectic-A, but it can be modified if the ground state is smectic-C.

The fourth term in Eq. (2) pertains to the energetic contribution of the electrical sources to the total energy, see for example Shu and Bhattacharya [18]. Specifically,

$$W_e = -\frac{\varepsilon_0}{2}\mathbf{E}\cdot\mathbf{E} - \mathbf{P}\cdot\mathbf{E} + \frac{1}{2\varepsilon_0\chi}\left(\mathbf{P}-\mathbf{P}^s\right)\cdot\left(\mathbf{P}-\mathbf{P}^s\right), \tag{6}$$

where $\mathbf{P}$ is the polarization, χ is the susceptibility and ε_0 is the permittivity in vacuum.

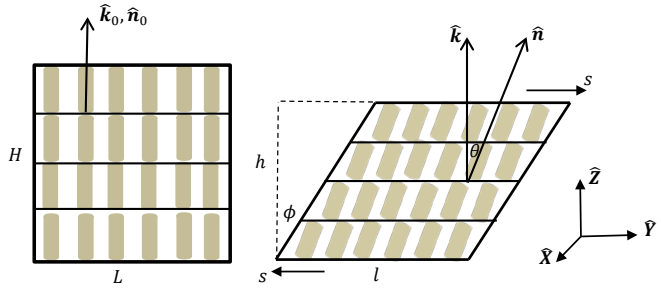

FIG. 1. A schematic of the electro-mechanical response of a Smectic-A elastomer. The chirality vector $\mathbf{q}$ points out of the plane.

To determine the polarization and the deformation under a prescribed coupled electro-mechanical loading, the energy-density function is minimized with respect to $\mathbf{P}$, $\mathbf{F}$ and the chirality vector $\mathbf{q}$. By computing $\frac{\partial W}{\partial \mathbf{P}} = 0$, we find that

$$\mathbf{P} = \varepsilon_0 \chi \mathbf{E} + \mathbf{P}^s = \varepsilon_0 \chi \mathbf{E} + C\,\mathbf{q}. \tag{7}$$

The last term in Eq. (2) concerns the external mechanical loading,

$$W_m = -\mathbf{S} \cdot \mathbf{F}, \tag{8}$$

where $\mathbf{S}$ is the first Piola-Kirchhoff stress tensor.

For convenience, we normalize the energy-density function by $\frac{\mu}{2}$ and substitute Eq. (7) into Eq. (2) to obtain

$$w = \frac{2}{\mu} W = \mathrm{Tr}\left(\mathbf{F}\,\boldsymbol{\ell}_0 \mathbf{F}^T\,\boldsymbol{\ell}^{-1}\right) + b\left(\frac{1}{\left|\mathbf{F}^{-T}\hat{\mathbf{k}}_0\right|\sqrt{1-q^2}} - 1\right)^2 + a\,q^2 - \frac{\varepsilon_0\,(1+\chi)}{\mu}\mathbf{E}\cdot\mathbf{E} - 2\,\frac{C}{\mu}\,\mathbf{q}\cdot\mathbf{E} - 2\frac{\mathbf{S}}{\mu}\cdot\mathbf{F}, \tag{9}$$

where $b = \frac{B}{\mu}$ and $a = \frac{A}{\mu}$ are the normalized layer stiffness and the normalized resistance to tilt, respectively.

The constants in the proposed model can be determined through experiments. The order of magnitude of the shear modulus and the layer stiffness are $\mu \sim 0.1 - 1$ MPa and $B \sim 10$ MPa, respectively [12]. Fitting to experimental findings resulted in a resistance to tilt parameter $A \sim 0.1 - 1$ MPa 19, 20. The anisotropy parameter r has been measured between $1.05 - 60$ in elastomers composed of prolate chains 10, 17. Lastly, we can deduce the order of magnitude of the chirality related parameter from experimental findings. It is found that $C \sim 10^{-3} - 10^{-4}\ \frac{\mathrm{C}}{\mathrm{m}^2}$.

III. THE ELECTRO-MECHANICAL RESPONSE OF SMECTIC-A ELASTOMERS

To examine the predictions of the proposed model, consider a thin film of a Smectic-A elastomer with an initial thickness t_0. We prescribe a global coordinate system $\left\{\hat{\mathbf{X}}, \hat{\mathbf{Y}}, \hat{\mathbf{Z}}\right\}$ such that at the reference configuration the director and the layer normal are along the $\hat{\mathbf{Z}}$- direction, i.e. $\hat{\mathbf{n}}_0 = \hat{\mathbf{Z}}$ and $\hat{\mathbf{k}}_0 = \hat{\mathbf{Z}}$ (See Fig. (1)). The faces of the film are covered with flexible electrodes such that an applied voltage induces an electric field $\mathbf{E} = \frac{V}{\lambda_E\, t_0}\hat{\mathbf{X}}$, where λ_E is the stretch along the thickness of the sheet. Mechanically, an applied stress $s = \mathbf{S}\,\hat{\mathbf{Z}} \cdot \hat{\mathbf{Y}}$ shears the film. The elastomer experiences the general deformation gradient

$$\mathbf{F} = \begin{pmatrix} \lambda_E & 0 & 0 \\ 0 & \lambda_y & \lambda_{yz} \\ 0 & 0 & \lambda_z \end{pmatrix}, \tag{10}$$

where $\lambda_z = \frac{h}{H}$, $\lambda_y = \frac{l}{L}$, $\lambda_{yz} = \lambda_z \tan\phi$ and ϕ is the shear angle (See Fig. (1)). Due to the incompressibility constraint $\lambda_E = \frac{1}{\lambda_y \lambda_z}$. As a result of an electro-mechanical loading, the director rotates such that at the current configuration $\hat{\mathbf{n}} = \sin\theta\,\hat{\mathbf{Y}} + \cos\theta\,\hat{\mathbf{Z}}$. The chirality vector is $\mathbf{q} = \sin(\theta)\,\hat{\mathbf{X}}$, where θ is the tilt angle between $\hat{\mathbf{n}}$ and $\hat{\mathbf{k}}$, in accordance to Eq. (1). Note that in accordance with its definition, the chirality vector is zero when the layer normal is parallel to the director. By making use of Eq. (1), we find that the layer normal at the current configuration $\hat{\mathbf{k}} = \hat{\mathbf{Z}}$.

With the above definitions, the normalized energy-density function in Eq. (9) can be written as

$$\begin{aligned} w(\lambda_y, \lambda_z, \lambda_{yz}, \theta) = {} & \frac{1}{\lambda_y \lambda_z} + \frac{r+1}{2\,r}\left(\lambda_y^2 + r\left(\lambda_{yz}^2 + \lambda_z^2\right)\right) \\ & + \frac{r-1}{2\,r}\Big(\left(\lambda_y^2 + r\left(\lambda_{yz}^2 - \lambda_z^2\right)\right)\cos(2\theta) \\ & - 2\,r\,\lambda_{yz}\,\lambda_z \sin(2\theta)\Big) + b\left(\frac{1}{\cos\theta} - 1\right)^2 \\ & + a\,\sin^2\theta - 2\,c\,V\,\lambda_y\,\lambda_z\,\sin\theta \\ & - \left(\frac{V}{V_d}\right)^2 \lambda_y \lambda_z - 2\,s\,\lambda_{yz}, \end{aligned} \tag{11}$$

where $c = \frac{C}{\mu\, t_0}$ and $V_d^2 = \frac{\mu\, t_0^2}{(1+\chi)\varepsilon_0}$. We also normalize the polarization (Eq. (7)),

$$\mathbf{p} = \frac{1}{\mu\, t_0}\,\mathbf{P} = \frac{1}{\lambda_E}\,\frac{V}{V_p^2}\hat{\mathbf{X}} + c\,\mathbf{q}, \tag{12}$$

where $V_p^2 = \frac{\mu\, t_0^2}{\chi \varepsilon_0}$. The constants V_d and V_p have units of voltage and are related via $\frac{1}{V_d^2} - \frac{1}{V_p^2} = \frac{\varepsilon_0}{\mu\, t_0^2}$.

In order to demonstrate the merit of the model, we adopt the typical values $a = 1$, $b = 5$, $c = 10^{-4}\,\frac{1}{\mathrm{V}}$ and $r = 1.5$. The constants $V_d = 13.7\,\mathrm{kV}$ and $V_p = 15\,\mathrm{kV}$ correspond to a film with a thickness $t_0 = 0.1\,\mathrm{mm}$ and a susceptibility $\chi = 5$.

A. Voltage induced deformation

Due to the electroclinic effect, Smectic-A elastomers deform in response to an applied electric field 1, 3, 4, 6, 7. The aim of this section is to examine the voltage induced deformations according to the proposed model. To this end, we take $s = 0$ and minimize the energy-density function $w(\lambda_y, \lambda_z, \lambda_{yz}, \theta)$ (Eq. (11)) with respect to λ_y, λ_z, λ_{yz} and θ to obtain the equilibrium state. This leads to nonlinear equations which can be solved numerically. We can also gain insight by simplifying the expressions for small voltages and retaining only the leading order terms in V. We obtain

$$\theta \approx \xi V, \tag{13}$$

$$\phi \approx \frac{r-1}{r}\,\xi V, \tag{14}$$

$$\lambda_y \approx 1 + \left(\frac{1}{2(3+b)} \left(\frac{b+2}{2} \left(\frac{1}{V_d^2} + \xi \left(c - \frac{1}{r}\xi \right) \right) - \frac{2}{r}\xi^2 \right) + \frac{1}{2}\xi^2 \right) V^2, \tag{15}$$

$$\lambda_z \approx 1 + \left(\frac{1}{2(3+b)} \left(\frac{1}{V_d^2} + \xi \left(c + \frac{3}{r}\xi \right) \right) - \frac{1}{2}\xi^2 \right) V^2, \tag{16}$$

$$\mathbf{p} \cdot \hat{\mathbf{X}} = \left(\frac{1}{V_p^2} + c\,\xi \right) V, \tag{17}$$

where $\xi = \frac{c}{a}$ is the initial slope between the tilt angle and the applied voltage. This quantity is related to the electroclinic coefficient $\alpha = \frac{\partial \theta}{\partial E}$. Specifically, in the examined parallel plate geometry $\xi \approx \frac{\alpha}{t_0}$. Subjected to relatively low voltages, Köhler *et al.* [21], Spillmann, Ratna, and Naciri [4] and Hiraoka *et al.* [6] measured an electroclinic coefficient $\alpha = 0.045\,\frac{\mathrm{m}}{\mathrm{MV}}$, $\alpha = 0.006\,\frac{\mathrm{m}}{\mathrm{MV}}$ and $\alpha = 0.07\,\frac{\mathrm{m}}{\mathrm{MV}}$, respectively. These findings correspond to tilt angles in the range $4^\circ - 10^\circ$. Since the thickness of the Smectic-A elastomers is around $t_0 \sim 0.01 - 1\,\mathrm{mm}$, we approximate the order of magnitude of the initial slope $\xi \sim 0.01 - 1\,\frac{1}{\mathrm{kV}}$.

The parameters b and c can be determined from a curve-fit of λ_y and λ_z as a function of the voltage via Eqs. (15) and (16) in the low voltage regime. The quadratic dependency of λ_y and λ_z on the applied voltage stems from the initial micro-structural alignment of the director and the layer normal in the Smectic-A elastomers.

Fig. (2a) and (2b) depict the shear angle ϕ and the tilt angle θ as a function of the applied voltage. The insets plot the electro-mechanical response of the Smectic-A elastomers under high voltages. The continuous and the dashed curves correspond to the exact predictions and the approximations (Eqs. (13) and (14)), respectively. A close inspection of the insets shows that the approximations hold for voltages up to $V \approx 5\,\mathrm{KV}$. It is noted that the application of the electric field leads to a tilt angle that is larger that the shear angle.

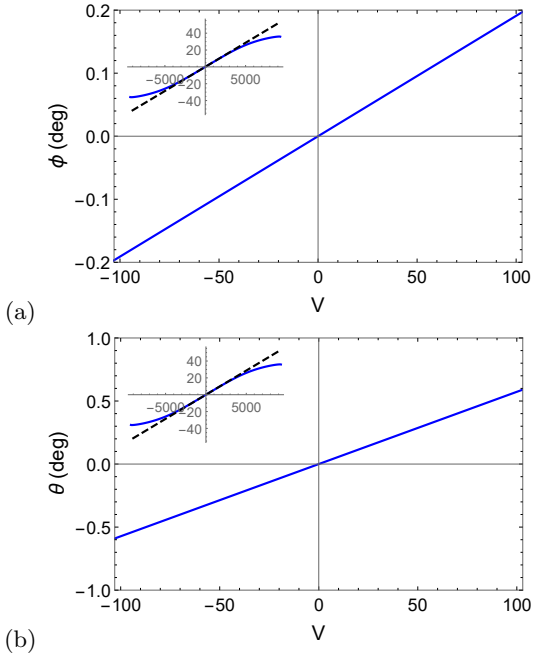

FIG. 2. (a) The shear angle ϕ and (b) the tilt angle θ as a function of the applied voltage V. The inset plots depict the predicted response subjected to high voltages. The continuous and the dashed curves correspond to the exact and the approximated predictions, respectively.

To explain this, note that Eqs. (13) and (14) reveal that in the range of small voltages the ratio between the shear and the tilt angles is $\frac{\phi}{\theta} = \frac{r-1}{r}$. Interestingly, in Smectic-A elastomers with $r > 1$ the ratio $\frac{\phi}{\theta} < 1$. This implies that the electrically induced deformation is characterized by a tilt angle θ that is greater than the shear angle ϕ. This observation is supported by the experimental findings of Hiraoka *et al.* [6], where the measured ratio $\frac{\phi}{\theta} \approx 0.02$ led to the anisotropy parameter $r = 1.02$. It should also be noted that the obtained value of the anisotropy parameter is very low.

The predicted polarization is plotted as a function of the voltage in Fig. (3). As before, the inset plots the predicted polarization as a result of high voltages. The continuous and the dashed curves correspond to the polarization with $c = 10^{-4}\,\frac{1}{\mathrm{V}}$ and $c = 0$, respectively. It is shown that the electrically induced tilt leads to an enhancement of the polarization of the elastomer by as much as 1.8 times. We also find that reversing the electric field flips the direction of the polarization, as seen in the experimental findings of Hiraoka *et al.* [6]. As previously discussed, this effect emanates from the chirality of the mesogens. We emphasize that since the direction of the chirality parameter $\mathbf{q}$ is linearly coupled to the polariza-

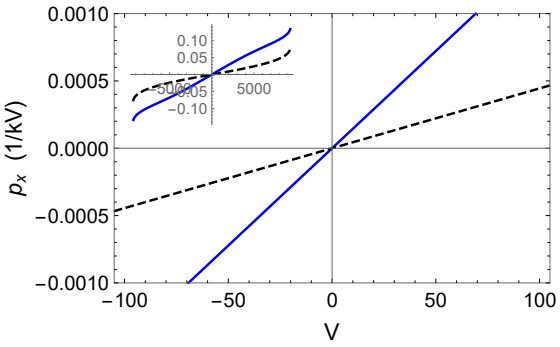

FIG. 3. $\mathbf{p}\cdot\hat{\mathbf{X}}$ as a function of the applied voltage V. The inset depicts the predicted response as a result of high voltages. The continuous and the dashed curves correspond to the polarization (Eq. (12)) with $c = 10^{-4}\,\frac{1}{\mathrm{V}}$ and $c = 0$, respectively.

tion, the spontaneous dipoles in a Smectic-A elastomer with $\mathbf{q} \to -\mathbf{q}$ decrease the polarization.

B. Deformation induced polarization

Next, we follow the experiment of Kramer and Finkelmann [16] and the theoretical work of Stenull *et al.* [13] and apply a shear stress s along the $\hat{\mathbf{Y}}$ direction which deforms the elastomer such that $\lambda_y = \lambda_z = 1$, corresponding to simple shear. By setting $V = 0$, the normalized energy density is

$$w = w\left(\lambda_y = 1, \lambda_z = 1, \lambda_{yz}, \theta\right). \tag{18}$$

To determine the equilibrium state, we minimize Eq. (18) with respect to the shear λ_{yz} and the tilt angle θ.

Figs. (4a) and (4b) depict the shear angle ϕ and the tilt angle θ as a function of s, respectively. We emphasize that the non-linear dependence of the shear angle ϕ on the shear force s corresponds to a linear dependence between the shear λ_{yz} and s. It is demonstrated that the mesogens tilt in the direction of the external shear force, as expected, and negating the force simply flips the direction of the tilt angle θ.

Interestingly, as opposed to the previous case the proposed model predicts that under the examined deformation the shear angle is always greater than the tilt angle. Furthermore, since $\lambda_z = 1$ is fixed and layer rotation is not permitted, the tilt of the mesogens is not accompanied by a reduction in the layer thickness. These observations are in accord with the experimental work of Kramer and Finkelmann [16], where the elastomer was subjected to a pure shear deformation such that the maximum imposed shear angle was $\phi = 21°$.

An examination of Fig. (4b) reveals that the dependence between the tilt of the mesogens and the applied shear force can be divided into two regimes. Initially, a small increase in the applied force leads to a large variation in the tilt angle. Beyond a threshold $s \approx 5$, the

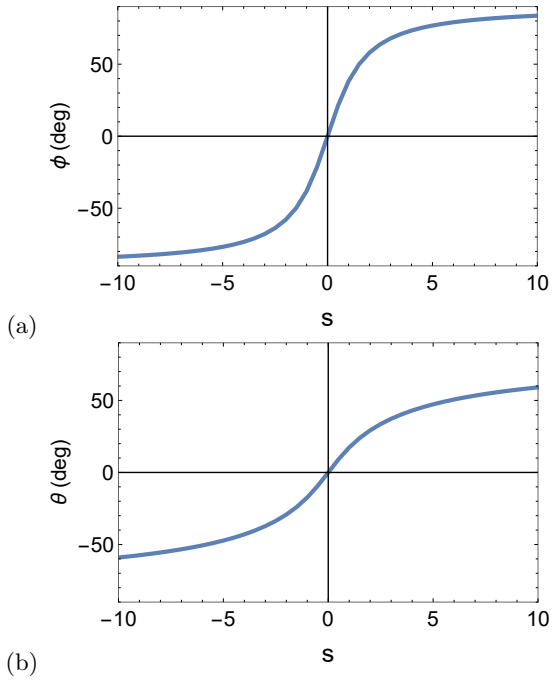

FIG. 4. (a) The shear angle ϕ and (b) the tilt angle θ as a function of the normalized nominal stress s.

cost of further tilting the director becomes very high. Specifically, a large increase in the shear force is required to attain a small increment in the tilt of the mesogens. Practically, elastomers fail at a shear angle $\phi \ll 90°$ and may not reach the second regime.

From Eq. (7) we deduce that the polarization in the absence of an electric field is directed along the chirality parameter $\mathbf{q}$, which is perpendicular to the plane spanned by $\hat{\mathbf{n}}$ and $\hat{\mathbf{k}}$. Fig. (5) plots the normalized polarization component $\mathbf{p}\cdot\hat{\mathbf{X}}$ as a function of s. It is demonstrated that the direction of the applied shear force determines the direction of the polarization. Two reasons lead to this behavior - the first is the anti-symmetric dependency of the tilt angle on the applied force. As discussed in relation to Fig. (4b), the directions of the shear force and the tilt are linearly dependent. Second, the observed trend is enforced by the direct coupling between the director and the chirality parameter $\mathbf{q}$. This coupling is defined such that the magnitude of the polarization is proportional to $\sin(\theta)$. We emphasize that the chirality of the mesogens determines the direction of the polarization. Specifically, a shear force along the $\hat{\mathbf{Y}}$-direction induces a polarization along the $-\hat{\mathbf{X}}$ direction in elastomers characterized by a flipped chirality vector (i.e. $\mathbf{q} \to -\mathbf{q}$).

IV. THE ELECTRICALLY INDUCED DEFORMATION OF A CYLINDER WITH HELICAL LAYERS

To illustrate the capabilities of the electroclinic effect, we explore the electro-mechanical response of a cylindri-

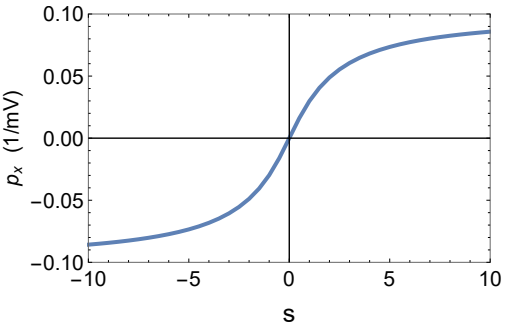

FIG. 5. $\mathbf{p}\cdot\hat{\mathbf{X}}$ as a function of the normalized nominal stress s.

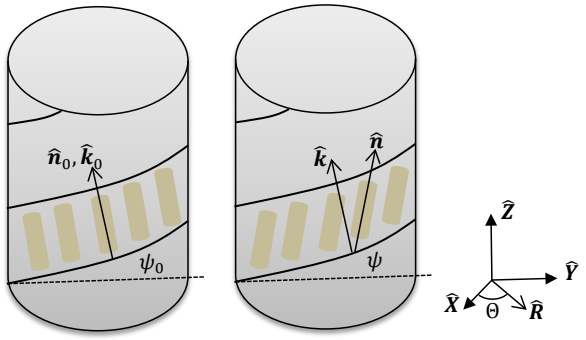

FIG. 6. A schematic of the electro-mechanical response of a cylinder with helical layers.

cal Smectic-A elastomer with helical layers. Consider a hollow thin-walled cylinder with an initial radius R, a wall thickness t_0 and a height H. The cylinder is made of a thin incompressible smectic-A elastomer film with helical layers. We prescribe a global cylindrical coordinate system $\left\{\hat{\mathbf{R}}, \hat{\mathbf{\Theta}}, \hat{\mathbf{Z}}\right\}$ such that at the reference configuration the director and the layer normal are

$$\hat{\mathbf{n}}_0 = \hat{\mathbf{k}}_0 = \sin(\psi_0)\,\hat{\mathbf{\Theta}} + \cos(\psi_0)\,\hat{\mathbf{Z}}, \tag{19}$$

where ψ_0 is the initial helix angle (see Fig. (6)). The position of the referential material points are denoted by $\{R, \Theta, Z\}$. The inner and outer faces of the cylindrical film are covered with flexible electrodes such that an applied voltage induces a radial electric field $\mathbf{E} \approx \frac{V}{t_0}\hat{\mathbf{R}}$. In addition, an internal pressure p is exerted on the inner surface. As a result, the cylinder deforms such that at the current deformation,

$$r = \sqrt{\lambda R^2 + \beta},\quad \theta = \Theta + \delta Z,\quad z = \frac{1}{\lambda} Z, \tag{20}$$

where $\{r, \theta, z\}$ denote the material points at the current configuration. Accordingly, the deformation gradient in cylindrical coordinates is

$$\mathbf{F} = \begin{pmatrix} \frac{\partial r}{\partial R} & 0 & 0 \\ 0 & \frac{r}{R}\frac{\partial \theta}{\partial \Theta} & r\frac{\partial \theta}{\partial Z} \\ 0 & 0 & \frac{\partial z}{\partial Z} \end{pmatrix} = \begin{pmatrix} \lambda\frac{R}{r} & 0 & 0 \\ 0 & \frac{r}{R} & \delta r \\ 0 & 0 & \frac{1}{\lambda} \end{pmatrix}. \tag{21}$$

At the current configuration, the chirality vector is

$$\mathbf{q} = \sin(\varphi)\,\hat{\mathbf{R}}, \tag{22}$$

and the director is

$$\hat{\mathbf{n}} = \sin(\psi + \varphi)\,\hat{\mathbf{\Theta}} + \cos(\psi + \varphi)\,\hat{\mathbf{Z}}, \tag{23}$$

where φ is the angle between the director $\hat{\mathbf{n}}$ and the layer normal $\hat{\mathbf{k}}$ and ψ is the helix angle at the current configuration. By employing Eq. (1), we find that

$$\hat{\mathbf{k}} = \sin(\psi)\,\hat{\mathbf{\Theta}} + \cos(\psi)\,\hat{\mathbf{Z}}. \tag{24}$$

The layer normal at the current configuration is directly dependent on the deformation gradient via 10, 12

$$\hat{\mathbf{k}} = \frac{\mathbf{F}^{-T}\hat{\mathbf{k}}_0}{\left|\mathbf{F}^{-T}\hat{\mathbf{k}}_0\right|}, \tag{25}$$

and therefore one finds that the helix angle at the current configuration is

$$\psi = \arctan\left(\frac{R\,\sin(\psi_0)}{\lambda r\,(\cos(\psi_0) - R\,\delta\,\sin(\psi_0))}\right). \tag{26}$$

The energy density function can be written as

$$w^{(h)}(\lambda, \beta, \varphi, \delta) = w(\lambda, \beta, \varphi, \delta) - \frac{2}{\mu}\,p\left(\frac{r}{R}\right)^2\frac{1}{\lambda}, \tag{27}$$

where $w(\lambda, \beta, \varphi, \delta)$ is computed with Eq. (9), p is the pressure inside the cylinder and $\left(\frac{r}{R}\right)^2\frac{1}{\lambda}$ accounts for the change in the enclosed volume of the cylinder. The electro-mechanical response of the cylinder is determined by minimizing the energy $w^{(h)}(\lambda, \beta, \varphi, \delta)$ with respect to λ, β, φ and δ.

Figs. (7a) and (7b) depict the twist δ and the change in the enclosed volume $\frac{v_t}{V_t} = \frac{\pi r^2 h}{\pi R^2 H}$, where h is the current height of the cylinder, as a function of the applied voltage, respectively, for various initial helix angles at $p = 0$. It is shown that elastomers with cylindrical layers (or helical layers with an angle $\psi_0 = 0°$) experience twist with a minor increase in the enclosed volume. As the helix angle increases from $\psi_0 = 0°$ to $\psi_0 = 45°$, the twist δ decreases while the enclosed volume increases. Interestingly, the twist angle in the Smectic-A elastomers with a helix angle $\psi_0 = 45°$ is extremely small. The proposed model also predicts that such an elastomer experiences the largest volumetric expansion. The trend is reversed as the helix angle increases from $\psi_0 = 45°$ to $\psi_0 = 90°$. Specifically, the cylinder twists in the other direction and the enclosed volume decreases.

We highlight that the volumetric expansion of the Smectic-A elastomers is due to the radial expansion and the longitudinal compression resulting from the amplification of the voltage. Additionally, it is important to note that the due to the low voltages that are practically applied in the actuation of these materials, the twist and the volumetric expansions are small.

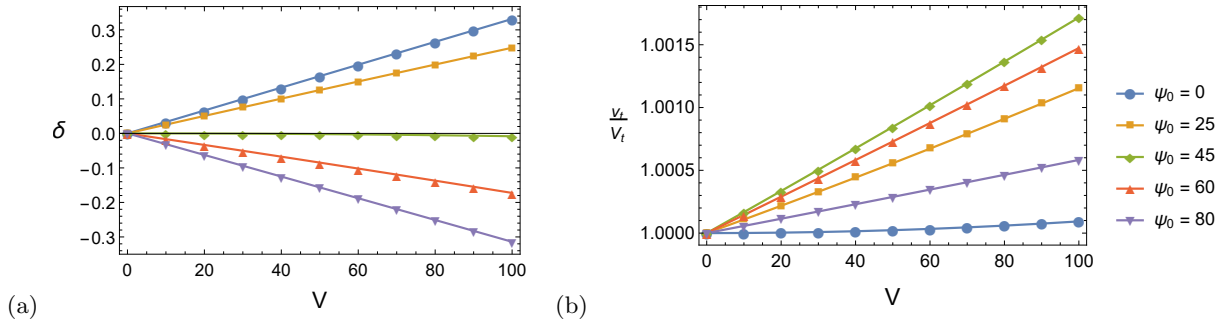

FIG. 7. (a) The twist δ and (b) the change in the enclosed volume $\frac{v_t}{V_t}$ as a function of the applied voltage for various helix angles at $p = 0$.

Next, we study the effects of pressure on the actuation of Smectic-A elastomers with helical layers at an angle $\psi_0 = 45°$. Figs. (8a) and (8b) plot the twist δ and the change in the enclosed volume $\frac{v_t}{V_t}$ as a function of the applied voltage, respectively, for a cylinder subjected to various pressures. Fig. (8a) reveals that the pressure causes the elastomer to twist. However, the increase in voltage does not significantly change the twist angle.

As shown in Fig. (8b), the enclosed volume increases with pressure. This stems from the radial expansion and the longitudinal elongation of the cylinder in response to the internal pressure. As the voltage is amplified, a linear increase in the enclosed volume is observed. Interestingly, we find that the linear dependence between $\frac{v_t}{V_t}$ and V is independent of the pressure in the examined range of voltages. It is again noted that even under an applied pressure, an increase in voltage leads to a radial expansion and a longitudinal compression.

V. CONCLUDING REMARKS

This work proposes a new model that captures the behavior of Smectic-A elastomers subjected to an electromechanical loading. Specifically, the energetic formulation of Adams *et al.* [12] is rewritten in terms of a new variable $\mathbf{q}$, that describes the chirality of the mesogens composing the elastomer, and an additional term is added to account for the effects of an electric stimulus. The current configuration of the elastomer is determined by minimizing the energy.

To demonstrate the merit of the model, two simple cases are presented. In the first case, an electric field is applied to the Smectic-A elastomer. In accordance with experimental findings, the proposed model predicts a shear deformation of the sample. In the second case, a mechanical shear deformation is imposed. As a consequence, the mesogens tilt and their inherent chirality gives rise to a polarization. The predictions qualitatively capture several experimental observations.

The potential of these elastomers is exhibited in Sec. IV, where a thin-wall cylindrical configuration of a Smectic-A film with helical layers is considered. Upon application of an electric field, we show that the pitch and the helix angle control the macroscopic behavior. Specifically, the enclosed volume and the twist depend on the formation of the lamellar structure. It is also shown that the application of internal pressure in the tube can effectively change the macroscopic response.

ACKNOWLEDGMENTS

This work was supported by MURI grant FA9550-16-1-0566 from the Air Force Office of Scientific Research (AFOSR).

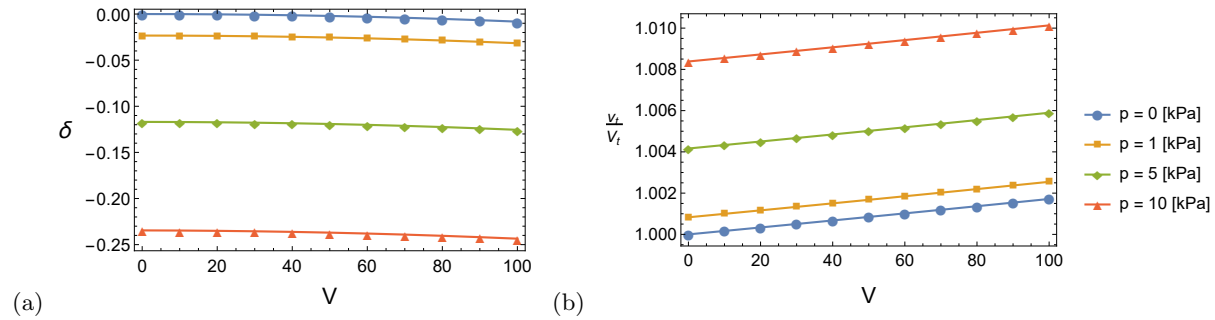

FIG. 8. (a) The twist δ and (b) the change in the enclosed volume $\frac{v_t}{V_t}$ as a function of the applied voltage for a cylinder comprising helical layers with a $\psi_0 = 45^\circ$ angle for various pressures.

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