Soft elasticity in smectic elastomers

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We investigate the soft elastic modes of smectic elastomers, that is shape change without energy cost. We use a microscopic model for their non-linear elasticity, similar those used for nematic elastomers. We consider two different phases of smectic elastomer; the biaxial smectic A for a simple illustration, and smectic C phases which are of great practical significance. We show that only one non-trivial trajectory of the director gives soft deformations. We give a geometrical interpretation of this soft elastic mode and give an explicit example.

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I.   INTRODUCTION

Soft modes in nematic elastomers are understood well theoretically from both symmetry arguments [1] and from microscopic models [2]. Regardless of the model chosen, the underlying symmetries of a nematic elastomer that must be present in any model result in the occurrence of soft modes [3]. These soft modes have also been investigated experimentally in both monodomain and polydomain nematic elastomers [4]. Quantitative, but not qualitative, deviations from characteristically soft deformations occur if the elastomer is only semi-soft, that is if it deviates from ideal symmetry requirements [5].

The elastic properties of monodomain smectic A (SmA) elastomers, synthesized by loading a swollen sample in the isotropic state and deswelling it into the smectic state, do not show any sign of soft elasticity. The smectic C (SmC) phase has also been synthesized experimentally and been formed into monodomains [6]. This phase is significant technologically because of its ferroelectric properties [7, 8]. Typically during soft deformation, shape changes accompany director rotation. Once the director is anchored perpendicular to the layers and loses its freedom to move relative to the solid matrix, softness is lost. In the SmC phase the director has sufficient additional freedom to be soft because the constraint on the layer thickness does not totally determine the orientation of the polymer shape tensor. Biaxial SmA is a conceptually simpler phase and also has sufficient freedom, but is of more limited experimental interest. Models of these two phases have been studied by different methods of non-linear elasticity from ours, but for small strains, and shown to be soft [9]. We aim here to give a geometrical interpretation of these soft modes by calculating the required deformation tensor for monodomains of these two phases of smectic elastomers. A simple illustration is first given for biaxial SmA, but our main aim is SmC elastomers. In section II, we summarise the model that will be used and introduce soft elasticity. We then calculate the soft modes of biaxial SmA elastomers in section III and finally we calculate the soft modes of an SmC elastomer in section IV. Our main conclusion is that as a result of the layer spacing constraint, the soft modes of a smectic elastomer are defined by only one parameter (excluding an arbitrary rotation).

II.   SMECTIC ELASTOMER MODEL

The models of biaxial SmA and SmC elastomers we will use here are extensions of the SmA elastomer model derived in [10] which we now summarise.

The end-to-end span of the polymer chains that make up the elastomer are modelled by an anisotropic Gaussian distribution, as in nematic elastomers, which obtains when chains are sufficiently long and which ensures that the matrix formed on crosslinking is rubbery and highly extensible. Additionally, in smectics the effect of the layers is to create a periodic potential in which biases where the ends of the polymer chains, and thus the crosslinks, sit. As a result of this potential the crosslink point distribution in the elastomer is only homogeneous within the plane of the layers. Parallel to the layer normal the distribution is periodic, as crosslink points sink into potential wells. This picture of smectic elastomers results in a strong coupling of the matrix to the layers. Consequently, SmA elastomers have a modulus comparable to that penalising the change of smectic layer spacing for any shape changes of the rubber matrix which convect layers to new spacings. This is a dramatic effect – smectic elastomers are rubbery in two dimensions and solid-like in the third; at larger strains they then suffer rotational instabilities. This reduces the cost of distortions involving layer spacing change to those involving the lower cost of rubbery distortion. In SmAs there is close agreement between experiment [11] and theory [10].

We shall need the tensor $\underline{\underline{\ell}}_0$ of effective step lengths of the polymer at the instant of crosslinking. It is defined by the second moment that specifies entirely the Gaussian distribution of chains at that time:

$$\langle R^0_\alpha R^0_\beta \rangle_{P_0} = \tfrac{1}{3} L \ell_{0\alpha\beta} . \tag{1}$$

In the uniaxial case in the principal frame there are two step lengths, $\ell_\parallel$ parallel to the (unit) director $\mathbf{n}$ specifying the direction of order, and $\ell_\perp$ perpendicular to $\mathbf{n}$. Note that on undergoing a phase change from the nematic to

a smectic state, the degree of anisotropy of the polymer, $r = \ell_\parallel/\ell_\perp$, may be changed because of the additional penalty of the polymer chains for crossing the smectic layers. In the SmA phase this effect can be described by decreasing the value of r for prolate chains on transition to the smectic phase, this effect in SmC elastomers is more complicated, as will be discussed in section IV.

We follow similar microscopic models of nematic elastomers in assuming that the smectic elastomer is composed of crosslinked polymers with identical strand length L, and that the crosslink points deform affinely, that is according to

$$\mathbf{R} = \underline{\underline{\lambda}} \cdot \mathbf{R}_0. \tag{2}$$

This is approximately true for individual chains. It is correct on average and is exact in the limit of infinite crosslink point functionality. For finite functionalities the effect is small, a change in the prefactor of the free energy. The quenched average of the elastic free energy density over the end positions, $\mathbf{R}_{1,2}$, and over the layers that the crosslink points occupy (assuming a strong potential localising crosslinks to layers) can be performed as in [10]. The resulting free energy density has a term proportional to $\mathcal{V}^{2/3}$ (where $\mathcal{V}$ is the volume of the system) penalising translations of the layers relative to the rubber matrix. This divergent intensive term enforces the affine deformation of the smectic layers with the matrix (crosslink points):

$$\mathbf{q} = \underline{\underline{\lambda}}^{-T} \cdot \mathbf{q}_0 \tag{3}$$

where $\mathbf{q}_0$ is the initial wavevector, and $\mathbf{q}$ the current wavevector of the smectic layers, and $\underline{\underline{\lambda}}^{-T}$ denotes the inverse transpose of the deformation matrix. The affine deformation of layers Eq. (3) makes the layer spacing

$$\frac{d}{d_0} = \frac{1}{|\underline{\underline{\lambda}}^{-T} \cdot \mathbf{k}_0|}, \tag{4}$$

where $\mathbf{k}_0$ is the initial (unit) direction of the layer normal in the solid.

If the rubber-elastic free energy terms and the smectic layer modulus terms are collected together then the resulting free energy expression is

$$f = \tfrac{1}{2}\mu \text{Tr}\left[\underline{\underline{\lambda}} \cdot \underline{\underline{\ell}}_0 \cdot \underline{\underline{\lambda}}^T \cdot \underline{\underline{\ell}}^{-1}\right] + \tfrac{1}{2}B\left(\frac{d}{d_0} - 1\right)^2. \tag{5}$$

where B is the smectic modulus associated with stretching the layers, d is the current layer spacing and d_0 is the initial layer spacing.

Both terms in the free energy density are non-negative. If we are to find deformations of zero cost, then the second term must vanish, that is $d = d_0$ during deformations. This condition is easily expressed from Eq. (4) or from Eq. (3) with $|\mathbf{q}| = q_0$, that is one must take $\underline{\underline{\lambda}}$ obeying the constraint:

$$\mathbf{k}_0^T \cdot \underline{\underline{\lambda}}^{-1} \cdot \underline{\underline{\lambda}}^{-T} \cdot \mathbf{k}_0 = 1 . \tag{6}$$

The first, rubber-elastic term of Eq. (5) is identical to that of purely nematic elastomers and arises from distorting the chain distribution away from its optimal shape. The minimum energy density is $3\mu/2$ which arises when the $\underline{\underline{\lambda}}$ is such that the argument of the trace reduces to the unit tensor $\underline{\delta}$. Trivially this can be satisfied by no deformation and no director rotation, or by a director rotation and a body rotation that are the same. The general form of non-trivial, soft modes in nematic elastomers [3] is:

$$\underline{\underline{\lambda}} = \underline{\underline{\ell}}_{\mathbf{n}}^{1/2} \cdot \underline{\underline{W}} \cdot \underline{\underline{\ell}}_0^{-1/2}, \tag{7}$$

where $\underline{\underline{W}}$ is a general rotation matrix, $\underline{\underline{\ell}}_0$ is the initial anisotropy tensor and $\underline{\underline{\ell}}_{\mathbf{n}}$ is the current anisotropy tensor of the polymer chain distribution, dependent on director $\mathbf{n}$. One can readily check that the $\underline{\underline{\lambda}}$ of Eq. (6) renders the argument of the Trace in Eq. (5) equal to $\underline{\delta}$ and hence gives the minimal free energy.

The tensor $\underline{\underline{\ell}} = (\ell_\parallel - \ell_\perp)\mathbf{nn} + \underline{\delta} \to \ell_\perp[(r-1)\mathbf{nn} + \underline{\delta}]$ can have a factor of $\ell_\perp$ extracted which we henceforth ignore since $\underline{\underline{\ell}}$ always appears in expressions with an $\underline{\underline{\ell}}^{-1}$, and thus with an associated, cancelling $1/\ell_\perp$ factor, if the latter tensor is also taken in its reduced form. In such forms $\underline{\underline{\ell}}^{1/2} = (\sqrt{r}-1)\mathbf{nn}+\underline{\delta}$ and $\underline{\underline{\ell}}^{-1/2} = (1/\sqrt{r}-1)\mathbf{nn}+\underline{\delta}$. Expressions we later require, such as $\underline{\underline{\ell}}^{1/2} \cdot \mathbf{n} \to \sqrt{r}\mathbf{n}$, are then easy to evaluate.

Physically, soft modes arise when the distribution of chain shapes is rotated at constant shape (and thus constant entropy and constant nematic energy) while the body changes shape to accommodate the chains [12]. A cartoon, Fig. 1, shows the initial chain shape distribution, an intermediate shape where the director rotation is not yet complete, and the shape when the director has rotated by 90º to be along the direction of imposed stretch and shape change at constant energy has come to its end.
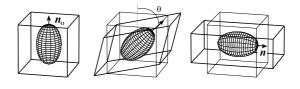

FIG. 1: Chain shape distribution is rotated by 90º from $\mathbf{n}_0$ to a perpendicular $\mathbf{n}$ with an intermediate state direction θ shown. The rubber deforms to accommodate the changing chain shape distribution without distorting it.

The general form Eq. (7) can be rewritten in terms of a particular vector $\mathbf{n}'$ such that $\mathbf{n} = \underline{\underline{W}} \cdot \mathbf{n}'$ by multiplying it by $\underline{\underline{W}} \cdot \underline{\underline{W}}^T$ from the left. The factor $\underline{\underline{W}}^T \cdot \underline{\underline{\ell}}_{\mathbf{n}}^{1/2} \cdot \underline{\underline{W}}$ becomes $\underline{\underline{\ell}}_{\mathbf{n}'}^{1/2}$ since the effect of the $\underline{\underline{W}}$ tensors is a rotation of the

tensor $\underline{\underline{\ell}}_{\mathbf{n}}^{1/2}$. Thus the soft mode (7) becomes:

$$\underline{\underline{\lambda}} = \underline{\underline{W}} \cdot \underline{\underline{\ell}}_{\mathbf{n}'}^{1/2} \cdot \underline{\underline{\ell}}_0^{-1/2} \ . \tag{8}$$

In rewriting the deformation in this way it is clear that the shape of the body after a soft deformation is determined by the choice of $\mathbf{n}'$ and is given by the term $\underline{\underline{\ell}}_{\mathbf{n}'}^{1/2} \cdot \underline{\underline{\ell}}_0^{-1/2}$ (which may contain an element of body rotation which we shall extract). It is followed by $\underline{\underline{W}}$ which is a pure body rotation and is of no elastic significance. We can choose the vector $\mathbf{n}'$ anywhere on the surface of a unit sphere. Consequently a nematic elastomer has several ways in which it can respond softly to an imposed deformation. In the case of a smectic elastomer however, there is considerably less freedom because of the layer spacing constraint.

III. BIAXIAL SMECTIC-A ELASTOMERS

Biaxial nematic phases, that is liquid crystalline phases where there is ordering along the director and perpendicular to it, are rare. Examples of both lyotropic and thermotropic biaxial nematic liquid crystals are known [13, 14]. Biaxial nematic phases have also been found in nematic polymers [15, 16] and in principle it should be possible to make a biaxial nematic liquid crystal elastomer. To our knowledge these have not yet been made, but as in [9] we calculate their soft elastic deformations as an illustration since they are so much simpler than SmC elastomers.

To model biaxial SmA elastomers we follow section II but include a biaxial polymer shape tensor, $\underline{\underline{\ell}}$, with principal axes $\mathbf{n}$, $\mathbf{m}$ and $\mathbf{l}$, and corresponding eigen values $\ell_\parallel$, ℓ_1 and ℓ_2. The shape tensor of a biaxial polymer is thus

$$\underline{\underline{\ell}} = \begin{pmatrix} \ell_1 & 0 & 0 \\ 0 & \ell_2 & 0 \\ 0 & 0 & \ell_\parallel \end{pmatrix} \tag{9}$$

In a biaxial SmA elastomer the layer normal $\mathbf{k}$ and the director $\mathbf{n}$ (the primary alignment direction) are identified. In the soft mode of Fig. 1, director rotation was about an axis perpendicular to $\mathbf{n}_0$. In SmA elastomers $\mathbf{n} \equiv \mathbf{k}$ is fixed along the layer normal and such a soft mode is now lost, as we prove below. However in a biaxial SmA, rotation about $\mathbf{n}$ is non-trivial. The secondary alignment axes are free to rotate in the plane of the layers. Since this section of $\underline{\underline{\ell}}$ is non-circular, $\ell_1 \neq \ell_2$, soft deformations in the perpendicular plane arise in complete analogy to Fig. 1 for nematics but in the plane perpendicular to $\mathbf{n}_0$, as we also prove below.

The layer spacing constraint Eq. (6) on identifying $\mathbf{k}$ and $\mathbf{n}$ becomes:

$$\mathbf{n}_0^T \cdot \underline{\underline{\lambda}}^{-1} \cdot \underline{\underline{\lambda}}^{-T} \cdot \mathbf{n}_0 = 1. \tag{10}$$

The general soft mode (7) in constraint (10) yields:

$$\frac{1}{r} = \mathbf{n}_0^T \cdot \underline{\underline{W}}^T \cdot \underline{\underline{\ell}}^{-1} \cdot \underline{\underline{W}} \cdot \mathbf{n}_0. \tag{11}$$

Deriving Eq. (11) from Eq. (10) requires expressions discussed after Eq. (7). The only solution to this equation is $\mathbf{n} = \mathbf{n}' \equiv \underline{\underline{W}} \cdot \mathbf{n}_0$ because the quadric surface associated with $\underline{\underline{\ell}}^{-1}$ only has the correct width at one point (the maximum or minimum width depending on whether $r < 1$ or $r > 1$). Returning this $\underline{\underline{W}}$ to Eq. (7) gives, on using the forms for $\underline{\underline{\ell}}^{1/2}$ and $\underline{\underline{\ell}}_0^{-1/2}$, the result $\underline{\underline{\lambda}} = \underline{\underline{W}}$, that is a trivial body rotation by the same amount as the director has been rotated. There has been no shape change of the body. Thus there is indeed no freedom for the primary alignment director to rotate with respect to the matrix, and hence no soft modes in uniaxial SmA elastomers except for pure rotations. This is because if the layer normal $\mathbf{n}$ is moved by any deformation other than a rotation then the layer spacing will be changed and the resulting state will be higher in energy. However, in biaxial SmA elastomers the secondary alignment axes are unconstrained, thus there is still enough freedom for a soft mode to exist, that is a manifold of solutions of Eq. (11) not involving rotation of $\mathbf{n}$.

The soft modes can be decomposed into their component rotations to gain a better understanding of them as follows. First we write the general deformation matrix as:

$$\underline{\underline{W}} = \underline{\underline{W}}_{\mathbf{R}} \cdot \underline{\underline{W}}_{\mathbf{n}_0}, \tag{12}$$

where $\underline{\underline{W}}_{\mathbf{n}_0}$ is a rotation about $\mathbf{n}_0$ and $\mathbf{n} = \underline{\underline{W}}_{\mathbf{R}} \cdot \mathbf{n}_0$. Using this in Eq. (7) results in the following expression for the soft mode:

$$\underline{\underline{\lambda}} = \underline{\underline{\ell}}^{1/2} \cdot \underline{\underline{W}}_{\mathbf{R}} \cdot \underline{\underline{W}}_{\mathbf{n}_0} \cdot \underline{\underline{\ell}}_0^{-1/2} \tag{13}$$

$$= \underline{\underline{W}}_{\mathbf{R}} \cdot \underline{\underline{W}}_{\mathbf{n}_0} \cdot \left[\underline{\underline{\ell}}_{\mathbf{n}_0, \mathbf{m}', \mathbf{l}'}^{1/2} \cdot \underline{\underline{\ell}}_0^{-1/2} \right] \ , \tag{14}$$

the second form arising from the same arguments that led to Eq. (8) from Eq. (7). The factor in square brackets here is the soft mode in which the secondary alignment axes rotate around but the primary axis remains fixed. It is given by:

$$\begin{pmatrix} 1 - (1 - 1/\sqrt{r_\perp}) \sin^2 \phi & (1 - \sqrt{r_\perp}) \sin \phi \cos \phi & 0 \\ (1/\sqrt{r_\perp} - 1) \sin \phi \cos \phi & 1 + (\sqrt{r_\perp} - 1) \sin^2 \phi & 0 \\ 0 & 0 & 1 \end{pmatrix}$$

where ϕ is the angle that the secondary alignment axis has rotated through and $r_\perp = \ell_1/\ell_2$ is a material parameter expressing the degree of biaxiality in the chain anisotropy. The $\underline{\underline{W}}_{\mathbf{R}} \cdot \underline{\underline{W}}_{\mathbf{n}_0}$ that follows this deformation is a trivial body rotation which we can neglect. Thus all soft modes in biaxial SmA elastomers can be decomposed into a rotation of the soft mode that has the primary alignment axis fixed and a secondary rotation axis displaced from its initial position.

A.   Example: Imposed λ_{yy}

For later comparison with the soft modes of a SmC elastomer, we present the soft mode associated with an imposed λ_{yy}. This mode has a fixed primary alignment direction, $\mathbf{n} = \mathbf{n}_0$, but a mobile secondary alignment direction, $\mathbf{m}$; see Fig. 2. The centre of the diagram depicts the secondary alignment axis, $\mathbf{m}$. Along the outside of the diagram the shape of the biaxial SmA elastomer is viewed from along the layer normal. Note that the maximum extension in the y direction is by a factor of $\sqrt{r_\perp}$ and occurs when the secondary alignment axis has rotated by $\phi = 90°$.
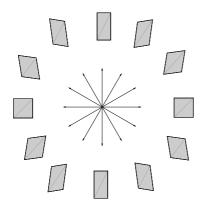

FIG. 2: The soft mode of a biaxial SmA elastomer of anisotropy $r_\perp = 2$, with the layer normal out of the page. The direction of the secondary director, $\mathbf{m}$, is shown in the centre. It rotates by $\phi = 30°$ from one image to the next.

IV.   SMECTIC C ELASTOMERS

For SmC elastomers we extend the SmA elastomer model presented in section II. Now the director is tilted with respect to the layer normal. We denote the layer normal by $\mathbf{k}$, the director by $\mathbf{n}$ and the direction in which the mesogens are tilted in by $\mathbf{c}$, all unit vectors (see Fig. 3). Thus if the director has a tilt angle of θ then:

$$\mathbf{n} = \mathbf{k}\cos\theta + \mathbf{c}\sin\theta . \qquad (15)$$

The constraint (6) on the layer spacing (4) must still be obeyed, but this does not constrain the whole of the director now, giving scope for soft modes.

Here we use a uniaxial tensor to represent the polymer shape anisotropy for simplicity. However, in the SmC phase there are *two* distinguished directions, the layer normal and the director, which should alter the polymer shape anisotropy. Thus it is anticipated that the shape tensor should really be biaxial. However we shall find
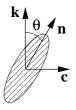

FIG. 3: In an SmC elastomer the director, $\mathbf{n}$, is tilted with respect to the layer normal, $\mathbf{k}$ by an angle θ.

that novel soft elasticity already arises because of the biaxial arrangement of a uniaxial $\underline{\underline{\ell}}$ about the layer normal. The same qualitative behaviour is expected for a truly biaxial $\underline{\underline{\ell}}$ as that generated below by the simplification we follow, that is of a uniaxial $\underline{\underline{\ell}}$ constrained to be tilted at a fixed angle to the layer normal.

Another term in this model penalising the tilt of the director away from making an angle θ with the layer normal could be included. However, since the main concern here is soft modes, deviation from tilt θ with any energy cost would remove softness during deformation. Consequently, this term is not included here and, as with the layer spacing, the tilt angle is simply regarded as being fixed. In any event, away from the SmA $\rightarrow$ SmC transition it is expected that the modulus associated with changes in θ is very high.

A.   General form of soft modes

Our model of SmC elastomers has underpinning nematic rubber elasticity with constraints of constancy of layer spacing. We again start with general soft modes of nematic elastomers Eq. (7), with the constraint on $\underline{\lambda}$ Eq. (6). We introduce a new auxiliary vector, $\mathbf{w}_0 = \underline{\underline{\ell}}_0^{1/2} \cdot \mathbf{k}_0$, much as we introduced the auxiliary $\mathbf{n}'$ to the director $\mathbf{n}$. With the help of $\underline{\underline{\ell}}_0^{1/2} = (\sqrt{r} - 1)\mathbf{n}_0\mathbf{n}_0^T + \underline{\underline{\delta}}$ one has:

$$\mathbf{w}_0 = \underline{\underline{\ell}}_0^{1/2} \cdot \mathbf{k}_0 = \mathbf{k}_0 + (\sqrt{r} - 1)\cos\theta\,\mathbf{n}_0 . \qquad (16)$$

Thus $\mathbf{n}_0$, $\mathbf{k}_0$ and $\mathbf{w}_0$ lie in the same plane, with $\mathbf{w}_0$ between $\mathbf{n}_0$ and $\mathbf{k}_0$ for $r > 1$. The modulus of $\mathbf{w}_0$ is not unity but rather $w_0^2 = 1 + (r-1)\cos^2\theta$.

Inserting the general soft mode (7) into the layer spacing constraint, Eq. (6), and using $\mathbf{w}_0$, one obtains:

$$\mathbf{w}_0^T \cdot \underline{\underline{W}}^T \cdot \underline{\underline{\ell}}^{-1} \cdot \underline{\underline{W}} \cdot \mathbf{w}_0 = 1 \qquad (17)$$

The tensor $\underline{\underline{\ell}}^{-1} = (\frac{1}{r} - 1)\mathbf{n}\mathbf{n}^T + \underline{\underline{\delta}}$ can be injected into Eq. (17), whence $1 = (\frac{1}{r}-1)[\mathbf{n}^T \cdot \underline{\underline{W}} \cdot \mathbf{w}_0]^2 + \mathbf{w}_0^T \cdot \mathbf{w}_0$. Using $w_0^2 = 1 + (r-1)\cos^2\theta$, this constraint can be rearranged into the more transparent form:

$$(\underline{\underline{W}} \cdot \mathbf{w}_0)^T \cdot \mathbf{n} = \pm\sqrt{r}\cos\theta. \tag{18}$$

That is, (18) is the equation of two planes that are a distance: $\pm 1/\left(1 + \frac{1}{r}\tan^2\theta\right)^{1/2}$ from the origin with normal vector along the $\underline{\underline{W}} \cdot \mathbf{w}_0$ direction. Additionally the director is a unit vector and thus the allowed directors $\mathbf{n}$ after distortion sit on the intersection of the planes with the unit sphere $\mathbf{n} \cdot \mathbf{n} = 1$. These circles of intersection are always guaranteed since the distance of the planes from the origin is less than unity.

The constraint of constant layer spacing having been implemented, we now need to ensure that the direction of the layer normal changes affinely with the matrix, Eq. (3). The new layer normal follows from

$$\mathbf{k} = \frac{\underline{\lambda}^{-T} \cdot \mathbf{k}_0}{|\underline{\lambda}^{-T} \cdot \mathbf{k}_0|} \rightarrow \underline{\lambda}^{-T} \cdot \mathbf{k}_0 \tag{19}$$

which is a normalised form of the transformed layer wavevector, Eq. (3), and simplifies as shown if the layer spacing is fixed, that is $d/d_0 = 1$ in Eq. (4) which means equally that Eq. (6) is satisfied. When considering a particular soft deformation constructed by choosing a particular $\underline{\underline{W}}$ in Eq. (7), the above determines the choice of $\underline{\underline{W}}$ if we want to achieve a desired $\mathbf{n}$ and $\mathbf{k}$:

We insert a soft $\underline{\lambda}^{-T}$ into this expression for the new $\mathbf{k}$. It is $\underline{\lambda}^{-T} = \underline{\underline{\ell}}^{-1/2} \cdot \underline{\underline{W}} \cdot \underline{\underline{\ell}}_0^{1/2}$, on inverting and transposing (7), whereupon the normal becomes $\mathbf{k} = \underline{\underline{\ell}}^{-1/2} \cdot \underline{\underline{W}} \cdot \mathbf{w}_0$. Multiplying both sides from the left by $\underline{\underline{\ell}}^{1/2}$, and using the explicit form for $\underline{\underline{\ell}}^{1/2}$ and recalling $\mathbf{n} \cdot \mathbf{k} = \cos\theta$, one obtains

$$\underline{\underline{W}} \cdot \mathbf{w}_0 = \mathbf{k} + (\sqrt{r} - 1)\cos\theta\,\mathbf{n}, \tag{20}$$

Thus the three vectors $\underline{\underline{W}} \cdot \mathbf{w}_0$, $\mathbf{k}$ and $\mathbf{n}$ obey a the relation of the same form as Eq. (16). This motivates the definition of a new auxiliary vector $\underline{\underline{W}} \cdot \mathbf{w}_0 = \mathbf{w}$.

The allowed soft modes correspond to the points where the two planes defined in Eq. (18) intersect a sphere. To analyse the soft modes first the case without $\underline{\underline{W}}$ is studied, followed by the more general case of those modes including a matrix $\underline{\underline{W}}$.

1. Geometrical interpretation of soft modes of the form $\underline{\underline{\ell}}_{\mathbf{n}}^{1/2} \cdot \underline{\underline{\ell}}_0^{-1/2}$

The algebraically simplest (but certainly not physically simplest) soft modes in an SmC elastomer are those without a $\underline{\underline{W}}$ matrix. In this case the final director must lie

on the intersection of the unit sphere and the planes of Eq. (18). An illustration of this is shown in Fig. 4. The figure shows the initial director $\mathbf{n}_0$ and the initial layer normal $\mathbf{k}_0$ and two circles on the surface of the sphere, corresponding to the final positions of the director and layer normal that obey the layer constraint. Note that
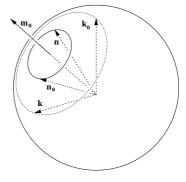

FIG. 4: An illustration of the director orientations that satisfy the layer constraint.

not all final positions of the director are possible with soft modes of this form. The appropriate $\underline{\underline{W}}$ must be included such that the required final director position still obeys the layer constraint.

2. Geometrical interpretation of general soft modes of the form $\underline{\underline{\ell}}_{\mathbf{n}}^{1/2} \cdot \underline{\underline{W}} \cdot \underline{\underline{\ell}}_0^{-1/2}$

It is convenient to break the $\underline{\underline{W}}$ down into two component rotations in order the exploit the simple case above, followed then by a body rotation. Thus if $\underline{\underline{W}} = \underline{\underline{W}}_{\mathbf{R}} \cdot \underline{\underline{W}}_{\mathbf{w}_0}(\xi)$, where the latter is a rotation by ξ about $\mathbf{w}_0$, we can write a general soft mode as:

$$\underline{\lambda} = \underline{\underline{\ell}}_{\mathbf{n}}^{1/2} \cdot \underline{\underline{W}}_{\mathbf{R}} \cdot \underline{\underline{W}}_{\mathbf{w}_0}(\xi) \cdot \underline{\underline{\ell}}_0^{-1/2}. \tag{21}$$

We multiply the right hand side from the left by unity in the form $\underline{\underline{W}}_{\mathbf{R}} \cdot \underline{\underline{W}}_{\mathbf{w}_0} \cdot \underline{\underline{W}}_{\mathbf{w}_0}^T \cdot \underline{\underline{W}}_{\mathbf{R}}^T$. Regrouping the $\underline{\underline{W}}$ terms rotates $\underline{\underline{\ell}}_{\mathbf{n}}^{1/2}$ to $\underline{\underline{\ell}}_{\mathbf{n}'}^{1/2} = \underline{\underline{W}}_{\mathbf{w}_0}^T \cdot \underline{\underline{W}}_{\mathbf{R}}^T \cdot \underline{\underline{\ell}}_{\mathbf{n}}^{1/2} \cdot \underline{\underline{W}}_{\mathbf{R}} \cdot \underline{\underline{W}}_{\mathbf{w}_0}$. Then (21) rearranges to:

$$\underline{\lambda} = \underline{\underline{W}}_{\mathbf{R}} \cdot \underline{\underline{W}}_{\mathbf{w}_0}(\xi) \cdot \underline{\underline{\ell}}_{\mathbf{n}'}^{1/2} \cdot \underline{\underline{\ell}}_0^{-1/2}. \tag{22}$$

Specifically one has applied the rotations $\underline{\underline{W}}_{\mathbf{R}}^T \cdot \mathbf{n} \rightarrow \mathbf{n}_0$ and $\underline{\underline{W}}_{\mathbf{w}_0}^T \cdot \mathbf{n}_0 \rightarrow \mathbf{n}'$. Thus the definitions of the rotations are $\underline{\underline{W}}_{\mathbf{w}_0}$ takes $\mathbf{n}'$ to $\mathbf{n}_0$ and $\underline{\underline{W}}_{\mathbf{R}}$ takes $\mathbf{n}_0$ to $\mathbf{n}$. The power of this method is that we can take any of the $\underline{\underline{\ell}}_{\mathbf{n}'}^{1/2} \cdot \underline{\underline{\ell}}_0^{-1/2}$ soft modes previously generated, see Fig. 5, to generate the shape change that is now defined by a single parameter, the rotation angle ξ. The rotation $\underline{\underline{W}}_{\mathbf{R}}$
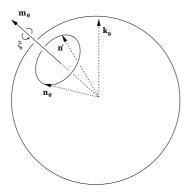
FIG. 5: The first stage of the calculation of a general soft mode: rotating the initial director $\mathbf{n}_0$ around to $\mathbf{n}'$.

that will determine the final director is not involved with this first stage. Then one can then independently select a general final director that is not confined to the circle about $\mathbf{w}_0$ by applying $\underline{W}_{\mathbf{R}}$ to $\mathbf{n}_0$, see Fig. 6. Having de-
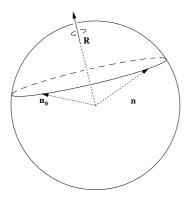

FIG. 6: The figure shows the second stage of the calculation of a soft mode: a general rotation about the axis $\mathbf{R}$.

cided where the final director is to point, one then applies the body rotation $\underline{W}_{\mathbf{R}} \cdot \underline{W}_{\mathbf{w}_0}(\xi)$ to the softly deformed sample to complete the deformation (22).

The element of body rotation of the soft mode can be separated out, leaving just a symmetric deformation, by using the polar decomposition theorem:

$$\underline{\lambda} = \underline{\ell}_{\mathbf{n}'}^{1/2} \cdot \underline{\ell}_0^{-1/2} = \underline{U} \cdot \underline{S}, \tag{23}$$

where $\underline{U}$ is a rotation matrix and $\underline{S}$ is a symmetric matrix. The rotation axis for this decomposition must be in the $\mathbf{n}' \times \mathbf{n}_0$ direction. We can use this information to construct $\underline{U}^T \cdot \underline{\lambda}$ and demand that it is symmetric to find $\underline{S}$. The resulting rotation angle is given by:

$$\tan \alpha = \frac{(1 - \sqrt{r})^2 (\mathbf{n}_0 \cdot \mathbf{n}') \sqrt{1 - (\mathbf{n}_0 \cdot \mathbf{n}')^2}}{(1 + r) - (\mathbf{n}_0 \cdot \mathbf{n}')^2 (1 - \sqrt{r})^2} \tag{24}$$

Now a particular example of Eq. (21) is considered. Here rigid clamping constraints are not included so there is no formation of microstructure.

B. Example: Imposed λ_{yy}

To illustrate the soft modes we could choose to impose an elongation in any direction, provided the director has scope to rotate into that direction and thereby to extend the sample. This excludes stretches parallel to the $\mathbf{n}_0$ direction. An elongation perpendicular to the layer normal is particularly simple because it does not induce the layer normal to rotate.

An extension in the $\mathbf{y}$ direction is imposed on an elastomer with its layer normal in the $\mathbf{z}$ direction and the in-plane component of the director in the $\mathbf{x}$ direction, i.e. $\mathbf{c} = \mathbf{x}$ initially. We take a deformation matrix is of the form:

$$\underline{\underline{\lambda}} = \begin{pmatrix} \lambda_{xx} & 0 & \lambda_{xz} \\ \lambda_{yx} & \lambda_{yy} & \lambda_{yz} \\ 0 & 0 & \lambda_{zz} \end{pmatrix}, \tag{25}$$

where the components λ_{xy} and λ_{zy} are not included. This is because these λs deform the sample by translating the y faces of the sample in the $\pm x$ and $\pm z$ directions. Any small y forces associated with the yy elongation would generate counter torques and quickly eliminate the λs. The λ_{zx} component is excluded because without compensating elongation in the $\mathbf{z}$ direction it would compress the layers, as well as rotating a component of the director perpendicular to the stretch direction.

The initial orientation of the layer normal and the director are given by:

$$\mathbf{n}_0 = (\sin \theta, 0, \cos \theta) \tag{26}$$
$$\mathbf{k}_0 = (0, 0, 1) \tag{27}$$

where θ is the tilt angle of the director (typically around $20°$). The current orientation of the director is assumed to be

$$\mathbf{n} = (\sin \theta \cos \phi, \sin \theta \sin \phi, \cos \theta), \tag{28}$$

and the layer normal, $\mathbf{k}$ unmoved. The layer normal ($\mathbf{k}_0 = \mathbf{z}$) cannot be moved by deformation tensors of the form Eq. (25) since it must be derived from the expression $\mathbf{k} = \underline{\lambda}^{-T} \cdot \mathbf{k}_0$. In addition, the only consistent rotation matrices $\underline{W}_{\mathbf{R}}$ that leave the layer normal unmoved, must have their rotation axis, $\mathbf{r}$, parallel to $\mathbf{k}_0$. This rotation must take $\mathbf{n}_0 \rightarrow \mathbf{n}$. Thus $\underline{W}_{\mathbf{R}}$ can be identified as a rotation of angle ϕ around an axis parallel to $\mathbf{k}_0$. Bearing this in mind, one constructs the tensor $\underline{\lambda} = \underline{W}_{\mathbf{R}} \cdot \underline{W}_{\mathbf{w}_0}(\xi) \cdot \underline{\ell}_{\mathbf{n}'}^{1/2} \cdot \underline{\ell}_0^{-1/2}$ (for the details of this see appendix). The only remaining variable is ξ, and this can

be determined by demanding that $\lambda_{xy} = 0$ in the above expression for $\underline{\lambda}$. This yields the following equation for ξ

$$0 = \cos\xi\sin\phi + \sqrt{\frac{1 + r + (r-1)\cos 2\theta}{2r}}\cos\phi\sin\xi. \quad (29)$$

Using this expression for ξ all the components of the deformation tensor can be obtained. Writing $\rho = \sin^2\theta + r\cos^2\theta$, and $a(\phi) = \sqrt{\cos^2\phi + \frac{\rho}{r}\sin^2\phi}$ we have the following matrix for $\underline{\underline{\lambda}}$

$$\begin{pmatrix} a(\phi) & 0 & \frac{(r-1)\sin 2\theta(-a(\phi)+\cos\phi)}{2\rho} \\ \frac{\left(1-\frac{\rho}{r}\right)\sin 2\phi}{2a(\phi)} & \frac{1}{a(\phi)} & \frac{(r-1)\sin 2\theta\left(-\frac{\left(1-\frac{\rho}{r}\right)\sin 2\phi}{2a(\phi)}+\sin\phi\right)}{2\rho} \\ 0 & 0 & 1 \end{pmatrix}.$$

This tensor is explicitly constructed to be a soft mode and evidently has $\det[\underline{\underline{\lambda}}] = 1$. To illustrate this mode Fig. 7 shows how this sample deforms for various different azimuthal angles, ϕ. The figure gives a view of a block of SmC rubber down the layer normal and should be compared with Fig. 2. Note that even after a rotation of the director of $\phi = \pi$ the rubber does not return to its original configuration. Because of the tilt of the director w.r.t. the layer normal a strain $\lambda_{xz} < 0$ is generated after $\phi \to \pi$ and this component has a $\cos\phi$ term. By contrast $\lambda_{xy} = \lambda_{zy} = 0$ and $\lambda_{yy} = 1$ at $\phi = \pi$, indeed λ_{zy} depends on 2ϕ. At the intermediate value of $\phi = \pi/2$ the elastomer has contracted along the direction of the original anisotropy tensor and so has developed a shear in both the λ_{xz} and λ_{yz} directions. The maximum extension in the y direction occurs at $\phi = \pi/2$, when the λ_{yy} component takes the value $\sqrt{r/(\sin^2\theta + r\cos^2\theta)}$. For the case with $\theta = 30°$ and $r = 2$ this gives a maximum extension of roughly 7%.
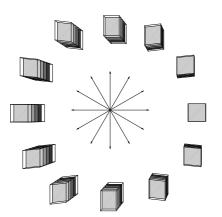

FIG. 7: An illustration of the soft mode of a SmC elastomer. In this case the layer normal remains out of the page and the **c** direction is shown in the centre of the diagram. A tilt angle of $\theta = 30°$ and an anisotropy of $r = 8$ were chosen.

V. CONCLUSIONS

We have presented a geometrical interpretation of the soft modes predicted in two phases of smectic elastomers. It was found that there is only one soft trajectory for the director, excluding body rotations, as a consequence of the restrictions imposed upon the elastomer by the fixed layer constraint. Specific examples of this mode were presented for a monodomains of biaxial SmA or SmC. They can respond softly to a single imposed component of the deformation tensor. An illustration of imposed λ_{xx} was given but in principle we could impose any other component, including λ_{zz}, through a combination of the single soft trajectory and a body rotation. The experimental boundary conditions of fixed ends near the clamps mean that any soft mode will be accompanied by microstructure, as is frequently the case in nematic elastomers.

APPENDIX: DETAILS OF THE DEFORMATION MATRIX

Here we give details of the calculation of the deformation tensor required for a soft mode when the λ_{yy} component is imposed. The following vectors are required:

$$\begin{align} \mathbf{k}_0 &= (0, 0, 1) & (A.1) \\ \mathbf{c}_0 &= (0, 1, 0) & (A.2) \\ \mathbf{w}_0 &= \underline{\underline{\ell}}_0^{1/2} \cdot \mathbf{k}_0 & (A.3) \\ \mathbf{c} &= (\cos\phi, \sin\phi, 0) & (A.4) \\ \mathbf{k} &= \mathbf{k}_0 & (A.5) \end{align}$$

From these vectors we can calculate the rotation matrices:

$$\underline{\underline{W}}_{\mathbf{R}} = \underline{\underline{\delta}}\cos\phi + (1-\cos\phi)\mathbf{k}_0\mathbf{k}_0^T + (\sin\phi)\mathbf{k}_0\wedge \quad (A.6)$$

$$\underline{\underline{W}}_{\mathbf{w}_0} = \underline{\underline{\delta}}\cos\xi + (1-\cos\xi)\mathbf{w}_0\mathbf{w}_0^T + (\sin\xi)\mathbf{w}_0\wedge \quad (A.7)$$

Using the expression given in Eq. (21) we obtain the following:

$$
\begin{pmatrix}
\cos\xi\cos\phi - \sqrt{\frac{\rho}{r}}\sin\xi\sin\phi & -\sqrt{\frac{r}{\rho}}\cos\phi\sin\xi - \cos\xi\sin\phi & \frac{(r-1)\sin 2\theta}{2\rho}\left(\cos\phi(1-\cos\xi) + \sqrt{\frac{\rho}{r}}\sin\xi\sin\phi\right) \\
\sqrt{\frac{\rho}{r}}\cos\phi\sin\xi + \cos\xi\sin\phi & \cos\xi\cos\phi - \sqrt{\frac{r}{\rho}}\sin\xi\sin\phi & \frac{(r-1)\sin 2\theta}{2\rho}\left(-\sqrt{\frac{\rho}{r}}\sin\xi\cos\phi + (1-\cos\xi)\sin\phi\right) \\
0 & 0 & 1
\end{pmatrix}
$$

To determine ξ we demand that the λ_{xy} component is zero and obtain the matrix given in the text.

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