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RAPID COMMUNICATION

A minimal model for the auxetic response of liquid crystal elastomers

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We develop a minimal phenomenological model to describe the auxetic response recently observed in liquid crystal elastomers, and further determine by theoretical calculation the critical condition required for the auxetic response to occur.

Keywords: auxetic response, liquid crystal elastomers, phenomenological model

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1. Introduction

Auxetic response is usually associated with materials with negative Poisson's ratios. The origin lies in the change of microscopic porous geometry of such materials when subjected to an external stress. Due to their abnormal mechanic properties, materials exhibiting auxeticity possess many potential applications in various fields, such as sports equipment, architecture, aerospace, and biomedical materials.[1–4] Recently, Mistry *et al.* reported that liquid crystal elastomers in certain circumstances can also exhibit auxetic response, *viz.*, stretching along a direction perpendicular to the orientation of nematic order causes an increase (rather than decrease) of the elastomer thickness (see Fig. 1).[5] Liquid crystal elastomers (LCEs) are a new class of materials, which combine the orientation properties of liquid crystals with the elastic properties of elastomers.[6] Due to coupling of these two properties, such materials exhibit many novel properties, including soft or semi-soft elasticity.[7–9] In this article, we develop a simple theoretical model to explain the observed auxetic response of LCEs, and further determine by theoretical calculation the critical condition required for appearance of auxetic response.

2. Model and results

To describe the elastic properties of LCEs, a powerful theory, the so-called "neoclassical" theory, has been developed by Warner and Terentjev, which generalizes the microscopic theory for the rubber elasticity to include anisotropic orientational properties of liquid crystals.[6] Alternatively, one can develop, based on symmetry argument, phenomenological theories to describe LCEs. In both types of theories, two essential physical quantities are the orientational order tensor of liquid crystals, $\boldsymbol{Q}$, and the deformation tensor of the elastomer, $\boldsymbol{\Lambda} = \mathrm{d}\boldsymbol{R}/\mathrm{d}\boldsymbol{R}_0$, where $\boldsymbol{R}$ and $\boldsymbol{R}_0$ are, respectively, the positions of a mass point after and before a deformation.

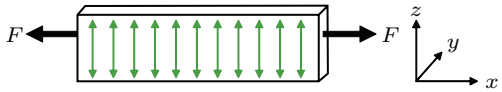


Fig. 1. Schematic illustration of the auxetic response of liquid crystal elastomers, where the vertical double-head arrows represent the initial nematic alignment orientation along the z-direction: when an external stress is applied along the x-direction, the thickness of the sample along the y-direction may increase.

In this article, we employ the phenomenological approach. As $\boldsymbol{Q}$ is currently not explicitly tuned, one can always integrate out $\boldsymbol{Q}$ and obtain a theory containing only the deformation tensor $\boldsymbol{\Lambda}$. To better describe the symmetry properties, one can further transform $\boldsymbol{\Lambda}$ to the strain tensor $\boldsymbol{u} = \frac{1}{2}(\boldsymbol{\Lambda}^{\mathrm{T}}\boldsymbol{\Lambda} - \boldsymbol{I})$, where $\boldsymbol{I}$ represents the identity matrix. Theories based on $\boldsymbol{u}$ only have been developed and studied in Refs. [8–11]. We use the one formally analogous to the de-Gennes–Maier–Saupe theory for nematics in an external field:

$$f' = \frac{1}{2}r'\,\mathrm{Tr}(\boldsymbol{uu}) - w\mathrm{Tr}(\boldsymbol{uuu}) + v\,[\mathrm{Tr}\,(\boldsymbol{uu})]^2 - h'u_{zz}, \quad (1)$$

where the symbol Tr represents the trace of a matrix, the coefficient r' is a parameter related to temperature and shear modulus of elastomers, and w and v are, respectively, the coefficients for the third- and fourth-order terms. The coefficient h' represents the strength of frozen-in anisotropy introduced

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during the cross-linking process of preparing LCE samples, where we have assumed the anisotropy direction aligns along the z-direction, which is also the initial alignment direction of the nematic. Following the rescaling approach in Ref. [10], we can further simplify the above energy expression so as to reduce the parameter number. We thus obtain the following free energy expression:

$$f = \frac{1}{2} r \mathrm{Tr}(\boldsymbol{u}\boldsymbol{u}) - \mathrm{Tr}(\boldsymbol{u}\boldsymbol{u}\boldsymbol{u}) + [\mathrm{Tr}(\boldsymbol{u}\boldsymbol{u})]^2 - h u_{zz}, \tag{2}$$

with $r = (v/w^2)r'$ and $h = \left(v^2/w^3\right)h'$. Here, f and $\boldsymbol{u}$ have been rescaled by v^3/w^4 and v/w, respectively, from their counterparts in Eq. (1). Note that the above free energy expression now has only two independent parameters r and h. In the above theory, we have assumed the strain tensor $\boldsymbol{u}$ to be traceless, i.e., $u_{xx} + u_{yy} + u_{zz} = 0$. This traceless constraint is a good approximation of the incompressibility requirement, $\mathrm{Det}\,\boldsymbol{\Lambda} = 1$, for small deformations. When considering large deformations, this traceless constraint does not quantitatively satisfy but still captures main characteristics of the incompressibility requirement. We therefore adopt this traceless constraint throughout the following parts to simplify calculations. A detailed theory following the exact incompressibility requirement will be studied in near future.

We first need to determine the equilibrium state of the free energy given in Eq. (2). Due to the presence of the linear term and the fact that the parameter r can be negative, the $\boldsymbol{u} = 0$ state is not clearly the equilibrium state. To determine the equilibrium state in the absence of external stress, we can assume that $\boldsymbol{u}$ takes a diagonal form, with

$$u_{xx} = u_{yy} = -\frac{1}{2} u_{zz} = u_0,$$

where the uniaxial symmetry of nematic elastomers and the traceless constraint have been used. The variation of the energy f with respect to u_0 gives

$$\frac{\partial f}{\partial u_0} = h + 3ru_0 + 9u_0^2 + 72u_0^3 = 0.$$

The value of u_0 can then be determined accordingly for given r and h.

We now proceed to investigate the elastic response of LCEs subjected to an external stress. Note that the new reference state is now the state with $\boldsymbol{u} = \boldsymbol{u}_0$, rather than the $\boldsymbol{u} = 0$ state. When the sample is stretched along the x-direction, u_{xx} gradually increases from u_0, and the other elements of the strain tensor $\boldsymbol{u}$ vary accordingly. As widely studied previously, a stress applied perpendicular to the anisotropy direction usually leads to the rotation of the nematic orientation, and such rotation, together with the boundary constraints, yields a stripe pattern with the nematic orientation alternating from stripe to stripe.[6] In other words, stretching along the x-direction usually causes the occurrence of nonzero off-diagonal elements of $\boldsymbol{u}$. However, in some circumstances, a perpendicular stress leads to no rotation of the nematic orientation, instead results in appearance of biaxial order.[12,13] In such cases, we can still assume that $\boldsymbol{u}$ takes a diagonal form, but with u_{yy} being not equal to u_{xx}. The free energy density f can thus be written as

$$\begin{aligned} f \;=\; & \frac{1}{2} r \left[u_{xx}^2 + u_{yy}^2 + (u_{xx} + u_{yy})^2\right] \\ & - \left[u_{xx}^3 + u_{yy}^3 - (u_{xx} + u_{yy})^3\right] \\ & + \left[u_{xx}^2 + u_{yy}^2 + (u_{xx} + u_{yy})^2\right]^2 \\ & + h\,(u_{xx} + u_{yy})\,. \end{aligned}$$

Then, for given u_{xx}, minimizing the above free energy with respect to u_{yy} yields

$$\begin{aligned} \frac{\partial f}{\partial u_{yy}} \;=\; & 16u_{yy}^3 + 24u_{xx}u_{yy}^2 + \left(2r + 6u_{xx} + 24u_{xx}^2\right) u_{yy} \\ & + 8u_{xx}^3 + 3u_{xx}^2 + ru_{xx} + h = 0. \end{aligned} \tag{3}$$

The value of u_{yy} can then be obtained by solving the above equation numerically. Figure 2 shows how u_{yy} varies with u_{xx} for various r at $h = 1$. For large r, u_{yy} decreases with u_{xx} monotonically; for small r, when u_{xx} increases, u_{yy} shows non-monotonic behavior, firstly decreasing, then rising up, and finally decreasing again. Such non-monotonic behavior corresponds exactly to LCEs' auxetic response reported recently by Mistry *et al.*[5]

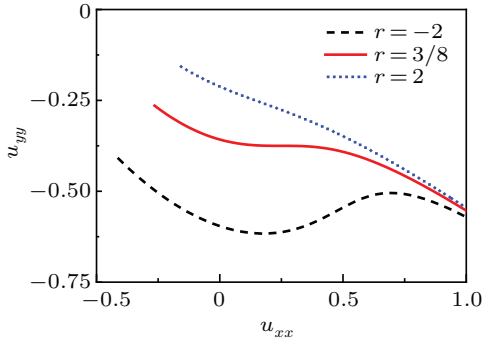


Fig. 2. Dependence of u_{yy} on u_{xx} for various r at $h = 1$. Note that the three curves start from different u_{xx} because the value change of r results in the change of u_0 as well.

After confirming the presence of the auxetic response, we proceed to investigate the exact condition under which the auxetic response may occur. As illustrated in Fig. 2, for small r, the curve of $u_{yy}(u_{xx})$ has two extreme points, and the auxetic response starts from the local minimum point and ends at the local maximum point. As the relation between u_{yy} and u_{xx} is given by Eq. (3), the derivative $\partial u_{yy}/\partial u_{xx}$ can then be easily calculated, and the positions of the two extreme points can be obtained by solving $\partial u_{yy}/\partial u_{xx} =$, which is equivalent to

$$24\,(u_{xx} + u_{yy})^2 + 6\,(u_{xx} + u_{yy}) + r = 0.$$

We thus have

$$C(r) = u_{xx} + u_{yy} = \frac{1}{8}\left(-1 \pm \sqrt{1 - \frac{8}{3}r}\right),$$

where the $-$ and $+$ signs are for the local minimum and maximum points, respectively. When $r = 3/8$, as shown by the red curve in Fig. 2, the two extreme points on the $u_{yy}(u_{xx})$ curve merge into one point, at which $\partial^2 u_{yy}/\partial^2 u_{xx} = 0$. We can thus easily conclude that the condition required for the auxetic response to occur is $r < r_c(= 3/8)$.

3. Discussion and conclusion

In conclusion, we have developed a minimal model to describe the auxetic response of liquid crystal elastomers, and obtained the condition required for the auxetic response to occur. Our theoretical result agrees well with the previous experimental results that liquid crystal elastomers are more likely to exhibit auxetic response at low temperatures.[12] Although being simplified, our model does capture the main feature that the auxetic response is related to the appearance of biaxiality of nematic alignment.[12] However, by assuming the off-diagonal elements of the strain tensor to be zero, the current work does not explain why a stress applied perpendicularly to the initial nematic orientation induces a modification of nematic order rather than a rotation of nematic orientation. Further investigation will be carried out in near future to explore the mechanisms inhibiting the rotation of nematic orientation.

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