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Positive or negative Poynting effect? The role of adscititious inequalities in hyperelastic materials

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Motivated by recent experiments on biopolymer gels whereby the reverse of the usual (positive) Poynting effect was observed, we investigate the effect of the so-called 'adscititious inequalities' on the behaviour of hyperelastic materials subject to shear. We first demonstrate that for homogeneous isotropic materials subject to pure shear, the resulting deformation consists of a triaxial stretch combined with a simple shear in the direction of the shear force if and only if the Baker–Ericksen inequalities hold. Then for a cube deformed under pure shear, the positive Poynting effect occurs if the 'sheared faces spread apart', whereas the negative Poynting effect is obtained if the 'sheared faces draw together'. Similarly, under simple shear deformation, the positive Poynting effect is obtained if the 'sheared faces tend to spread apart', whereas the negative Poynting effect occurs if the 'sheared faces tend to draw together'. When the Poynting effect occurs under simple shear, it is reasonable to assume that the same sign Poynting effect is obtained also under pure shear. Since the observation of the negative Poynting effect in semiflexible biopolymers implies that the (stronger) empirical inequalities may not hold, we conclude that these inequalities must not be imposed when such materials are described.

Keywords: nonlinear elasticity; Poynting effect; pure shear stress; simple shear deformation; adscititious inequalities; biopolymers

1. Introduction

In a recent set of experiments on semiflexible biopolymers, Janmey *et al.* (2006) showed that when sheared between two plates, some bio-gels 'tend to pull the plates together' rather than away from each other as would be expected from experiments (and theory) on hyperelastic material. The tendency to develop a stress in the direction perpendicular to the applied shear stress is known as the (general) Poynting effect (Truesdell & Noll 2004). The experimental observations in Janmey *et al.* (2006) suggest that semiflexible polymer gels exhibit a *negative* Poynting effect in the sense that the 'material tends to contract in the direction perpendicular to the applied shear stress', while for most materials the opposite, *positive* Poynting effect whereby the 'material tends to expand in the direction perpendicular to the applied shear stress' is observed. Further, the authors conclude that 'this property is directly related to the nonlinear strain–stiffening behaviour of biopolymer gels'. This conclusion is rather surprising in the context

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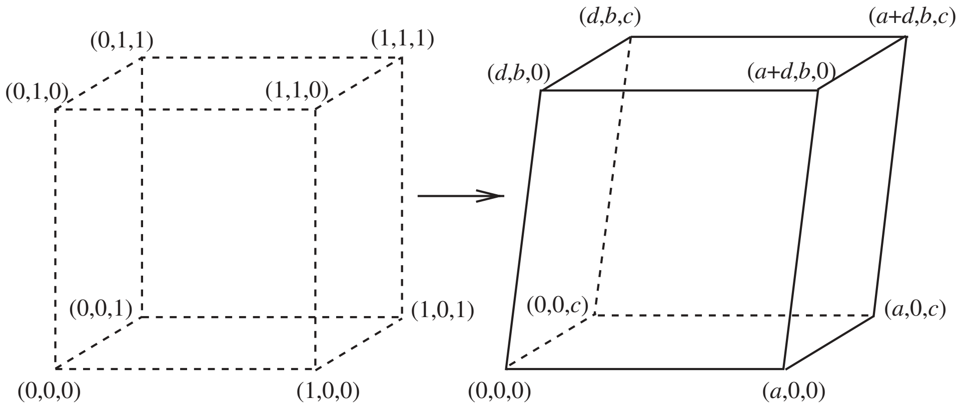

Figure 1. A cube deformed under pure shear stress ($d = \sqrt{b^2 - a^2}$).

of nonlinear elasticity, where it has been long appreciated that the sign of the Poynting effect depends not on the nonlinearity of the hyperelastic material but on the so-called 'adscititious inequalities', a set of inequalities on the coefficients of the constitutive equations derived empirically from experiments mostly on rubber materials (see Truesdell 1952 and references therein). It is important to note that the statement of the authors given above could be interpreted as relating to the micro-structural behaviour of the gels rather than its overall behaviour as an elastic material. Nevertheless, these observations raise a fundamental question of classical nonlinear elasticity, namely 'under what conditions should we obtain the inversion of the Poynting effect?'.

Here, we focus on the effect of the following adscititious inequalities on the behaviour of hyperelastic materials subject to shear: the Baker–Ericksen (BE) inequalities, which is a statement of the observation that *the greater principal stress occurs in the direction of the greater principal stretch* (cf. Baker & Ericksen 1954), the more restrictive empirical (E) inequalities, which are often conveniently assumed for hyperelastic models, and the ordered forces (OF) inequalities, which state that *the greater stretch occurs in the direction of the greater force* and hence are similar but not always equivalent to the BE inequalities.

We first show that for a homogeneous isotropic hyperelastic material subject to *pure* (*uniform*) *shear stress*, the resulting deformation consists of a triaxial stretch (pure strain deformation) combined with a simple shear in the direction of the shear force *if and only if* the BE inequalities hold. Specifically, if the Cauchy stress is given by

$$\boldsymbol{\sigma} = \begin{pmatrix} 0 & S & 0 \\ S & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}, \tag{1.1}$$

where $S > 0$ is constant, the corresponding homogeneous deformation takes the form (figure 1)

$$x = aX + \sqrt{b^2 - a^2}\,Y, \quad y = bY \quad \text{and} \quad z = cZ. \tag{1.2}$$

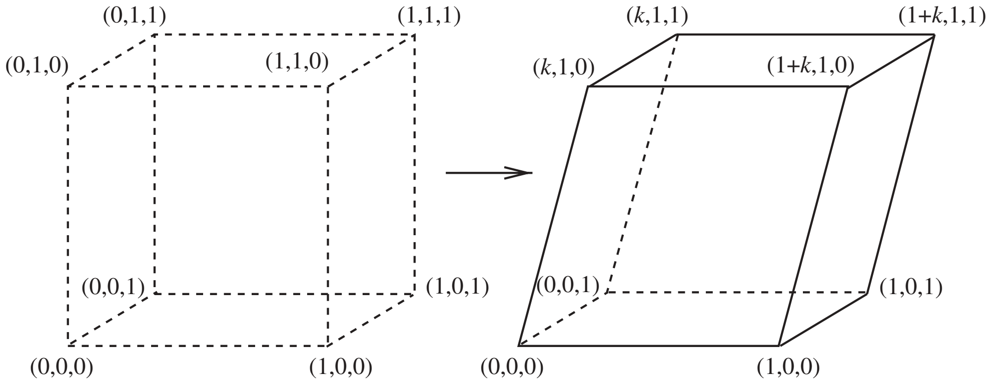

Figure 2. A cube deformed by simple shear.

For the deformation (1.2), obtained under pure shear, we define the *Poynting effect* as $0 < b \neq 1$ (Moon & Truesdell 1974). Specifically, if a cube is deformed under pure shear, we say that the *positive Poynting effect* occurs when $b > 1$, i.e. the 'sheared faces spread apart', whereas the *negative Poynting effect* is obtained when $b < 1$, i.e. the 'sheared faces draw together'.

Note that, if pre-stretching in the second direction is applied before pure shear, then the Poynting effect should be defined relative to the length in the second direction after pre-streching instead of the initial length 1. In Rajagopal & Wineman (1987), a general shear problem is presented, which, if the angle of shear is less than 45°, can be formulated as pre-stretching in the second direction followed by pure shear. It is then an interesting exercise to verify that, for the neo-Hookean example discussed there, the positive Poynting effect is obtained.

We are further interested in the occurrence of the Poynting effect in materials subject to the *simple shear deformation* (figure 2)

$$x = X + kY, \quad y = Y \quad \text{and} \quad z = Z, \tag{1.3}$$

where $k > 0$ is constant. In order for this deformation to be maintained, in addition to the pure shear stress

$$\sigma_{12} = (\beta_1 - \beta_{-1})k, \tag{1.4}$$

the following stresses in the axial directions need to be applied:

$$\sigma_{11} = \beta_0 + \beta_1 + \beta_{-1} + \beta_1 k^2, \tag{1.5}$$

$$\sigma_{22} = \beta_0 + \beta_1 + \beta_{-1} + \beta_{-1} k^2 \tag{1.6}$$

and

$$\sigma_{33} = \beta_0 + \beta_1 + \beta_{-1}, \tag{1.7}$$

where

$$\beta_0 = \frac{2}{\sqrt{I_3}}\left(I_2\frac{\partial W}{\partial I_2} + I_3\frac{\partial W}{\partial I_3}\right), \quad \beta_1 = \frac{2}{\sqrt{I_3}}\frac{\partial W}{\partial I_1} \quad \text{and} \quad \beta_{-1} = -2\sqrt{I_3}\frac{\partial W}{\partial I_2}$$

are scalar functions of the principal invariants of the Cauchy–Green strain tensor $\mathbf{B}$

$$I_1(\mathbf{B}) = \operatorname{tr}\mathbf{B}, \quad I_2(\mathbf{B}) = \frac{1}{2}((\operatorname{tr}\mathbf{B})^2 - \operatorname{tr}\mathbf{B}^2) \quad \text{and} \quad I_3(\mathbf{B}) = \det\mathbf{B},$$

and $W(I_1, I_2, I_3)$ is the stored energy function of the hyperelastic material (Ogden 1997).

For the simple shear deformation (1.3), we define the *Poynting effect* as the existence of unequal pressures $\sigma_{11} \neq \sigma_{22}$, (Truesdell & Noll 2004, pp. 176–177; Gurtin *et al.* 2010, p. 296). Then, for a cube deformed by simple shear, we say that the *positive Poynting effect* occurs if $\sigma_{22} < 0$, i.e. the 'sheared faces tend to spread apart', and hence a compressive stress σ_{22} is necessary to maintain the deformation, whereas the *negative Poynting effect* is obtained if $\sigma_{22} > 0$, i.e. the 'sheared faces tend to draw together', and therefore a tensile stress σ_{22} is required to maintain this deformation.

When the positive/negative Poynting effect is obtained under the simple shear deformation (1.3), it is reasonable to assume that the positive/negative Poynting effect will occur also under the pure shear stress (1.1), where $S = (\beta_1 - \beta_{-1})k$ and vice versa (Truesdell & Noll 2004, p. 176). We refer to this as the 'same sign Poynting effect assumption' in pure and in simple shear. Under this physical assumption, we demonstrate that if the E inequalities are valid, then for incompressible materials only the positive Poynting effect is obtained.

Concerning the experimental results in Janmey *et al.* (2006), we conclude that the observation of the reverse (negative) Poynting effect in semiflexible biopolymers implies that the (stronger) E inequalities may not hold and therefore they must not be imposed when such materials are described. In the general context, this can be regarded as evidence that 'such strong conditions are necessarily more restrictive than those they imply and their imposition might prevent a theory from describing interesting phenomena' (Truesdell & Noll 2004, preface to the 3rd edn).

2. Pure shear stress

For a hyperelastic body under *uniaxial tension*, Marzano (1983) shows that the corresponding deformation is a simple extension in the direction of the (positive) tensile force, where the ratio between the tensile strain and the strain in the perpendicular direction is greater than one *if and only if* the BE inequalities hold. Employing a similar argument, we derive the following result concerning the deformation of a body under *pure shear*. An equivalent result was obtained by Moon & Truesdell (1974). However, our formulation is different and more suitable for the subsequent analysis.

Theorem 2.1. *For a homogeneous isotropic hyperelastic material subject to the pure shear stress* (*1.1*), *the following assertions are equivalent*:

(i) *The Baker–Ericksen inequalities hold:*

$$\lambda_i \neq \lambda_j \quad \Rightarrow \quad (\sigma_i - \sigma_j)(\lambda_i - \lambda_j) > 0, \quad i, j = 1, 2, 3 \tag{2.1}$$

where $\{\sigma_i\}_{i=1,2,3}$ *and* $\{\lambda_i\}_{i=1,2,3}$ *are the principal stresses and principal stretches, respectively.*

(ii) *The corresponding left Cauchy–Green strain tensor has the representation*

$$\boldsymbol{B} = \begin{pmatrix} B_{11} & B_{12} & 0 \\ B_{12} & B_{11} & 0 \\ 0 & 0 & B_{33} \end{pmatrix}, \tag{2.2}$$

where

$$B_{11} + B_{12} > B_{33} > B_{11} - B_{12} > 0. \tag{2.3}$$

Proof. For a homogeneous and isotropic material, by the Rivlin–Ericksen (RE) representation, the Cauchy stress (1.1) can be expressed equivalently as follows:

$$\boldsymbol{\sigma} = \beta_0 \mathbf{I} + \beta_1 \mathbf{B} + \beta_{-1}\mathbf{B}^{-1}, \tag{2.4}$$

where $\mathbf{B}$ is the corresponding left Cauchy–Green strain tensor, which is symmetric and positive definite, and hence takes the general form

$$\mathbf{B} = \begin{pmatrix} B_{11} & B_{12} & B_{13} \\ B_{12} & B_{22} & B_{23} \\ B_{13} & B_{23} & B_{33} \end{pmatrix}.$$

Since by the above RE representation, $\boldsymbol{\sigma}$ and $\mathbf{B}$ have the same eigenvectors, this implies $\boldsymbol{\sigma}\mathbf{B} = \mathbf{B}\boldsymbol{\sigma}$, and hence

$$\begin{pmatrix} SB_{12} & SB_{22} & SB_{23} \\ SB_{11} & SB_{12} & SB_{13} \\ 0 & 0 & 0 \end{pmatrix} = \begin{pmatrix} SB_{12} & SB_{11} & 0 \\ SB_{22} & SB_{12} & 0 \\ SB_{23} & SB_{13} & 0 \end{pmatrix}.$$

Thus

$$B_{11} = B_{22} \quad \text{and} \quad B_{13} = B_{23} = 0, \tag{2.5}$$

and $\mathbf{B}$ takes the form (2.2).

In order to prove (i) $\Rightarrow$ (ii), it remains to show that, if the BE inequalities hold, then relations (2.3) are satisfied.

If $\{\sigma_i\}_{i=1,2,3}$ are the principal stresses, i.e. the eigenvalues of $\boldsymbol{\sigma}$, and $\{\lambda_i\}_{i=1,2,3}$ are the principal stretches, i.e. the square roots of the eigenvalues of $\mathbf{B}$, then the RE representation (2.4) implies

$$\sigma_i = \beta_0 + \beta_1\lambda_i^2 + \beta_{-1}\lambda_i^{-2}, \quad i = 1, 2, 3. \tag{2.6}$$

It is easy to verify that:

$$\sigma_1 = S, \quad \sigma_2 = -S \quad \text{and} \quad \sigma_3 = 0 \tag{2.7}$$

and

$$\lambda_1^2 = B_{11} + B_{12}, \quad \lambda_2^2 = B_{11} - B_{12} \quad \text{and} \quad \lambda_3^2 = B_{33}. \tag{2.8}$$

Then, by representation (2.6)

$$S = \beta_0 + \beta_1\lambda_1^2 + \beta_{-1}\lambda_1^{-2}, \tag{2.9}$$

$$-S = \beta_0 + \beta_1\lambda_2^2 + \beta_{-1}\lambda_2^{-2} \tag{2.10}$$

and

$$0 = \beta_0 + \beta_1\lambda_3^2 + \beta_{-1}\lambda_3^{-2}, \tag{2.11}$$

and subtracting equation (2.11) from equations (2.9) and (2.10), respectively, implies

$$S=(\lambda_1^2-\lambda_3^2)(\beta_1-\beta_{-1}\lambda_1^{-2}\lambda_3^{-2}) \tag{2.12}$$

and

$$-S=(\lambda_2^2-\lambda_3^2)(\beta_1-\beta_{-1}\lambda_2^{-2}\lambda_3^{-2}). \tag{2.13}$$

Since the BE inequalities are equivalent to

$$\lambda_i\neq\lambda_j \quad \Rightarrow \quad \beta_1-\beta_{-1}\lambda_i^{-2}\lambda_j^{-2}>0, \quad i,j=1,2,3 \tag{2.14}$$

from relations (2.12)–(2.14) we obtain:

$$\lambda_1^2>\lambda_3^2>\lambda_2^2, \tag{2.15}$$

which by the identities (2.8) is equivalent to condition (2.3).

Conversely, we show that (ii) $\Rightarrow$ (i). If the double inequality (2.15) holds, then by equations (2.12)–(2.13)

$$\beta_1-\beta_{-1}\lambda_1^{-2}\lambda_3^{-2}>0$$

and

$$\beta_1-\beta_{-1}\lambda_2^{-2}\lambda_3^{-2}>0$$

and after subtracting equation (2.13) from equation (2.12)

$$\beta_1-\beta_{-1}\lambda_1^{-2}\lambda_2^{-2}>0.$$

Therefore, relations (2.14), or equivalently the BE inequalities, are satisfied. ■

(a) *Homogeneous deformation*

When the BE inequalities are satisfied, the deformation under the pure shear stress (1.1) is a combination of a simple shear in the direction of the shear force and a triaxial stretch, and takes the general form (1.2), where

$$a=\sqrt{\frac{B_{11}^2-B_{12}^2}{B_{11}}}, \quad b=\sqrt{B_{11}} \quad \text{and} \quad c=\sqrt{B_{33}}. \tag{2.16}$$

It is easy to verify that the above deformation leads to the strain tensor (2.2). Then the strain tensor and its inverse can be written, respectively, as follows:

$$\mathbf{B}=\begin{pmatrix} b^2 & b\sqrt{b^2-a^2} & 0\\ b\sqrt{b^2-a^2} & b^2 & 0\\ 0 & 0 & c^2\end{pmatrix}$$

and

$$\mathbf{B}^{-1}=\frac{1}{a^2b^2c^2}\begin{pmatrix} b^2c^2 & -bc^2\sqrt{b^2-a^2} & 0\\ -bc^2\sqrt{b^2-a^2} & b^2c^2 & 0\\ 0 & 0 & a^2b^2\end{pmatrix}$$

and by the representation (2.4):

$$S=b\sqrt{b^2-a^2}\left(\beta_1-\beta_{-1}\frac{1}{a^2b^2}\right). \tag{2.17}$$

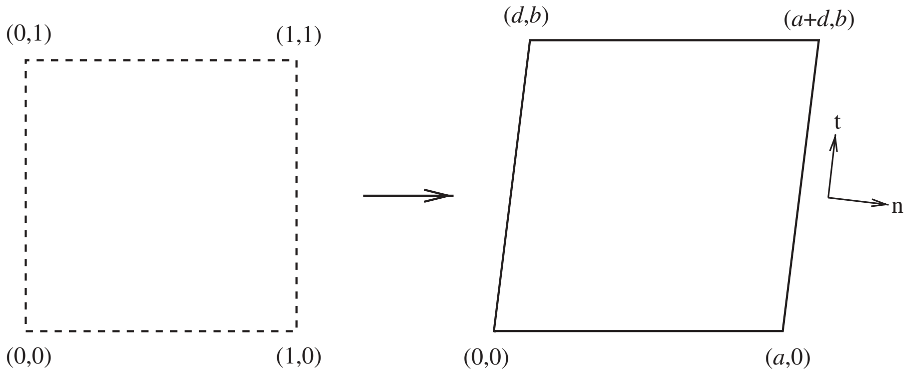

Figure 3. Cross section in the plane of shear for a cube deformed under pure shear stress ($d=\sqrt{b^2-a^2}$).

In the plane of shear, for a cube deformed under pure shear, the normal and shear (tangent) tractions on the inclined faces (the faces that were perpendicular to the sheared faces) are as follows (see also Destrade *et al.* in press):

$$\sigma_n = \mathbf{n}\cdot\boldsymbol{\sigma}\mathbf{n} = -2S\frac{b\sqrt{b^2-a^2}}{2b^2-a^2} < 0 \quad \text{and} \quad \sigma_t = \mathbf{t}\cdot\boldsymbol{\sigma}\mathbf{n} = S\frac{a^2}{2b^2-a^2} > 0$$

where

$$\mathbf{n} = \pm\frac{1}{\sqrt{2b^2-a^2}}\left(b, -\sqrt{b^2-a^2}, 0\right) \quad \text{and} \quad \mathbf{t} = \pm\frac{1}{\sqrt{2b^2-a^2}}\left(\sqrt{b^2-a^2}, b, 0\right)$$

are the normal and tangent unit vectors, respectively (figure 3). Therefore, the necessary normal traction is compressive while extension occurs in the shear direction. The corresponding normal and shear strains are, respectively,

$$B_n = \mathbf{n}\cdot\mathbf{B}\mathbf{n} = \frac{a^2b^2}{2b^2-a^2} \quad \text{and} \quad B_t = \mathbf{t}\cdot\mathbf{B}\mathbf{n} = \frac{a^2b\sqrt{b^2-a^2}}{2b^2-a^2}.$$

The shear modulus is given by (Moon & Truesdell 1974):

$$\mu = \frac{\sigma_t}{B_t/B_{11}}.$$

Hence

$$\mu = S\frac{b}{\sqrt{b^2-a^2}} > 0. \tag{2.18}$$

(*b*) *Linearized pure shear*

We now investigate the case of small pure shear. Clearly, if $a, b, c \to 1$, then relations (2.17) and (2.18) imply $0 < \mu \to \beta_1 - \beta_{-1}$ and $S \to 0$. Thus, in the initial state, i.e. before the shear stress is applied, $\beta_1 > \beta_{-1}$. Conversely, assuming that the initial state is stress free, let the applied shear stress satisfy $S \to 0$. Then, by equations (2.17) and (2.18), $\mu = \beta_1 b^2 - \beta_{-1} a^{-2} > 0$ and $b^2 - a^2 \to 0$, hence by representation (2.6), $\lambda_i \to 1$, for all $i=1,2,3$, or $\lambda_i \to \beta_{-1}/\beta_1$, for all $i=1,2,3$. However, the latter implies $b^2 \to \beta_{-1}/\beta_1$ and $a^2 \to \beta_{-1}/\beta_1$, and since $\mu > 0$, it follows that $\beta_{-1} > \beta_1$ in the initial state, which contradicts the reverse inequality obtained earlier. Therefore, $\lambda_i \to 1$, for all $i=1,2,3$, i.e. $a, b, c \to 1$ as $S \to 0$. If we denote $k=\sqrt{b^2-a^2}$, then $k \to 0$ and the strain tensor (2.2) is approximated to the first order in k by

$$\mathbf{B} = \begin{pmatrix} 1 & k & 0 \\ k & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix}. \tag{2.19}$$

(*c*) *The ordered forces inequalities*

Similar to the BE inequalities are the so-called OF inequalities, which state that *the greater stretch occurs in the direction of the greater force*, but in general, these two sets of inequalities do not imply each other (Truesdell & Noll 2004, pp. 157–159). However, it can be shown that if two of the three principal stresses are non-negative, then the BE inequalities follow from the OF inequalities, and if two of the three principal stresses are non-positive, then the OF inequalities are implied by the BE inequalities (Truesdell & Noll 2004, p. 160). Since for the pure shear stress (1.1) the principal stresses are expressed by relations (2.7), we deduce that, *under pure shear, the BE and the OF inequalities are simultaneously satisfied.*

(*d*) *The empirical inequalities*

We also investigate the effect of the BE inequalities on the following E inequalities (Moon & Truesdell 1974):

$$\beta_0 \leq 0, \quad \beta_1 > 0 \quad \text{and} \quad \beta_{-1} \leq 0. \tag{2.20}$$

If we multiply equations (2.9) and (2.11) by λ_3^{-2} and $-\lambda_1^{-2}$, respectively, then add the two results, we obtain

$$S\lambda_3^{-2} = \beta_0(\lambda_3^{-2} - \lambda_1^{-2}) + \beta_1(\lambda_1^2\lambda_3^{-2} - \lambda_1^{-2}\lambda_3^2).$$

Since in the above identity, the left-hand side is positive and by the double inequality (2.15), the result in each of the two brackets on the right-hand side is also positive, assuming that the first E inequality is valid, then the second E inequality follows, i.e.

$$\beta_0 \leq 0 \;\Rightarrow\; \beta_1 > 0.$$

Therefore, under pure shear, the BE inequalities imply that the first two E inequalities may hold, but not necessarily the third.

On the other hand, by relations (2.14) and (2.15), we obtain

$$\beta_{-1} < \beta_1 \lambda_2^2 \lambda_3^2, \tag{2.21}$$

where λ_2^2 and λ_3^2 are the smallest principal strains. Conversely, if relation (2.21) holds, then the BE inequalities follow.

In particular, if all three E inequalities are valid, then the BE inequalities are satisfied. Moreover, adding equations (2.9) and (2.10) yields

$$\beta_0 + \beta_1 + \beta_{-1} = \beta_1(1 - b^2) + \beta_{-1}(1 - a^{-2}), \tag{2.22}$$

while equation (2.11) implies

$$\beta_0 + \beta_1 + \beta_{-1} = (1 - c^2)(\beta_1 - \beta_{-1}c^{-2}). \tag{2.23}$$

Thus, we obtain the following result.

Proposition 2.2. *Under pure shear stress, assuming that the E inequalities hold, the following assertions are valid*:

(i) *If the negative Poynting effect occurs, i.e.* $b < 1$, *then* $a < 1$ *and* $c < 1$;
(ii) *If* $c > 1$, *then* $b > 1$, *i.e. the positive Poynting effect is obtained*; *and*
(iii) *If* $c = 1$, *then either* $b > 1$, *i.e. the positive Poynting effect occurs*, *or* $b = 1$, *i.e. there is no Poynting effect*, *in which case* $\beta_{-1} = 0$.

Proof.

(i) If the negative Poynting effect occurs, i.e. $b < 1$, then, since $a < b$, we obtain $a < 1$, hence by equation (2.22) $\beta_0 + \beta_1 + \beta_{-1} > 0$ and by equation (2.23) $c < 1$.
(ii) If $c > 1$, then on the one hand by equation (2.23), $\beta_0 + \beta_1 + \beta_{-1} < 0$, and on the other hand by (i) $b \geq 1$. Assuming $b = 1$, since $a < b$, we obtain $a < 1$, hence by equation (2.22) $\beta_0 + \beta_1 + \beta_{-1} \geq 0$, which is a contradiction. Thus, $b > 1$, i.e. the positive Poynting effect occurs.
(iii) If $c = 1$, then on the one hand by equation (2.23), $\beta_0 + \beta_1 + \beta_{-1} = 0$, and on the other hand by (i): either $b > 1$, i.e. the positive Poynting effect occurs, or $b = 1$, i.e. there is no Poynting effect, in which case, since $a < b$, by equation (2.22) $\beta_{-1} = 0$. ■

(*e*) *Incompressible materials*

For incompressible materials, since the volume must be preserved by the deformation (1.2), i.e. $abc = 1$, an immediate consequence of proposition 2.2 is that when the negative Poynting effect occurs, the E inequalities cannot hold. In this case, if the first two E inequalities are valid, then $\beta_{-1} > 0$.

Alternatively, if the E inequalities are assumed, then the positive Poynting effect is obtained. Indeed, if the Poynting effect does not occur, i.e. $b = 1$, then on the one hand, since $a < b$, by equation (2.22) $\beta_0 + \beta_1 + \beta_{-1} \geq 0$, and on the other hand, since $a < 1$ and $abc = 1$, it follows that $c > 1$, hence by equation (2.23) $\beta_0 + \beta_1 + \beta_{-1} < 0$, which is a contradiction.

3. Simple shear deformation

We now investigate the occurrence of the Poynting effect in a material under the simple shear deformation (1.3). The left Cauchy–Green strain tensor for this

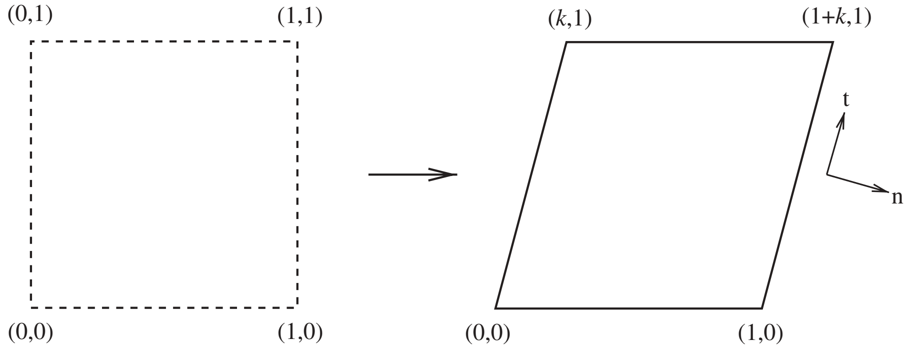

Figure 4. Cross section in the plane of shear for a cube deformed by simple shear.

deformation is

$$\mathbf{B} = \begin{pmatrix} 1+k^2 & k & 0 \\ k & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix} \tag{3.1}$$

and the corresponding Cauchy stress tensor takes the general form

$$\boldsymbol{\sigma} = \beta_0 \mathbf{I} + \beta_1 \mathbf{B} + \beta_{-1} \mathbf{B}^{-1}.$$

In this case, the eigenvalues of the strain tensor (3.1) are as follows:

$$\lambda_1^2 = 1 + \frac{k^2 + k\sqrt{k^2+4}}{2}, \quad \lambda_2^2 = 1 + \frac{k^2 - k\sqrt{k^2+4}}{2} \quad \text{and} \quad \lambda_3^2 = 1, \tag{3.2}$$

and the corresponding principal stresses have the representation (2.6). Thus

$$\lambda_2^2 < \lambda_3^2 < \lambda_1^2,$$

and the BE inequalities, or equivalently (2.14), are satisfied if and only if

$$\beta_1 > \beta_{-1} \lambda_2^{-2}.$$

Thus,

$$\beta_1 > \beta_{-1}. \tag{3.3}$$

In the plane of shear, for a cube deformed by simple shear, the normal and tangent unit vectors on the inclined faces are, respectively (figure 4),

$$\mathbf{n} = \pm \frac{1}{\sqrt{1+k^2}} (1, -k, 0) \quad \text{and} \quad \mathbf{t} = \pm \frac{1}{\sqrt{1+k^2}} (k, 1, 0).$$

On these faces, the corresponding normal and shear tractions are, respectively,

$$\sigma_n = \frac{\beta_0 (1+k^2) + \beta_1 + \beta_{-1}(1+3k^2+k^4)}{1+k^2} \quad \text{and} \quad \sigma_t = \frac{(\beta_1 - \beta_{-1})(k - k^3 + k^4)}{1+k^2}$$

and the normal and shear strains take the form:

$$B_n = \frac{1}{1+k^2} \quad \text{and} \qquad B_t = \frac{k}{1+k^2}.$$

The shear modulus is

$$\mu = (\beta_1 - \beta_{-1})(1 - k^2 + k^3) > 0. \tag{3.4}$$

(*a*) *Linearized simple shear*

In the case of small shear $k \to 0$, if the initial state is stress free, then by the identities (3.2) $\lambda_i \to 1$, for all $i = 1, 2, 3$, and by equations (1.4)–(1.7), $\sigma_{ij} \to 0$, for all $i, j = 1, 2, 3$. Conversely, if $\sigma_{ij} \to 0$, for all $i, j = 1, 2, 3$, since by expression (3.4), $\beta_1 > \beta_{-1}$, then $k \to 0$. In this case, the strain tensor (3.1) is approximated to the first order in k by the strain tensor (2.19) obtained in the pure shear situation. However, we note that the Poynting effect is not captured by this linear approximation.

(*b*) *The empirical inequalities*

When the BE inequalities are satisfied, we multiply the first and the third principal stresses by λ_3^{-2} and $-\lambda_1^{-2}$, respectively, then add the two results to obtain

$$\sigma_1 - \sigma_3 \lambda_1^{-2} = \beta_0 (1 - \lambda_1^{-2}) + \beta_1 (\lambda_1^2 - \lambda_1^{-2}).$$

Since in the above identity the left-hand side is positive, and on the right-hand side, the result in each bracket is also positive, $\beta_0 \leq 0$ implies $\beta_1 > 0$.

Thus, under simple shear also, the BE inequalities imply that the first two E inequalities may hold, but not necessarily the third.

Assuming that all three E inequalities are valid, if the negative Poynting effect is obtained under the pure shear stress (1.1) for some $S = (\beta_1 - \beta_{-1})k$ and $\beta_{-1} = 0$, then under the same sign Poynting effect assumption, the negative Poynting effect occurs also in simple shear, for all $k > 0$. Hence, the negative Poynting effect will always occur under pure shear, for all $S > 0$. Alternatively, if $\beta_{-1} \neq 0$, there exists $k_0 = \sqrt{-(\beta_0 + \beta_1 + \beta_{-1})/\beta_{-1}}$, such that if $k < k_0$, then $\sigma_{22} > 0$, i.e. the negative Poynting effect occurs, while if $k = k_0$, then $\sigma_{22} = 0$, i.e. there is no Poynting effect. Since by the definition of the Poynting effect in simple shear, the latter implies $\sigma_{22} = \sigma_{11}$, i.e. $\beta_{-1} = \beta_1 > 0$, which contradicts the E inequalities, we conclude that $\beta_{-1} = 0$.

Therefore, under the same sign Poynting effect assumption, if a cube is subject to pure shear, it is not possible for the sheared faces to first draw together and then return to their initial mutual distance as the shear increases.

Note that, in Moon & Truesdell (1974), where only the pure shear stress is treated, it is stated that it is possible for $1 - c$ to change sign at some $S = S_0$. Here, by also considering the simple shear deformation, under the same sign Poynting effect assumption, we have proved that if the negative Poynting effect occurs first, then by proposition 2.2 (i) $1 - c > 0$ and therefore, $1 - c$ cannot change sign.

Thus, we arrive at the following result.

Proposition 3.1. *Under the same sign Poynting effect assumption, if the E inequalities are valid and the negative Poynting effect occurs in pure shear, then the negative Poynting effect is obtained also in simple shear and* $\beta_{-1}=0$*. Consequently, if the negative Poynting effect occurs and* $\beta_{-1}\neq 0$*, then the E inequalities cannot hold.*

(*c*) *Incompressible materials*

For isotropic incompressible materials, equation (2.4) becomes

$$\boldsymbol{\sigma}=-p\mathbf{I}+\beta_1\mathbf{B}+\beta_{-1}\mathbf{B}^{-1}, \tag{3.5}$$

where β_1 and β_{-1} are as before, and p is the arbitrary hydrostatic pressure. By setting $\sigma_{33}=0$, we obtain

$$p=\beta_1+\beta_{-1},$$

and the stresses (1.5) and (1.6) now are

$$\sigma_{11}=\beta_1 k^2$$

and

$$\sigma_{22}=\beta_{-1}k^2.$$

In this case, it is easy to see that the necessary and sufficient condition for the positive Poynting effect to occur is $\beta_{-1}<0$, while the negative Poynting effect is obtained if and only if $\beta_{-1}>0$. Thus, when the negative Poynting effect occurs, the E inequalities are not valid.

(*d*) *Experimental observations*

In the experimental results reported by Janmey *et al.* (2006), where bio-gels were subjected to simple shear deformation, on the one hand, since the shear stresses σ_{12} are positive, it is reasonable to assume that the BE inequalities are satisfied for both flexible and semiflexible polymers (see Hayes 1969). On the other hand, for semiflexible biopolymers, where the negative Poynting effect was obtained, from the experimental observations and the results derived here, we infer that these bio-gels can be modelled as homogeneous isotropic hyperelastic materials, such that $\beta_0\leq 0$ and $\beta_1>\beta_{-1}\geq 0$. If the material is incompressible and $p=\beta_1+\beta_{-1}$, then $\beta_{-1}>0$, hence the E inequalities do not hold. This conclusion was also reached by Murphy & Horgan (in preparation), where the authors use an incompressible (and nearly incompressible) material model to fit the data from Janmey *et al.* (2006). Moreover, since the shear modulus (3.4) increases with the deformation parameter k, one expects strain–stiffening to occur when simple shear deformation increases. Although the experimental results in Janmey *et al.* (2006) are for simple shear deformation, where displacements can be controlled, under the same sign Poynting effect assumption, one expects the same Poynting effect to occur in pure shear as well. While simple shear deformation is easier to quantify experimentally, the sign of the Poynting effect under pure shear could be directly observed, in a qualitative manner, from the movement of the sheared faces.

4. Conclusion

We have shown that, in the context of hyperelasticity, the sign of the Poynting effect does not depend on whether the material is compressible or not, or whether the material is strain–stiffening or not, but on the validity of certain adscititious inequalities. In the case of the experimental results from Janmey *et al.* (2006), the magnitude of the effect observed in semiflexible biopolymer gels and the overall description of the material suggest that a negative Poynting effect has been observed, which is consistent with the fact that the BE inequalities hold but the E inequalities may not. Therefore, a possible model for these bio-gels is that of a homogeneous isotropic nonlinear elastic material (compressible or incompressible), such that $\beta_0 \leq 0$ and $\beta_1 > \beta_{-1} \geq 0$. If the material is incompressible and $\beta_0 + \beta_1 + \beta_{-1} = 0$, then $\beta_{-1} > 0$, and the E inequalities do not hold. In hindsight, this is not truly surprising. Indeed, there is no reason to expect that a bio-gel, that is a hydrogel made out of semiflexible polymers, should behave as a classical elastomer. Decades of theory and experiments on elastomers have lulled us into the acceptance that, despite their names, the adscititious inequalities should be considered as laws of nature and that most elastic materials should behave as rubber in large deformations. The realm of biological materials offers us many different and counterintuitive mechanical behaviours, and biological systems are constantly forcing us to revisit our understanding of fundamental concepts of mechanics and material modelling. Nevertheless, the question 'what is the physical origin of the negative Poynting effect in some bio-gels?' remains. The obvious suspect is indeed the nonlinear stress response of the semiflexible polymers under deformation and their network structure. However, the relationship between the microscopic properties of the semiflexible polymers and the overall macroscopic response of the gel viewed as a material remains elusive.

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