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# On the Non Linear Behaviour of Nematic Liquid Single Crystal Elastomers (NLSCE) Under Biaxial Mechanic and Electrical Force Fields

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*A continuum theory has been developed to give account of the three characteristic zones appearing in the “semisoft” behaviour of nematic single crystal elastomers under the effect of biaxial mechanic and electrical force fields considering higher order coupling terms. The analysis is based on a model which takes into account two coexistent coupled components showing a preferred direction. Two cases are analysed according to the type of input: stress and displacement. Bifurcation phenomena are predicted in the first case. Stress-strain curves show the characteristic features experimentally observed. The case where shear terms in the strain are considered is approximately analysed.*

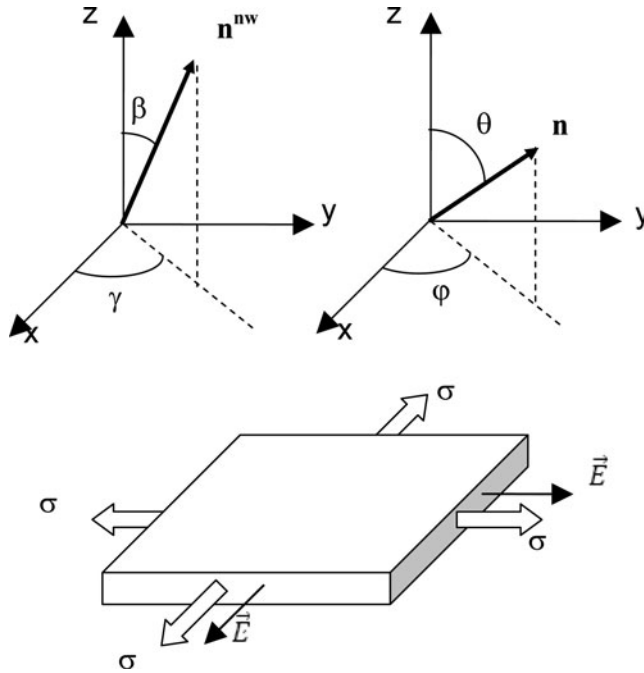
**Keywords** nematic single crystal; elastomers; biaxial case

## Introduction

The reorientation of the director in a nematic liquid crystal and the concomitant “soft elasticity” has been predicted by Golubovic and Lubensky [1] and experimentally checked by Finkelmann [2]. A theory of “soft elasticity” containing details about the molecular structure of the materials predicting the reorientation of the director has been developed by Warner and Terentjev [3]. This theory predicts that the director rotation should start immediately after the sample is stretched. The shear modulus should be zero until the rotation is completed. However, the actual stress-strain curves show a “semi-soft” behaviour in which three characteristic zones are present. The first and the third correspond to the stretch before and after the director rotation. Between these two zones a plateau is shown where the shear modulus can be relatively high. A static pattern in the form of stripe domains has been also found [2]. A continuum theory avoiding details about the molecular structure of the materials has been developed in the Chapter 10 of the Reference [3]. In the present work a non linear analysis using higher order coupling terms for the rotation of the director than those proposed by de Gennes [4] has been carried out. The analysis is based on a model proposed by Menzel, Pleiner and Brand [5,6] which takes into account two coexisting coupled components both showing a preferred direction. However, only uniaxial stretching has been considered in these previous papers. For this reason it seems

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**Figure 1.** Squared slab of a single crystal nematic elastomer  $i$  subjected to a pair of equal mechanic and electrical force fields in the plane  $x$ - $y$ . The orientations of the director and the network (respectively  $\mathbf{n}_0$ ,  $\mathbf{n}_0^{nw}$ ) are initially parallel to the  $z$ -direction.

convenient to examine the biaxially stretching case. Moreover, the simultaneous application of mechanic and electrical force field should be analysed. The relevant terms of the free energy are: a) the elastic energy of the network, b) the coupling energy between the relative rotations of the director and the network and the strain and c) the electrostatic energy. It should be noted that the elastic contribution of the network to the free energy is, in general, several orders of magnitude larger than the contribution becoming from the Frank energy. For this reason the Frank energy will be disregarded as a first approach. Moreover, if we are only interested in the bulk phenomena the effect of the boundaries can be neglected and consequently only spatially homogeneous solutions for the rotation of the director are derived. Bifurcation phenomena are predicted and discussed. The two types of inputs, namely by displacement and stress, are analysed [7]. In the same way, the two cases with and without shear are also studied.

### Geometry of the Problem

A squared slab of a single crystal nematic elastomer (typically  $2.5 \times 2.5 \text{ cm}^2$ ) is subjected to a pair of equal mechanic and electrical force fields in the plane  $x$ - $y$  (figure 1). The sample has been prepared in such a way that the orientations of the director and the network, respectively represented by the unit vectors  $\mathbf{n}_0$ ,  $\mathbf{n}_0^{nw}$  are initially parallel to the  $z$ -direction. After the application of the mechanic and electrical inputs the unit vectors are expressed by

$$\begin{aligned} \mathbf{n} &= (\sin \theta \cos \varphi, \sin \theta \sin \varphi, \cos \theta) \\ \mathbf{n}^{nw} &= (\sin \beta \cos \gamma, \sin \beta \sin \gamma, \cos \beta) \end{aligned} \quad (1)$$

The relative rotations of the director and the network are defined according to the Ref [5,6] by the following expressions

$$\begin{aligned}\boldsymbol{\omega} &:= \mathbf{n} - [\mathbf{n} \cdot \mathbf{n}^{nw}] \mathbf{n}^{nw} \\ \boldsymbol{\omega}^{nw} &:= -\mathbf{n}^{nw} + [\mathbf{n} \cdot \mathbf{n}^{nw}] \mathbf{n}\end{aligned}\quad (2)$$

### Specific Free Energy of the System and Preliminary Consequences of the Equilibrium Equations

a) If the input is by equibiaxial loads, one has the following expression

$$\begin{aligned}F = & c_1 (\varepsilon_{xx}^2 + \varepsilon_{yy}^2 + \varepsilon_{zz}^2 + 2\varepsilon_{xz}^2 + 2\varepsilon_{yz}^2) + \frac{1}{2} c_2 (\varepsilon_{xx} + \varepsilon_{yy} + \varepsilon_{zz})^2 \\ & + \frac{1}{2} D_1 \omega_i \omega_i + D_1^{(2)} (\omega_i \omega_i)^2 + D_2 n_i \varepsilon_{ij} \omega_j + D_2^{nw} n_i^{nw} \varepsilon_{ij} \omega_j^{nw} \\ & + D_2^{(2)} n_i \varepsilon_{ij} \varepsilon_{jk} \omega_k + D_2^{nw, (2)} n_i^{nw} \varepsilon_{ij} \varepsilon_{jk} \omega_k^{nw} - \\ & - \frac{1}{4} \varepsilon_0 \Delta \varepsilon_a E^2 \sin^2 \theta (1 + \sin 2\varphi)\end{aligned}\quad (3)$$

In Eq. (3) isotropy and linear elastic energy of the network are considered in order to reveal more clearly the effect of the complex coupling of the strain with the network and director rotations. The coefficients  $c_1, c_2$  are the Lamé constants of the material,  $\varepsilon_{ij}$  is the strain tensor,  $D, s$  are material coupling constants related to the network and the director,  $\varepsilon_0$  the permittivity of the evacuated space,  $\Delta \varepsilon_a$  the dielectric anisotropy,  $E$  the electric field and the angles  $\theta$  and  $\varphi$  are defined in Figure 1. According to the Eq. (3) and the definitions given by Eq. (1) and (2) the pertinent expression of the free energy is

$$\begin{aligned}F = & \frac{1}{2} c_2 (\varepsilon_{xx} + \varepsilon_{yy} + \varepsilon_{zz})^2 + c_1 (\varepsilon_{xx}^2 + \varepsilon_{yy}^2 + \varepsilon_{zz}^2 + 2\varepsilon_{xz}^2 + 2\varepsilon_{yz}^2) \\ & - \frac{1}{4} \varepsilon_0 \Delta \varepsilon E^2 \sin^2 \theta (1 + \sin 2\varphi) + \\ & + \frac{1}{2} D_1 [1 - (\bullet)^2] + D_1^{(2)} [1 - (\bullet)^2]^2 + \\ & + D_2 \left[ \frac{1}{2} (\varepsilon_{xx} + \varepsilon_{yy}) \sin \theta [\sin \theta - (\bullet) \sin \beta] + \right. \\ & \left. + \varepsilon_{zz} \cos \theta [\cos \theta - (\bullet) \cos \beta] + \right. \\ & \left. + \frac{\sqrt{2}}{2} (\varepsilon_{xz} + \varepsilon_{yz}) [\sin 2\theta - (\bullet) \sin (\theta + \beta)] \right] + \\ & + D_2^{nw} \left[ \frac{1}{2} (\varepsilon_{xx} + \varepsilon_{yy}) \sin \beta [-\sin \beta + (\bullet) \sin \theta] + \right. \\ & \left. + \varepsilon_{zz} \cos \beta [-\cos \beta + (\bullet) \cos \theta] + \right. \\ & \left. + \frac{\sqrt{2}}{2} (\varepsilon_{xz} + \varepsilon_{yz}) [-\sin 2\beta + (\bullet) \sin (\theta + \beta)] \right] + \\ & + D_2^{(2)} \left[ \frac{1}{2} (\varepsilon_{xx}^2 + \varepsilon_{yy}^2 + (\varepsilon_{xz} + \varepsilon_{yz})^2) [\sin \theta (\sin \theta - (\bullet) \sin \beta)] + \right. \\ & \left. + (\varepsilon_{zz}^2 + \varepsilon_{xz}^2 + \varepsilon_{yz}^2) \cos \theta (\cos \theta - (\bullet) \cos \beta) + \right. \\ & \left. - \sqrt{2} (\varepsilon_{xx} \varepsilon_{yz} + \varepsilon_{yy} \varepsilon_{xz}) [\sin 2\theta - (\bullet) \sin (\theta + \beta)] \right] +\end{aligned}$$

$$+D_2^{nw,(2)} \left[ \begin{aligned} & \frac{1}{2} \left( \varepsilon_{xx}^2 + \varepsilon_{yy}^2 + (\varepsilon_{xz} + \varepsilon_{yz})^2 \right) [\sin \beta (-\sin \beta + (\bullet) \sin \theta)] + \\ & + \left( \varepsilon_{zz}^2 + \varepsilon_{xz}^2 + \varepsilon_{yz}^2 \right) \cos \beta (-\cos \beta + (\bullet) \cos \theta) + \\ & - \sqrt{2} (\varepsilon_{xx} \varepsilon_{yz} + \varepsilon_{yy} \varepsilon_{xz}) [-\sin 2\beta + (\bullet) \sin (\theta + \beta)] \end{aligned} \right] \quad (4)$$

where

$$(\bullet) = (\cos \theta \cos \beta + \sin \theta \sin \beta \cos (\varphi - \gamma)) \quad (5)$$

From the equilibrium conditions

$$\frac{\partial F}{\partial \varepsilon_{xz}} = \frac{\partial F}{\partial \varepsilon_{yz}} = \frac{\partial F}{\partial \varphi} = \frac{\partial F}{\partial \gamma} = 0 \quad (6)$$

one obtains  $\varphi = \gamma$ ,  $\varepsilon_{xz} = \varepsilon_{yz}$ . Moreover, one concludes that  $\varphi = \gamma = \pm 45^\circ$  as expected and  $\sin \beta \approx \pm \sqrt{2} \varepsilon_{xz}$  (for simplicity the sign plus will be taken in the following). It should be noted that this result predicts a bifurcation for the meridian angle  $\beta$  near  $\beta = 0$ . Then Eq. (5) becomes

$$(\bullet) = \cos (\theta - \beta) \quad (7)$$

Moreover, shear terms  $\varepsilon_{xz} = \varepsilon_{yz}$  and  $\varepsilon_{yz} = \varepsilon_{zy}$  only differs from zero when the director rotation is in course (from  $\theta = 0$  to  $\theta = \pi/2$ ).

In the same way, from

$$\frac{\partial F}{\partial \varepsilon_{xx}} = \frac{\partial F}{\partial \varepsilon_{yy}} = \sigma \rightarrow \frac{\partial F}{\partial \varepsilon_{xx}} - \frac{\partial F}{\partial \varepsilon_{yy}} = 0 \quad (8)$$

one obtains

$$(\varepsilon_{xx} - \varepsilon_{yy}) \left[ 2c_1 + \left( D_2^{(2)} \sin^2 \theta - D_2^{nw,(2)} \sin^2 \beta \right) - \left( D_2^{(2)} - D_2^{nw,(2)} \right) \cos (\theta - \beta) \sin \theta \sin \beta \right] = 0 \quad (9)$$

Eq. (9) is relevant because it leaves open the possibility of a pitchfork bifurcation when the second factor of this equation becomes zero and  $\varepsilon_{xx} \neq \varepsilon_{yy}$ . If this is not the case, then the following equality holds,  $\varepsilon_{xx} = \varepsilon_{yy}$  and no bifurcation is predicted. If incompressibility is assumed, then for very small deformations one has  $\varepsilon_{zz} = -2\varepsilon_{xx}$  and consequently the term with coefficient  $c_2$  disappears in the main equation for the free energy. On these premises the free energy equation reduces to

$$\begin{aligned} F = & c_1 (6\varepsilon_{xx}^2 + 4\varepsilon_{xz}^2) - \frac{1}{2} \varepsilon_0 \Delta \varepsilon_a E^2 \sin^2 \theta + \frac{1}{2} D_1 \sin^2 (\theta - \sqrt{2} \varepsilon_{xz}) \\ & + D_1^{(2)} \sin^4 (\theta - \sqrt{2} \varepsilon_{xz}) + \\ & + D_2 \left[ \begin{aligned} & \varepsilon_{xx} \sin \theta \left( \sin \theta - \cos (\theta - \sqrt{2} \varepsilon_{xz}) \sin \sqrt{2} \varepsilon_{xz} \right) + \\ & + \sqrt{2} \varepsilon_{xz} \left( \sin 2\theta - \cos (\theta - \sqrt{2} \varepsilon_{xz}) \sin (\theta + \sqrt{2} \varepsilon_{xz}) \right) + \\ & - 2\varepsilon_{xx} \cos \theta \left( \cos \theta - \cos (\theta - \sqrt{2} \varepsilon_{xz}) \cos \sqrt{2} \varepsilon_{xz} \right) \end{aligned} \right] + \end{aligned}$$

$$\begin{aligned}
& + D_2^{nw} \left[ \begin{aligned} & \varepsilon_{xx} \sin \sqrt{2} \varepsilon_{xz} \left( -\sin \sqrt{2} \varepsilon_{xz} + \cos \left( \theta - \sqrt{2} \varepsilon_{xz} \right) \sin \theta \right) + \\ & + \sqrt{2} \varepsilon_{xz} \left( -\sin 2\sqrt{2} \varepsilon_{xz} + \cos \left( \theta - \sqrt{2} \varepsilon_{xz} \right) \sin \left( \theta + \sqrt{2} \varepsilon_{xz} \right) \right) + \\ & - 2\varepsilon_{xx} \cos \sqrt{2} \varepsilon_{xz} \left( -\cos \sqrt{2} \varepsilon_{xz} + \cos \left( \theta - \sqrt{2} \varepsilon_{xz} \right) \cos \theta \right) \end{aligned} \right] + \\
& + D_2^{(2)} \left[ \begin{aligned} & \left( \varepsilon_{xx}^2 + 2\varepsilon_{xz}^2 \right) \sin \theta \left( \sin \theta - \cos \left( \theta - \sqrt{2} \varepsilon_{xz} \right) \sin \sqrt{2} \varepsilon_{xz} \right) + \\ & - \sqrt{2} \varepsilon_{xz} \varepsilon_{xx} \left( \sin 2\theta - \cos \left( \theta - \sqrt{2} \varepsilon_{xz} \right) \sin \left( \theta + \sqrt{2} \varepsilon_{xz} \right) \right) + \\ & + \left( 4\varepsilon_{xx}^2 + 2\varepsilon_{xz}^2 \right) \cos \theta \left( \cos \theta - \cos \left( \theta - \sqrt{2} \varepsilon_{xz} \right) \cos \sqrt{2} \varepsilon_{xz} \right) \end{aligned} \right] + \\
& + D_2^{nw,(2)} \left[ \begin{aligned} & \left( \varepsilon_{xx}^2 + 2\varepsilon_{xz}^2 \right) \sin \sqrt{2} \varepsilon_{xz} \left( -\sin \sqrt{2} \varepsilon_{xz} - \cos \left( \theta - \sqrt{2} \varepsilon_{xz} \right) \sin \theta \right) + \\ & - \sqrt{2} \varepsilon_{xz} \varepsilon_{xx} \left( -\sin 2\sqrt{2} \varepsilon_{xz} + \cos \left( \theta - \sqrt{2} \varepsilon_{xz} \right) \sin \left( \theta + \sqrt{2} \varepsilon_{xz} \right) \right) + \\ & + \left( 4\varepsilon_{xx}^2 + 2\varepsilon_{xz}^2 \right) \cos \sqrt{2} \varepsilon_{xz} \left( -\cos \sqrt{2} \varepsilon_{xz} + \cos \left( \theta - \sqrt{2} \varepsilon_{xz} \right) \cos \theta \right) \end{aligned} \right]
\end{aligned} \quad (10)$$

for the case where the director rotation is in course. In the case where no shear is present the free energy reduces still more to

$$\begin{aligned}
F = & 6c_1 \varepsilon_{xx}^2 - \frac{1}{2} \varepsilon_0 \Delta \varepsilon_a E_n^2 (1 - \varepsilon_{xx})^4 \sin^2 \theta + \frac{1}{2} D_1 \sin^2 \theta + D_1^{(2)} \sin^4 \theta \\
& + D_2 \varepsilon_{xx} \sin^2 \theta + 2D_2^{nw} \varepsilon_{xx} \sin^2 \theta + D_2^{(2)} \varepsilon_{xx}^2 \sin^2 \theta - 4D_2^{nw,(2)} \varepsilon_{xx}^2 \sin^2 \theta \quad (11)
\end{aligned}$$

In Eq. (11) the true electric field  $E$  has been expressed in terms of the nominal field  $E_n$ .

b) If the displacement is taken as the input, then the following *ansatz* can be assumed for homogeneous solutions

$$u_x = Ax + Tz; \quad u_y = Ay + Tz; \quad u_z = Cz \quad (12)$$

where the amplitudes, which are numerical coefficients,  $A$ ,  $C$ ,  $T$ , reflect the strain deformation of the elastomer.

Under the hypothesis of incompressibility in Eulerian coordinates one obtains

$$C = -\frac{A(2-A)}{(1-A)^2} \quad (13)$$

which in the linear regime reduces to

$$C = -2A \quad (14)$$

After the pertinent calculations, retaining only linear terms in the strain tensor, and after deleting higher order (or combined products) than three in  $A$  and  $T$  one can write a new formal expression for the specific free energy as follows

$$F = c_1 (6A^2 + T^2) + \frac{1}{2} D_1 \sin^2 \left( \theta - \frac{\sqrt{2}}{2} T \right) + D_1^{(2)} \sin^4 \left( \theta - \frac{\sqrt{2}}{2} T \right) +$$

$$\begin{aligned}
& + \frac{1}{2} D_2 \left\{ \begin{aligned} & 2A \sin \theta \left( \sin \theta - \cos \left( \theta - \frac{\sqrt{2}}{2} T \right) \sin \frac{\sqrt{2}}{2} T \right) \\ & + \sqrt{2} T \left( \sin 2\theta - \cos \left( \theta - \frac{\sqrt{2}}{2} T \right) \sin \left( \theta + \frac{\sqrt{2}}{2} T \right) \right) + \\ & - 2(2A + T^2) \cos \theta \left( \cos \theta - \cos \left( \theta - \frac{\sqrt{2}}{2} T \right) \cos \frac{\sqrt{2}}{2} T \right) \end{aligned} \right\} + \\
& + \frac{1}{2} D_2^{nw} \left\{ \begin{aligned} & 2A \sin \frac{\sqrt{2}}{2} T \left( -\sin \frac{\sqrt{2}}{2} T + \cos \left( \theta - \frac{\sqrt{2}}{2} T \right) \sin \theta \right) + \\ & + \sqrt{2} T \left( -\sin \sqrt{2} T + \cos \left( \theta - \frac{\sqrt{2}}{2} T \right) \sin \left( \theta + \frac{\sqrt{2}}{2} T \right) \right) + \\ & - 2(2A + T^2) \cos \frac{\sqrt{2}}{2} T \left( -\cos \frac{\sqrt{2}}{2} T + \cos \left( \theta - \frac{\sqrt{2}}{2} T \right) \cos \theta \right) \end{aligned} \right\} + \\
& + \frac{1}{4} D_2^{(2)} \left\{ \begin{aligned} & 2T^2 \sin^2 \left( \theta - \frac{\sqrt{2}}{2} T \right) + 4A^2 \sin \theta \left( \sin \theta - \cos \left( \theta - \frac{\sqrt{2}}{2} T \right) \sin \frac{\sqrt{2}}{2} T \right) + \\ & + 16A^2 \cos \theta \left( \cos \theta - \cos \left( \theta - \frac{\sqrt{2}}{2} T \right) \cos \frac{\sqrt{2}}{2} T \right) + \\ & - 2\sqrt{2} AT \left( \sin 2\theta - \cos \left( \theta - \frac{\sqrt{2}}{2} T \right) \sin \left( \theta + \frac{\sqrt{2}}{2} T \right) \right) \end{aligned} \right\} + \\
& + \frac{1}{4} D_2^{nw,(2)} \left\{ \begin{aligned} & -2T^2 \sin^2 \left( \theta - \frac{\sqrt{2}}{2} T \right) + 4A^2 \sin \frac{\sqrt{2}}{2} T \left( -\sin \frac{\sqrt{2}}{2} T + \cos \left( \theta - \frac{\sqrt{2}}{2} T \right) \sin \theta \right) + \\ & + 16A^2 \cos \frac{\sqrt{2}}{2} T \left( -\cos \frac{\sqrt{2}}{2} T + \cos \left( \theta - \frac{\sqrt{2}}{2} T \right) \cos \theta \right) + \\ & - 2\sqrt{2} AT \left( -\sin \sqrt{2} T + \cos \left( \theta - \frac{\sqrt{2}}{2} T \right) \sin \left( \theta + \frac{\sqrt{2}}{2} T \right) \right) \end{aligned} \right\} - \\
& - \frac{1}{2} \varepsilon_0 \Delta \varepsilon_a E_n^2 (1 - A)^4 \sin^2 \theta \tag{15}
\end{aligned}$$

If no rotation of the network occurs  $\beta = T = 0$ . Furthermore  $\varepsilon_{xx} = \varepsilon_{yy}$  and  $\varepsilon_{zz} = C = -2A$ .

The resulting free energy is given by

$$\begin{aligned}
F &= 6c_1 A^2 + \\
& + \left[ 0.5D_1 + D_1^{(2)} \sin^2 \theta + D_2 A + 2D_2^{nw} A + D_2^{(2)} A^2 - 4D_2^{nw,(2)} A^2 - \frac{1}{2} \varepsilon_0 \Delta \varepsilon_a E_n^2 (1 - A)^4 \right] \sin^2 \theta \tag{16}
\end{aligned}$$

which is formally identical to Eq. (11).

### Stress-strain Relationship Predicted by the Model

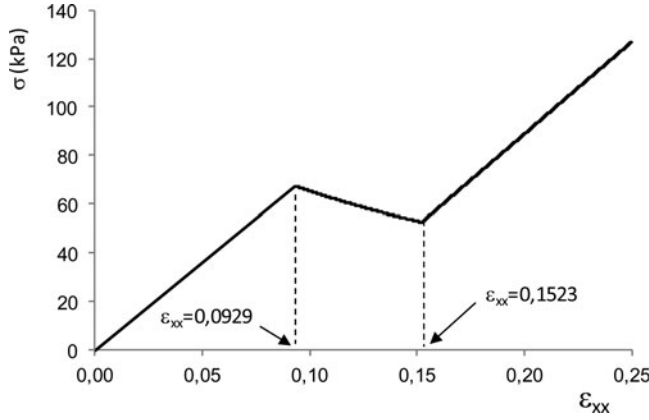
The following parameters will be taken for the calculations

$$\begin{aligned}
c_1 &= 121 \times 10^3 \text{ Jm}^{-3}, D_1 = 22.9 \times 10^3 \text{ Jm}^{-3}, D_1^{(2)} = 3.5 \times 10^3 \text{ Jm}^{-3}, \\
D_2 &= -42 \times 10^3 \text{ Jm}^{-3}, D_2^{nw} = -42.2 \times 10^3 \text{ Jm}^{-3}, \\
D_2^{(2)} &= -53.5 \times 10^3 \text{ Jm}^{-3}, D_2^{nw,(2)} = -22 \times 10^3 \text{ Jm}^{-3}
\end{aligned}$$

a) Case where neither shear deformation nor bifurcation occurs.

From Eq. (10) and the conditions given by Eq. (8) and the remaining equilibrium condition

$$\frac{\partial F}{\partial \varepsilon_{zz}} = 0 \tag{17}$$



**Figure 2.** Stress-strain curve in absence of electric field and shear.

one obtains for incompressible materials  $\varepsilon_{xx} = \varepsilon_{yy} = \frac{\sigma}{6c_1}$ ,  $\varepsilon_{zz} = -\frac{\sigma}{3c_1}$ , for  $\theta = \beta = 0$ , as in linear elasticity theory.

$$\varepsilon_{xx} = \varepsilon_{yy} = \frac{\sigma - \frac{1}{2}D_2 - D_2^{nw}}{6c_1 + D_2^{(2)} - 4D_2^{nw,(2)}},$$

$$\varepsilon_{zz} = -2 \frac{\sigma - \frac{1}{2}D_2 - D_2^{nw}}{6c_1 + D_2^{(2)} - 4D_2^{nw,(2)}},$$

for

$$\theta = \pi/2, \beta = 0 \quad (18)$$

From (11) or (16) and in absence of electric field  $[\sigma_{xx} (= \sigma_{yy}) = \frac{1}{2} \frac{\partial F}{\partial \varepsilon_{xx}}]$  one obtains

$$\sigma_{xx} = 6c_1 \varepsilon_{xx}, \quad \varepsilon_{xx} \leq 0.0929$$

$$\sigma_{xx} = 6c_1 \varepsilon_{xx} - \left[ 0.5D_2 + D_2^{nw} + \varepsilon_{xx} \left( D_2^{(2)} - 4D_2^{nw,(2)} \right) \right] \times$$

$$\times \left[ \frac{D_1 + 2\varepsilon_{xx} \left( D_2 + 2D_2^{nw} + D_2^{(2)} \varepsilon_{xx} - 4D_2^{nw,(2)} \varepsilon_{xx} \right)}{4D_1^{(2)}} \right], \quad 0.0929 \leq \varepsilon_{xx} \leq 0.1523$$

$$\sigma_{xx} = 6c_1 \varepsilon_{xx} + \left[ 0.5D_2 + D_2^{nw} + \varepsilon_{xx} \left( D_2^{(2)} - 4D_2^{nw,(2)} \right) \right], \quad \varepsilon_{xx} \geq 0.1523 \quad (19)$$

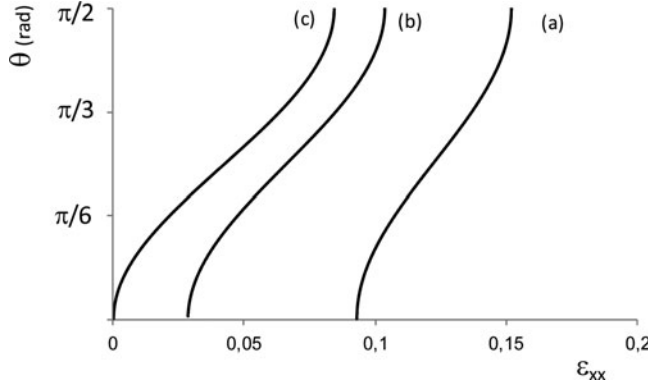
Limiting values for the strain,  $\varepsilon_{xx}$  in Figure 2, (0.0929, 0.1523) can be obtained from a stability analysis.

The electric field tends to reduce the threshold strain, and in fact for a nominal electric field of  $E_n = 22.744 \text{ kV/mm}$ . and taking  $\Delta \varepsilon_a = 5$  the threshold strain reduces to zero. A plot of  $\theta$  vs.  $\varepsilon_{xx}$

$$\theta = \sin^{-1} \sqrt{\frac{\varepsilon_o \Delta \varepsilon_a E_n^2 (1 - \varepsilon_{xx})^4 - D_1 - 2\varepsilon_{xx} \left( D_2 + 2D_2^{nw} + D_2^{(2)} \varepsilon_{xx} - 4D_2^{nw,(2)} \varepsilon_{xx} \right)}{4D_1^{(2)}}} \quad (20)$$

is shown in Figure 3.





**Figure 3.** Direct rotation angle as a function of the strain for three different electric fields (a)  $E = 0$  kV/mm (b)  $E = 20$  kV/mm (c)  $E = 22.74$  kV/mm.

b) Case where shear is present.

In this case it is convenient start from the Eq. (15). On account of the dependence of the free energy of  $T$  (or  $\beta$ ) and  $A$  the two equilibrium conditions are now

$$\frac{\partial F}{\partial \theta} = \frac{\partial F}{\partial T} = 0 \quad (21)$$

The corresponding algebra implied in the calculations becomes quite complex. However, in order to test the stability of the system, in absence of electric field, the signs of the minors of the following matrix

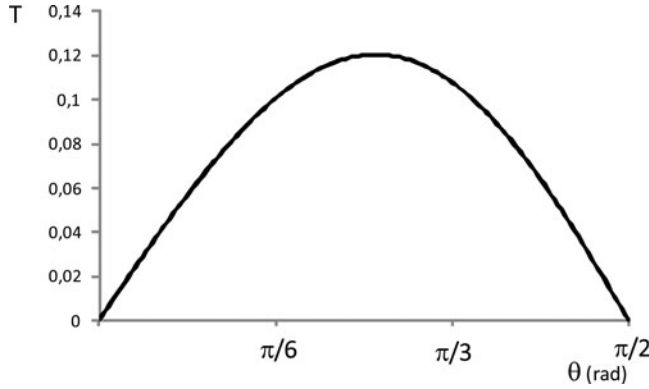
$$\begin{pmatrix} \frac{\partial^2 F}{\partial T^2} & \frac{\partial^2 F}{\partial T \partial \theta} \\ \frac{\partial^2 F}{\partial \theta \partial T} & \frac{\partial^2 F}{\partial \theta^2} \end{pmatrix} \quad (22)$$

are considered for the two extreme cases  $\theta = T = 0$  and  $\theta = \pi/2, T = 0$  according to the strategy outlined in [8]. If both minors are positive (negative), then the system is stable (unstable). After the pertinent calculations one obtains

$$\begin{aligned} & 4c_1 \left( D_1 + 2D_2 A + 4D_2^{nw} A + 2D_2^{(2)} A^2 - 8D_2^{nw,(2)} A^2 - \varepsilon_o \Delta \varepsilon_a E_n^2 (1 - A)^4 \right) - \\ & - \left( D_2 + D_2^{nw} - D_2^{(2)} A + D_2^{nw,(2)} A \right)^2 > 0, \quad \theta = T = 0 \\ & 2 \left( 2c_1 + D_2^{(2)} - D_2^{nw,(2)} \right) \left( D_1 + 2D_2 A + 4D_2^{nw} A + 2D_2^{(2)} A^2 - 8D_2^{nw,(2)} A^2 \right. \\ & \quad \left. + \varepsilon_o \Delta \varepsilon_a E_n^2 (1 - A)^4 \right) - \\ & + \left( D_2 + D_2^{nw} - D_2^{(2)} A + D_2^{nw,(2)} A \right)^2 > 0, \quad \theta = \pi/2, T = 0 \end{aligned} \quad (23)$$

In absence of electric field the following values for  $A$  are respectively obtained

$$A(\theta = T = 0) = 0.03445, 3.578 \text{ and } A(\theta = \pi/2, T = 0) = 0.1563, 3.563.$$



**Figure 4.** Shear as a function of the rotation angle for  $A = 0.1$ .

The electric field tends to diminish the lower figures found. In fact, when  $\Delta\epsilon_a = 5$ , an electric field of  $E = 1.365 \times 10^7 \text{ V m}^{-1}$  provokes the rotation of the director in absence of mechanical effects.

A rough estimation of the dependence of  $T$  upon the rotation angle can be obtained by assuming the following approximations:  $\sin(\theta - \beta) \approx \sin \theta$ ,  $\cos(\theta - \beta) \approx \cos \theta$ . This is justified if the rotation angle of the network is small in comparison with the rotation angle of the director. The pertinent calculations lead to (see Figure 4)

$$T = -\frac{\sqrt{2}}{2} \frac{\left(D_2 + D_2^{nw} - D_2^{(2)}A - D_2^{nw,(2)}A\right) \sin \theta \cos \theta}{2c_1 + \left(D_2^{(2)} - D_2^{nw,(2)}\right) \sin^2 \theta} \quad (24)$$

At a first approach the angle for which  $T$  attains to a maximum is independent of  $A$  and results  $47^\circ$  for  $T_{\max} = 0.120$ .

## Discussion and Conclusions

A continuum model to describe the macroscopic behaviour of NSCLSCE under biaxially applied mechanical and electrical force fields has been used. The reason for this choice was to analyse the actual behaviour of these elastomers that show a non linear plateau together with a non-vanishing shear modulus. The non linear terms used in our analysis are specifically related to the relative rotations of the network and the director. In contrast with previous work [5, 6], an electric field has been also applied in the same directions that the biaxial mechanical stresses. The electric field tends to reduce the value of the critical stresses appearing in the different analyzed cases because it helps the rotation of the director. The physical picture of the present problem is that when the rotation angle of the director starts to move from its initial value ( $\theta = 0$ ) the angle corresponding to the network rotation first increases from  $\beta = 0$  to a certain maximum value and then diminishes once more to  $\beta = 0$  when the director rotation is completed ( $\theta = \pi/2$ ).

Disregarding the network rotation the effect of the bifurcation in incompressible materials under electric fields has been examined. If no bifurcation is possible, the equality of the strains along the x- and y-axis is a direct consequence of the equality of the biaxial loads. Then equations for the principal strains corresponding to the starting and finishing director rotation are obtained for the case of incompressible materials. Implications of the

equilibrium equations are the equality of the meridian angles for the director and the network and the equality of the two shear terms. The two possible meridian angles obtained as well as the appearance in the relationship between the network meridian angle and the shear terms of two signs indicates the existence of a further bifurcation close to the starting the director rotation.

Finally, a stability analysis has been carried out following the route outlined in Ref. [8]. Stability threshold together with the corresponding critical values for the stress has been found for some particular cases. A rough estimation of the dependence of the shear  $S$  vs.  $\theta$  should also be obtained.

In this way, the more characteristic experimental facts experimentally observed [2] have been reproduced. In particular the curve corresponding to the stress-strain expression, shown in the Figure 2, is qualitatively consistent with previous result obtained by Urayama et al. [9].

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