# Electro-mechanical coupling in nematic elastomers: statics and dynamics

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SUMMARY. New analytical and computational results on the static and dynamic response of nematic elastomers to applied electric fields and mechanical loads are reviewed, and compared with the available experimental evidence.

## 1  INTRODUCTION

Nematic elastomers display a peculiar mechanical response arising from the interplay between the electro-optical properties of liquid crystals and the elastic properties of a rubbery solid. They have been the subject of numerous studies in the recent literature, see [21] and the many references cited therein. Envisaged by the late P.G. de Gennes [5], nematic elastomers were synthesized at the end of the 80s [22], [15]. Early experiments [16], [17] have immediately shown their potential as materials for soft actuators (large spontaneous deformations which, as it is now known, can be activated thermally, by electric fields, or UV light). At the same time, these experiments have revealed a very rich and complex behavior: unusually soft elastic response to uniaxial stretching, akin to the phenomenon of super-elasticity in shape memory alloys, and stripe domain instabilities, which are reminiscent of mechanical twinning.

The interest in the theoretical understanding of the behavior of nematic elastomers has been strong and sustained, starting from the pioneering paper of de Gennes [6], and including [14], [1], [7], [9], [10], [2], [3], [4].

The goal of this paper is to present some new analytical and computational results on the static and dynamic response of nematic elastomers to applied electric fields and mechanical loads. As discussed in [11], these results can be used to understand both the classical stretching experiments in [16], [17] and the more recent experimental measurements contained in [18], [19], and [12].

## 2  THE BASIC EXPERIMENTAL EVIDENCE

Electro-opto-mechanical effects arise in nematic elastomers from the coupling between nematic order and elasticity. Variable optical properties reflect the variability of nematic order, which controls dielectric anisotropy. Thus an applied electric field may reorient the nematic director in a free-standing film, inducing both a change of optical properties and spontaneous deformation. This experiment is described in [12], and it illustrates the *direct* electro-opto-mechanical effect.

Since nematic elastomers are not piezolectric, there is no inverse electro-opto-mechanical effect in the sense that one cannot generate electric fields through stresses. However, thanks to the coupling between elastic and nematic degrees of freedom, one can achieve director reorientation through mechanical stretching, hence an *inverse* opto-mechanical effect. The ability of the material to accommodate externally imposed stretches by rearranging internal degrees of freedom leads to

an unusually soft response. The most striking stretching experiments are those in which the sample is somewhat constrained, for example by rigid clamps, see [16], [17]. In this case one observes complex optical and deformation patterns, typically in the form of narrow bands (*stripe domains*), accompanied by surprisingly low build-up of internal stresses (*soft elasticity*).

## 3 THE MODEL

### 3.1 *Elastic energies*

For the sake of simplicity, we will illustrate the model starting from an idealized case, namely, the ideally soft material arising from isotropic elasticity. We later discuss more realistic anisotropic models, following [11].

Specifying an energy density for an elastic material amounts to identifying its natural states (i.e., the states of minimal energy density) and the way the energy grows away from this set. The first prescription is rather generic, and is strongly guided by material symmetry. The second one is much more material-specific since it determines, in particular, all the elastic moduli and all the nonlinearities of the stress-strain response.

We denote by $\mathbf{u}(x) = \mathbf{y}(x) - x$ the displacement at a point $x$ of the body $\Omega$, where $\mathbf{y}$ is the deformation. We write $\mathbf{F} = \nabla \mathbf{y}$ for the deformation gradient, and $\mathbf{B} = \mathbf{F}\mathbf{F}^T$ for the left Cauchy-Green strain tensor. We denote the nematic director, a unit vector field on $\Omega$, by $\mathbf{n}$ and set $\mathbf{N} := \mathbf{n} \otimes \mathbf{n}$, where $\otimes$ is the tensor product defined by $(\mathbf{a} \otimes \mathbf{b})\mathbf{v} = (\mathbf{b} \cdot \mathbf{v})\mathbf{a}$ for all $\mathbf{a}$, $\mathbf{b}$. The elastic energy describing an (incompressible) ideally soft nematic elastomer is

$$W(\mathbf{F}, \mathbf{N}) = \frac{1}{2}\mu \, \mathbf{B} \cdot \mathbf{L}^{-1}(\mathbf{N}) \tag{1}$$

where

$$\mathbf{L}(\mathbf{N}) := a^{\frac{2}{3}} \, \mathbf{N} + a^{-\frac{1}{3}} (\mathbf{I} - \mathbf{N}) \tag{2}$$

while $\mu > 0$ and $a > 1$ are material parameters. Energy (1) is isotropic, and it describes a material with a large set of natural states. These are all the states of deformation such that the left Cauchy-Green tensor $\mathbf{B}$ matches $\mathbf{L}(\mathbf{N})$. They are volume-preserving uniaxial deformation with (current) direction of largest principal stretch aligned with the (current) direction of the nematic director $\mathbf{n}$.

While very useful fron the conceptual point of view, energy (1) leads to predictions of the mechanical response that are not satisfactory. A minimal correction comes by recognizing that the actual material has a memory of a distinguished direction, namely, the orientation of the nematic molecules during the crosslinking process. Thus, their response should be transversely isotropic, with preferred axis given by the director orientation at cross-linking $\mathbf{n}_r$, rather than being isotropic. We consider two model anisotropic enegies, namely,

$$W_\alpha(\mathbf{F}, \mathbf{N}) = \frac{1}{2}\alpha\mu(1 - \mathbf{N} \cdot \mathbf{N}^*(\mathbf{F})) + W(\mathbf{F}, \mathbf{N}) \tag{3}$$

and

$$W_\beta(\mathbf{F}, \mathbf{N}) = \frac{1}{2}\beta\mu\mathbf{C} \cdot \mathbf{L}^{-1}(\mathbf{N}_r) + W(\mathbf{F}, \mathbf{N}) \,. \tag{4}$$

Here $\mathbf{C} = \mathbf{F}^T\mathbf{F}$ is the right Cauchy-Green tensor, $\alpha$ and $\beta$ are two positive dimensionless material parameters, $\mathbf{N}_r = \mathbf{n}_r \otimes \mathbf{n}_r$, and finally

$$\mathbf{N}^* := \mathbf{n}^* \otimes \mathbf{n}^*, \quad \mathbf{n}^* = \mathbf{n}^*(\mathbf{F}) := \frac{\mathbf{F}\mathbf{n}_r}{|\mathbf{F}\mathbf{n}_r|} \tag{5}$$

describes the current (i.e., after deformation) orientation of $\mathbf{n}_r$.

In the regime of small strains and large director rotations, denoting by $\mathbf{E} = (\nabla\mathbf{u} + \nabla\mathbf{u}^T)/2$ the infinitesimal strain, the energies above are appoximated by the following expressions

$$\Phi(\mathbf{E}, \mathbf{N}) = \mu(\mathbf{E} - \mathbf{E}_0(\mathbf{N})) \cdot (\mathbf{E} - \mathbf{E}_0(\mathbf{N})) \,, \tag{6}$$

$$\Phi_\alpha(\mathbf{E}, \mathbf{N}) = \frac{1}{2}\alpha\mu(1 - \mathbf{N} \cdot \mathbf{N}_r) + \Phi(\mathbf{E}, \mathbf{N}) \,, \tag{7}$$

$$\Phi_\beta(\mathbf{E}, \mathbf{N}) = \beta\mu(\mathbf{E} - \mathbf{E}_0(\mathbf{N}_r)) \cdot (\mathbf{E} - \mathbf{E}_0(\mathbf{N}_r)) + \Phi(\mathbf{E}, \mathbf{N}) \,, \tag{8}$$

where

$$\mathbf{E}_0(\mathbf{N}) := \frac{1}{2}\gamma(3\mathbf{N} - \mathbf{I}) \,, \quad \gamma := a^{\frac{2}{3}} - 1 \,. \tag{9}$$

In what follows, we will work within the setting of the geometrically linear theory and use the small strain energy densities (6), (7), and (8).

### 3.2 *Applied electric fields and total energy*

The elastic energy is not the only contribution. In addition, one typically also includes a curvature elasticity term (Frank energy) penalizing spatial variations of the nematic order, and an electrostatic energy giving the total work by the batteries to mantain a prescribed electric potential difference between selected points of the sample. In the isotropic case, we are led to the following free-energy functional

$$\mathscr{E}(\mathbf{u}, \mathbf{n}, \varphi) = \frac{1}{2} \int_\Omega \big( k_F |\nabla\mathbf{n}|^2 + \Phi(\mathbf{E}, \mathbf{N}) \big) - \frac{1}{2} \int_\Omega \big( \varepsilon_{\mathrm{o}}(\mathbb{D}(\mathbf{N})\,\nabla\varphi) \cdot \nabla\varphi \big) \tag{10}$$

where $k_F > 0$ is Frank constant, $\varepsilon_{\mathrm{o}}$ is the free-space permittivity, $\varphi$ is the electric potential, and $\mathbb{D}$ is the dielectric tensor. We take

$$\mathbf{d} := -\varepsilon_{\mathrm{o}}\mathbb{D}(\mathbf{N})\nabla\varphi = -\varepsilon_{\mathrm{o}}\big( \varepsilon_\perp \nabla\varphi + \varepsilon_a(\nabla\varphi \cdot \mathbf{n})\mathbf{n} \big) \tag{11}$$

where $\mathbf{d}$ is the electric displacement, while $\varepsilon_\perp$ and $\varepsilon_a > 0$ are dielectric coefficients. We are omitting here the potential of applied body forces or traction, since these are either neglected or zero in the simple experiments we consider in what follows. In the anisotropic cases, we denote the free-energy by either $\mathscr{E}_\alpha$ or $\mathscr{E}_\beta$: their expressions are obtained from (10) by replacing $\Phi$ with $\Phi_\alpha$ and $\Phi_\beta$.

### 3.3 *Dynamics*

The simplest nontrivial dynamics is obtained from the free–energy functional (10) by assuming insantaneous relaxation to equilibrium of the electric and elastic variables, and a viscous (over-damped) dynamics for the nematic director. Formally, we write

$$0 = \frac{\delta\mathscr{E}}{\delta\varphi} \,, \tag{12}$$

$$0 = \frac{\delta\mathscr{E}}{\delta\mathbf{u}} \,, \tag{13}$$

$$\frac{1}{m}\,\dot{\mathbf{n}} \times \mathbf{n} = -\frac{\delta\mathscr{E}}{\delta\mathbf{n}} \times \mathbf{n} \,, \tag{14}$$

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where $m > 0$ is a mobility parameter and the right hand–sides contain the variational derivative of $\mathscr{E}$ with respect to $\varphi$, $\mathbf{u}$, and $\mathbf{n}$. More explicitly, the equations above read

$$0 = \operatorname{div} \mathbf{d}\,, \tag{15}$$

$$0 = \operatorname{div} \mathbf{S}\,, \tag{16}$$

where $\mathbf{S} = -p\mathbf{I} + \mu\mathbf{E}_d$ is the stress tensor (here $p$ is the pressure, a reaction to the incompressibility constraint, and $\mathbf{E}_d$ is the deviatoric part of $\mathbf{E}$) and, finally,

$$\frac{1}{m}\,\dot{\mathbf{R}}\mathbf{R}^\top = [\,\mathbf{S}\,, \mathbf{E}_0\,] + \frac{1}{2}\,\varepsilon_o\,\varepsilon_a\,[\,\nabla\varphi \otimes \nabla\varphi\,, \mathbf{n}\otimes\mathbf{n}\,] + \operatorname{skw}\left(\operatorname{div}\left(k_F\nabla\mathbf{n}\right)\otimes\mathbf{n}\right)\,. \tag{17}$$

In the last equation, $\mathbf{R}$ is a rotation parametrizing the current orientation of $\mathbf{n}$ with respect to $\mathbf{n}_r$, i.e., $\mathbf{n} = \mathbf{R}\mathbf{n}_r$, $[\mathbf{A}, \mathbf{B}] = \mathbf{A}\mathbf{B} - \mathbf{B}\mathbf{A}$ is the commutator of the matrices $\mathbf{A}$ and $\mathbf{B}$, and finally $\operatorname{skw}(\mathbf{A}) = (\mathbf{A} - \mathbf{A}^\top)/2$ denotes the skew-symmetric part of the matrix $\mathbf{A}$. Equation (17) is obtained from (14) by introducing the microspin $\dot{\mathbf{R}}\mathbf{R}^\top$ (notice that $\dot{\mathbf{R}}\mathbf{R}^\top\mathbf{n} = \dot{\mathbf{n}} = \boldsymbol{\omega} \times \mathbf{n}$, where $\boldsymbol{\omega}$ is the director angular velocity), and by representing vector products with the action of the corresponding skew tensors.

The structure of equation (17) reveals in a rather transparent way the conditions such that a spatially uniform director field $\mathbf{n}$ be in equilibrium. In particular, the condition $[\mathbf{S}, \mathbf{E}_0] = 0$ is satisfied if and only if the stress $\mathbf{S}$ and the spontaneous distorsion $\mathbf{E}_0(\mathbf{n})$ have the same principal directions, see [13, p. 12]. The condition $[\nabla\varphi \otimes \nabla\varphi, \mathbf{n} \otimes \mathbf{n}] = 0$ is satisfied if and only if $\mathbf{n}$ is either parallel or perpendicular to the electric field $\mathbf{e} = -\nabla\varphi$.

The equation governing the dynamics of the system in the anisotropic cases are obtained with a similar procedure, by replacing $\mathscr{E}$ in (12)–(14) with $\mathscr{E}_\alpha$ and $\mathscr{E}_\beta$.

### 4  NUMERICAL RESULTS

We use the finite element method to solve numerically the evolution equations (15), (16), and (17) (or their anisotropic counterparts) supplemented with initial and boundary conditions suitable to represent the key experiments described in the introduction.

The first one is the application of an electric field in direction perpendicular to $\mathbf{n}_r$ to a free-standing film whose director is initially parallel to $\mathbf{n}_r$. Using the dynamics associated with the free–energy $\mathscr{E}_\beta$, we let the system relax to the equilibrium state under an applied electric field. The plots describing director and strain (along $\mathbf{n}_r$) at equilibrium as functions of the applied voltage are given in Figure 1. The agreement between theoretical prediction and experimental measurements is quite convincing. The characteristic times of response to instantaneous switching on and off of the electric field can also be estimated, and they compare rather well with experiments.

We remark that the model based on the isotropic elastic energy $\Phi$ and the one based on the anisotropic energy $\Phi_\beta$ are unable to represent correctly the physical picture because, according to them, the director would not go back to the initial orientation $\mathbf{n}_r$ when the electric field is switched off, contrary to what is actually observed.

The second key experiment consists in stretching a film with rigid clamps in direction perpendicular to $\mathbf{n}_r$ and in the absence of an electric field, at various stretching rates. The resulting force–stretch curves for the case of the isotropic elastic energy density are given in Figure 2. The initial plateau, with small build up of forces at the clamps is due to the fact that the specimen can accommodate the imposed elengation by rotating the director towards the stretching direction. This reorientation process occurs in a nonhomogenous way throughout the sample, hence the formation of stripe domains.

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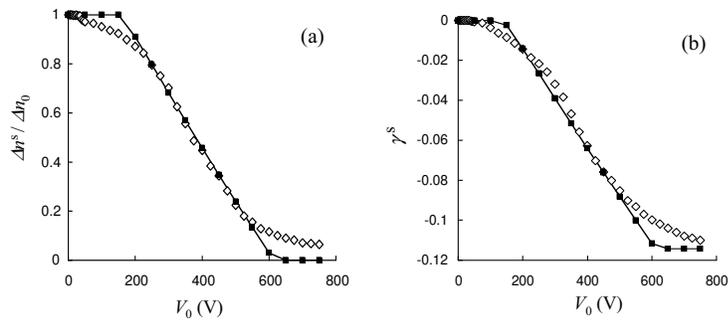
Figure 1: Numerical (filled symbols) versus experimental (open symbols) results for (a) effective optical birefringence and (b) strain along the initial director in the steady state as a function of applied voltage. The specimen is a free standing film under an electric field applied perpendicularly to the initial director orientation $\mathbf{n}_r$.
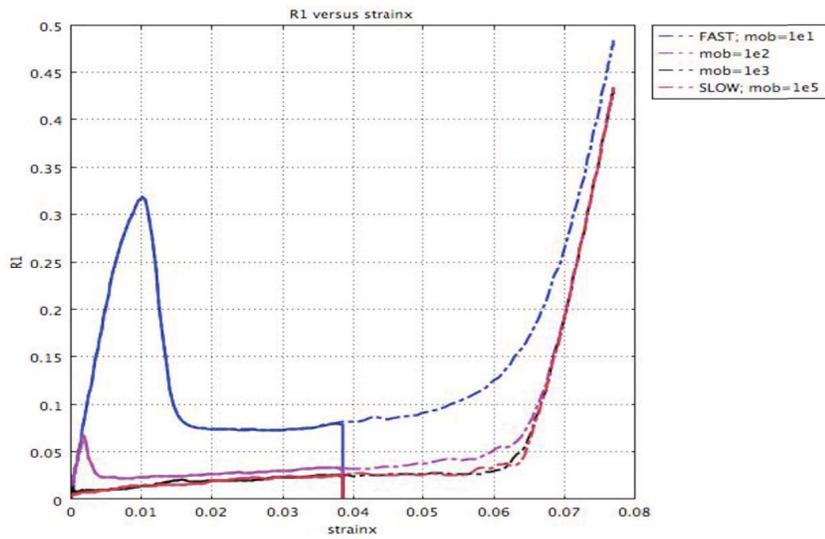
Figure 2: Force-strain curve in a purely mechanical stretching experiment at several stretching rates. The specimen is clamped and stretched in direction perpendicular to the initial director orientation $\mathbf{n}_r$.

Figure 2 shows a behavior which, as the loading rate is decreased, tends to an ideally soft response, with zero force until the director reorientation process is complete. In actual experiments, the response is not ideally soft and a threshold force must be overcome to trigger director reorientation. Both anisotropic models, either the one based on $\Phi_\alpha$ or the one based on $\Phi_\beta$, are able to reproduce this fact.

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