Chapter 12
Modeling of Dielectric Elastomers Accounting for Electrostriction by Means of a Multiplicative Decomposition of the Deformation Gradient Tensor

Elisabeth Staudigl, Michael Krommer, and Alexander Humer

Abstract Nonlinear modeling of inelastic material behavior by a multiplicative decomposition of the deformation gradient tensor is quite common for finite strains. The concept has proven applicable in thermoelasticity, elastoplacticity, as well as for the description of residual stresses arising in growth processes of biological tissues. In the context of advanced materials, the multiplicative decomposition of the deformation gradient tenser has been introduced within the fields of electro-elastic elastomers, shape-memory alloys as well as piezoelastic materials. In the present paper we apply this multiplicative approach to the special case of dielectric elastomers in order to account for the electrostrictive effect. Therefore, we seek to include the two main sources of electro-mechanical coupling in dielectric elastomers. These are elastostatic forces acting between the electric charges and electrostriction due to intramolecular forces of the material. In particular we intend to study the significance of electrostriction for the particular case of dielectric elastomers, in the form of a thin layer with two compliant electrodes.

12.1 Introduction

In this work we study constitutive modeling within the field of nonlinear electro-elasticity, with special application to dielectric elastomer films. Dielectric elastomers are capable of a mechanical response upon application of an external electric field, which is why they are commonly termed electro-active polymers (EAPs). These

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types of actuators are predicted to have a large variety of promising application fields, due to their characteristic to resist large strains while having a low stiffness and low density. These features, make them especially prone to smart or bio-inspired structural technologies e.g. artificial muscles. An EAP is typically assembled of a dielectric film sandwiched between two electrodes. When applying a potential difference to the electrode layers, they attract each other due to Coulomb forces, causing a pressure on the surface of the dielectric film, enforcing a deformation. Practical applications of this effect have been developed rather intensively, see examples such as soft and flexible keyboards (Xu et al, 2016) or artificial caterpillars demonstrated by various groups e.g. in SPIE (2017).

However, the full theoretical background of the deformation is yet not fully exploited. It has been reported, that the electric field might also interact with the dielectric layer as it gets polarized. This effect is reasoned in the micro-structure of the material, where different polarization mechanisms prevail. Therefore, we seek to include the two main sources of electro-mechanical coupling in dielectric elastomers as pointed out in Mehnert et al (2016). Polarization on the molecular level, called electrostriction, poses a process for which a full geometric nonlinear electro-mechanically coupled theory is necessary in order to model the impact of this effect adequately. Among the first theoretical works referring to electrostriction we refer to Zhao and Suo (2008), while earlier Zhenyi et al (1994) already presented experimental results.

Typical candidates for the material choice of dielectrics are silicon rubber and polyurethane elastomers, while special graft elastomers have been developed in the 1990s whose improved properties among high elastic-modulus count also the capability of nonlinear behavior at large strain regimes; therefore, a geometrical nonlinear framework is also necessary to model the mechanical behavior accurately. Theoretical works on the field of nonlinear electro-mechanical coupling date back into the 1950s. Toupin (1956) was among the first to address this field. A comprehensive presentation has later been given by Landau et al (2013); Maugin and Eringen (2012). Within the framework of nonlinear elasticity the book of Bonet and Wood (1997), has proven to be a handy reference, while special emphasis on the electric-coupling procedure can be found in the works of Dorfmann and Ogden (2005); Bustamante et al (2009a); Dorfmann and Ogden (2017) as well as in McMeeking and Landis (2004).

Constitutive modeling techniques incorporating the multiplicative decomposition of the deformation gradient tensor are quite common for finite strains. The concept has proven applicable in thermoelasticity, elastoplacticity, as well as for the description of residual stresses arising in growth processes of biological tissues (Lubarda, 2004). In the context of advanced materials, electro-elastic elastomers have been investigated in Skatulla et al (2012), shape-memory alloys in Arghavani et al (2010) and piezoelastic materials in Humer and Krommer (2015). A geometric nonlinear formulation on the constitutive modeling for the coupled electrostrictive-viscoelastic problem has been published by Ask et al (2012) using an additive decomposition of the free energy function. However, the comparison to experimental results, published by Diaconu and Dorohoi (2005); Diaconu et al (2006), suggest to put still further

investigations into this field. Bortot et al (2016); Ask et al (2015) made already use of the multiplicative decomposition of the deformation gradient tensor in their viscoelastic constitutive relation. Applications within the fields of electro-elastic coupled fields can be found in Zäh and Miehe (2015), and with special application to piezoelectricity in the works of Humer and Krommer (2015). Special interest is put on the physical bounds of the actuation at high strain magnitude. Phenomena called pull in or ceasing instability currently limit further increase in the actuation strains, as either imperfections or localization effects lead to the breakdown of the EAP. Analytical efforts to investigated these phenomena were made in Xu et al (2010), making use of the Hessian. They derived analytical expressions for the critical strain values, above which no stable stretch configuration exists any more.

This work is organized into five sections. First, we are going to introduce the basic electro-elastic coupled balance equations of continuum-mechanics, following Maugin and Eringen (2012) and the recent work of Humer et al (2017). The relevant electric quantities are reviewed and the Maxwell equations of electrostatics for dielectric materials are addressed. Within the second part, we introduce these quantities into the continuum mechanic theory in order to derive the spatial balance equations rendering the basis for the electro-mechanically coupled theory. With the spatial balance equations at hand, the material counterparts are obtained by which some preliminary constitutive relations are derived shortly, in order to demonstrate the general approach to the constitutive modeling framework. We close this part by introducing the incompressibility constraint using a Lagrange multiplier, which allows the physical interpretation of an electrostatic force. Higher order effects are then incorporated into the theory in Sect. 12.3. There, we extend the constitutive model by introducing the multiplicative decomposition of the deformation gradient. Results of the derivations show, that additional electro-mechanical coupling stresses increase the electro-elastic entanglement while still the overall physical relations can be retained. Section 12.4 provides additional background on the electrostrictive effect, unveiling the approach to include this effect into the constitutive model. In Sect. 12.5, we finally apply the resulting equations to the simple example of a homogeneous in-plane deformation of a plate. We choose this example on the one hand, as it allows for a comparison to experimental results, while on the other hand theoretical investigations allow for generalized statements on the impact of the electrostrictive effect.

12.2 Electromechanical Coupling by Electrostatic Force

12.2.1 Kinematics in Nonlinear Elasticity

Beginning with the kinematic quantities required in the framework of nonlinear continuum mechanics, we consider a material body, whose material points in the undeformed reference configuration V_r are denoted by upper case letters $\mathbf{X}$. Its bound-

ary is referred as ∂V_r. We consider only quasi-static, time independent deformation, which lead to a deformed current configuration, denoted by V with the boundary ∂V, and assume that a mapping function χ exists, such that $\mathbf{x} = \chi(\mathbf{X})$ uniquely maps the position vector of the material point into the current configuration $\mathbf{x}$, see Fig. 12.1. Hence the deformation gradient tensor $\mathbf{F}$ can be defined with respect to χ given by,

$$\mathbf{F} = \nabla_0 \mathbf{x} = \nabla_0 \chi(\mathbf{X}). \tag{12.1}$$

∇_0 is the differential operator with respect to the reference configuration. The volume change throughout the configuration is defined by $J = \det\mathbf{F}$ assuming $J > 0$ holds.

Knowing the deformation gradient tensor $\mathbf{F}$ allows to introduce the nonlinear strain measures

$$\mathbf{B} = \mathbf{F} \cdot \mathbf{F}^T, \qquad \mathbf{C} = \mathbf{F}^T \cdot \mathbf{F}, \tag{12.2}$$

where $\mathbf{B}$ is referred to as the left, and $\mathbf{C}$ as the right Cauchy-Green Tensor.

Orientation and position of a deformed surface element are defined by the unit outward normal vector $\mathbf{n}$ on ∂V, while $\mathbf{t}$ is the force per unit area on ∂V, which allows to introduce the second rank Cauchy stress tensor $\boldsymbol{\sigma}$ through $\boldsymbol{\sigma}^T \cdot \mathbf{n} = \mathbf{t}$.

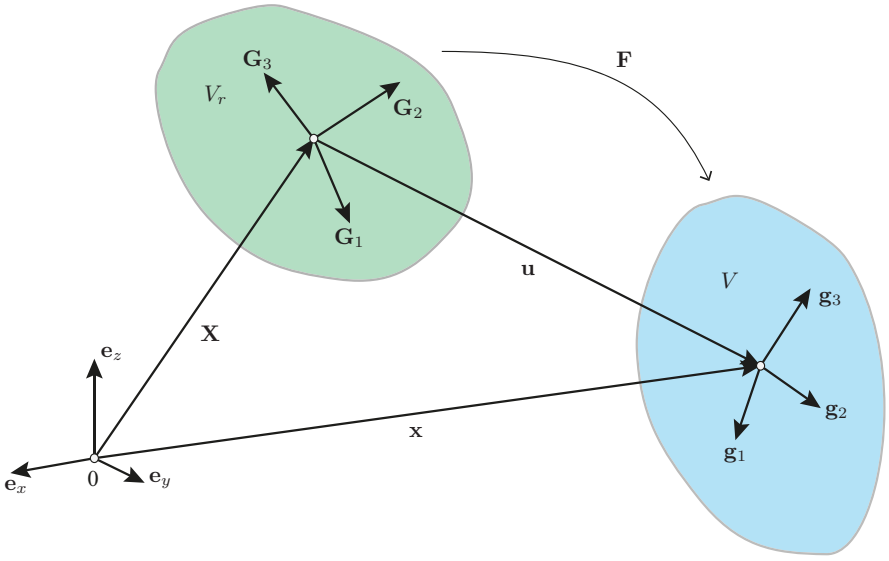

Fig. 12.1: Field mapping of a general deformable body.

12.2.2 Electro-Elastic Balance Laws

In order to derive the balance equations for the electro-elastic continuum, we start by defining the dependent variables within the micro-continuum, which upon statistical averaging over the continuum volume, lead to the relevant expressions of the macro-continuum. Within the continuum mechanic framework, only the quantities arising when applying external fields have to be taken into account, internal field quantities are incorporated by the concepts of electro-mechanical stress and internal energy. Upon application of an external electric field $\boldsymbol{e}$, charges q are encouraged to move slightly forming dipole-moments within the continuum; this process called polarization $\boldsymbol{p}$, see Fig. 12.2, is reflected by electrostatic volume force $\boldsymbol{f}^E$, the corresponding couple $\boldsymbol{c}^E$, and the power of the electrostatic force W^E:

$$\boldsymbol{f}^E = (\nabla \boldsymbol{e}) \cdot \boldsymbol{p}, \qquad \boldsymbol{c}^E = \boldsymbol{p} \times \boldsymbol{e}, \qquad W^E = \rho \boldsymbol{e} \cdot \frac{d}{dt}\left(\frac{\boldsymbol{p}}{\rho}\right) + \boldsymbol{f}^E \cdot \mathbf{v}. \tag{12.3}$$

12.2.2.1 Maxwell Equation and Electric Body Forces

In case of electrostatics for an ideal dielectric, terms with free charges and magnetic interactions can be dropped in the Maxwell equations, which then read:

$$\nabla \cdot \boldsymbol{d} = 0, \tag{12.4}$$

$$\nabla \times \boldsymbol{e} = \mathbf{0}; \tag{12.5}$$

respectively, the *Gauss Law* and the *Faraday Law* of electrostatics. Here, we already used the electric displacement vector, defined by $\boldsymbol{d} = \varepsilon_0 \boldsymbol{e} + \boldsymbol{p}$, in which ε_0 denotes the vacuum permittivity. One may find an exact solution for the *Faraday Law* immediately by engaging a scalar potential function Φ, which satisfies $\boldsymbol{e} = -\nabla\Phi$; ∇ is the differential operator with respect to the current configuration. Additionally, the fields $\boldsymbol{e}, \boldsymbol{p}$ and $\boldsymbol{d}$ have to satisfy the jump conditions

$$\mathbf{n} \cdot [\![\boldsymbol{d}]\!] = 0, \qquad \mathbf{n} \times [\![\boldsymbol{e}]\!] = \mathbf{0}. \tag{12.6}$$

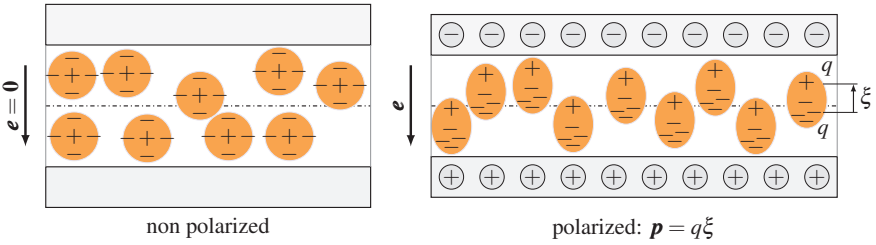

Fig. 12.2: Polarization of a continuum.

The bracketed terms denote jumps in the relevant field quantity. In case of vacuum the fields reduce to $\boldsymbol{d} = \varepsilon_0 \boldsymbol{e}$. Following Bustamante et al (2009b), the boundary conditions can be set as a function in $\boldsymbol{p}$:

$$\varepsilon_0 [\![\boldsymbol{e}]\!] = (\mathbf{n} \cdot \boldsymbol{p})\mathbf{n}, \qquad [\![\boldsymbol{d}]\!] = (\mathbf{n} \cdot \boldsymbol{p})\mathbf{n} - \boldsymbol{p}; \tag{12.7}$$

hence, by introducing the so called Maxwell stress which is present in the free field, the jump on the boundary ∂V of a material can be defined as:

$$[\![\boldsymbol{\sigma}^M]\!] \cdot \mathbf{n} = \frac{1}{2} \varepsilon_0^{-1} (\boldsymbol{p} \cdot \mathbf{n})^2 \mathbf{n}. \tag{12.8}$$

12.2.2.2 Conservation Laws

Turning now to the equations of the theory of electro-elasticity, we start with the macroscopic law of conservation of mass $m = \rho V$ in form of the *Continuity equation:*

$$\dot{m} = \frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \mathbf{v}) = 0, \tag{12.9}$$

where $\mathbf{v} = \dot{\mathbf{x}}$ denotes the current velocity of the continuum at the point $\mathbf{x}$.

For the electro-elastic coupling, the electrostatic body-force $\boldsymbol{f}^E$ of the continuum is first introduced into the *Balance of linear momentum*. Hence, the momentum of a body, which is balanced by body forces $\rho \boldsymbol{f}$ per unit volume, mechanical surface loads $\boldsymbol{t} = \boldsymbol{\sigma}^T \cdot \boldsymbol{n}$ per unit area, and the effect of the electrostatic force per unit volume $\boldsymbol{f}^E$ reads

$$\frac{d}{dt} \int_V \rho \mathbf{v} dV = \int_V (\rho \boldsymbol{f} + \boldsymbol{f}^E) dV + \int_{\partial V} \boldsymbol{t} dS \tag{12.10}$$

in its global form. Using the Gauss integral theorem, yields the local form,

$$\nabla \cdot \boldsymbol{\sigma} + \rho \boldsymbol{f} + \boldsymbol{f}^E - \rho \dot{\mathbf{v}} = \mathbf{0}. \tag{12.11}$$

In analogy to the pure mechanical case, also an electrostatic stress tensor $\boldsymbol{f}^E = \nabla \cdot \boldsymbol{\sigma}^E$ can be introduced. Adding the electrostatic stress tensor to the Cauchy stress tensor $\boldsymbol{\sigma}$, results in the total electro-mechanical stress tensor $\boldsymbol{\sigma}^{tot}$,

$$\boldsymbol{\sigma}^{tot} = \boldsymbol{\sigma} + \boldsymbol{\sigma}^E, \qquad \boldsymbol{\sigma}^E = \boldsymbol{e}(\varepsilon_0 \boldsymbol{e} + \boldsymbol{p}) - \frac{1}{2} \varepsilon_0 (\boldsymbol{e} \cdot \boldsymbol{e}) \mathbf{I}. \tag{12.12}$$

Furthermore, one has to incorporate the electric couple $\boldsymbol{c}^E$ in the *Balance of Moment of Momentum*,

$$\frac{d}{dt} \int_V \mathbf{x} \times \rho \mathbf{v} dV = \int_V \mathbf{x} \times (\rho \boldsymbol{f} + \boldsymbol{f}^E) dV + \int_V \boldsymbol{c}^E dV + \int_{\partial V} \mathbf{x} \times \mathbf{t} dS. \tag{12.13}$$

This yields, in contrast to the pure mechanical theory of elasticity, a non-symmetric mechanical Cauchy stress tensor. Using the identity

$$\nabla \cdot (\mathbf{x} \times \boldsymbol{\sigma}) = \mathbf{x} \times (\nabla \cdot \boldsymbol{\sigma}) + {}^3\boldsymbol{\varepsilon} \cdot\cdot \boldsymbol{\sigma}, \tag{12.14}$$

where ${}^3\boldsymbol{\varepsilon}$ is the third rank *Levi-Civita tensor*, and the Gauss integral theorem the local form eventually yields to the symmetry of the sum of the Cauchy stress tensor, and the polarization stress $\boldsymbol{\sigma}^P$:

$$\boldsymbol{c}^{\boldsymbol{E}} + {}^3\boldsymbol{\varepsilon} \cdot\cdot \boldsymbol{\sigma} = \mathbf{0} \quad , \qquad \boldsymbol{c}^{\boldsymbol{E}} = \boldsymbol{p} \times \boldsymbol{e} = {}^3\boldsymbol{\varepsilon} \cdot\cdot \boldsymbol{e}\boldsymbol{p} = {}^3\boldsymbol{\varepsilon} \cdot\cdot \boldsymbol{\sigma}^P. \tag{12.15}$$

The last equation states, that the negative antisymmetric part of the Cauchy stress tensor is identically the antisymmetric part of the dipole moment tensor $\boldsymbol{\sigma}^P$. Hence,

$$\operatorname{skew}\left(\boldsymbol{\sigma} + \boldsymbol{\sigma}^P\right) = \mathbf{0} \quad \text{and} \quad \boldsymbol{\sigma}^S = \boldsymbol{\sigma} + \boldsymbol{\sigma}^P, \tag{12.16}$$

where a symmetric stress tensor $\boldsymbol{\sigma}^S = (\boldsymbol{\sigma}^S)^T$ has been introduced. If we now use the last part of Eq. (12.16) and insert it into the electro-mechanical stress tensor, we find the famous Maxwell stress tensor $\boldsymbol{\sigma}^M$:

$$\boldsymbol{\sigma}^{tot} = \boldsymbol{\sigma}^S - \boldsymbol{\sigma}^P + \boldsymbol{\sigma}^E = \boldsymbol{\sigma}^S + \boldsymbol{\sigma}^M, \tag{12.17}$$

$$\boldsymbol{\sigma}^M = \boldsymbol{\sigma}^E - \boldsymbol{\sigma}^P = \varepsilon_0 \boldsymbol{e}\boldsymbol{e} - \frac{1}{2}\varepsilon_0 (\boldsymbol{e} \cdot \boldsymbol{e})\mathbf{I}. \tag{12.18}$$

Finally, in order to get hands on a thermodynamically consistent constitutive relation, the *Balance of Energy* for the electro-elastic body reads

$$\frac{d}{dt}\int_V \rho\left(\frac{1}{2}\mathbf{v}^2 + e\right) dV = \int_V \left[\left(\rho \boldsymbol{f} + \boldsymbol{f}^E\right) \cdot \mathbf{v} + \rho \boldsymbol{e} \cdot \dot{\boldsymbol{\pi}}\right] dV + \int_{\partial V} \mathbf{t} \cdot \mathbf{v} dS, \tag{12.19}$$

which upon using the balance of momentum Eq. (12.11), yields the local form:

$$\rho \dot{e} - \boldsymbol{\sigma} \cdot\cdot (\nabla \mathbf{v})^T - \rho \boldsymbol{e} \cdot \dot{\boldsymbol{\pi}} = 0, \tag{12.20}$$

where $\dot{\boldsymbol{\pi}} = \frac{d}{dt}\frac{\boldsymbol{p}}{\rho}$. Finally, following Maugin and Eringen (2012) we change the dependent variable by using a Legendre transform for the internal energy e in order to gain the Helmholtz free energy $\psi = e - \frac{1}{\rho}\boldsymbol{e} \cdot \boldsymbol{p}$

$$\rho \dot{\psi} + \frac{d}{dt}\boldsymbol{e} \cdot \boldsymbol{p} - \boldsymbol{\sigma} \cdot\cdot (\nabla \mathbf{v})^T - \rho \boldsymbol{e} \cdot \dot{\boldsymbol{\pi}} = 0, \tag{12.21}$$

which after differentiation gives the final form of the rate of free energy:

$$\rho \dot{\psi} = \boldsymbol{\sigma} \cdot\cdot (\nabla \mathbf{v})^T - \dot{\boldsymbol{e}} \cdot \boldsymbol{p}. \tag{12.22}$$

12.2.3 Lagrangian (Material) Framework

Within this part the framework of the derivation of the constitutive relations is presented in a generalized state, in order to present the overall procedure. We start by transforming the relations, which were obtained previously in the current (spatial) configuration into the reference configuration, resulting into the Lagrangian (material) framework. The transformation rules of the dependent variables read:

$$\rho_0 = \rho J, \tag{12.23}$$
$$\boldsymbol{\mathcal{P}} = J\mathbf{F}^{-1}\cdot\boldsymbol{p}, \tag{12.24}$$
$$\boldsymbol{\mathcal{E}} = \boldsymbol{e}\cdot\mathbf{F}, \tag{12.25}$$
$$\mathbf{S} = J\mathbf{F}^{-1}\cdot\boldsymbol{\sigma}\cdot\mathbf{F}^{-T}. \tag{12.26}$$

$\boldsymbol{\sigma}$ defines the Cauchy stress tensor and $\mathbf{S}$ the second Piola-Kirchoff stress tensor. $\boldsymbol{\mathcal{E}}$ and $\boldsymbol{\mathcal{P}}$ define the material electric field and polarization vector respectively. The electric displacement vector $\boldsymbol{d}$, transforms in the same manner as the polarization vector, using Nanson's formula $\mathbf{n}da = J\mathbf{F}^{-T}\cdot\mathbf{N}dA$:

$$\boldsymbol{\mathcal{D}} = J\boldsymbol{d}\cdot\mathbf{F}^{-T}, \quad \boldsymbol{\mathcal{D}} = \varepsilon_0\boldsymbol{\mathcal{E}}\cdot\mathbf{C}^{-1} + \boldsymbol{\mathcal{P}}. \tag{12.27}$$

Maxwell equations can be written as

$$\boldsymbol{d} = \varepsilon_0\boldsymbol{e} + \boldsymbol{p}, \qquad \nabla_0\cdot\boldsymbol{\mathcal{D}} = \nabla_0\cdot(\varepsilon_0\boldsymbol{\mathcal{E}}\cdot\mathbf{C}^{-1} + \boldsymbol{\mathcal{P}}) = 0, \tag{12.28}$$
$$\nabla_0\times\boldsymbol{\mathcal{E}} = \mathbf{0}, \tag{12.29}$$

while by carefully applying the volume, surface and line element transformation rules, on can find the electrostatic material force and couple:

$$\boldsymbol{\mathcal{F}}^E = (\nabla_0\boldsymbol{\mathcal{E}})\cdot\boldsymbol{\mathcal{P}}, \tag{12.30}$$
$$\boldsymbol{\mathcal{C}}^E = -\mathbf{F}^{-T}\cdot\boldsymbol{\mathcal{E}}\times\boldsymbol{\mathcal{P}}\cdot\mathbf{F}^T. \tag{12.31}$$

Next, we write the material form of the balance of momentum, while from the balance of moment of momentum, the material form of the polarization stress tensor is derived:

$$\rho_0\mathbf{v} = \nabla_0\cdot(\boldsymbol{S} + \boldsymbol{S}^E) + \rho_0\boldsymbol{f}, \tag{12.32}$$
$$\boldsymbol{S}^P = J\mathbf{F}^{-1}\cdot\boldsymbol{e}\boldsymbol{p}\cdot\mathbf{F}^{-T} = \mathbf{C}^{-1}\cdot\boldsymbol{\mathcal{E}}\boldsymbol{\mathcal{P}} \tag{12.33}$$

The material electrostatic stress tensor follows to

$$\boldsymbol{S}^E = \mathbf{C}^{-1}\cdot\boldsymbol{\mathcal{E}}(\varepsilon_0\boldsymbol{\mathcal{E}}\cdot\mathbf{C}^{-1} + \boldsymbol{\mathcal{P}}) - \frac{1}{2}\varepsilon_0 J(\boldsymbol{\mathcal{E}}\cdot\mathbf{C}^{-1}\cdot\boldsymbol{\mathcal{E}})\cdot\mathbf{C}^{-1} \tag{12.34}$$
$$= \mathbf{C}^{-1}\cdot\boldsymbol{\mathcal{E}}\boldsymbol{\mathcal{D}} - \frac{1}{2}\varepsilon_0 J(\boldsymbol{\mathcal{E}}\cdot\mathbf{C}^{-1}\cdot\boldsymbol{\mathcal{E}})\cdot\mathbf{C}^{-1}. \tag{12.35}$$

In order to obtain the material form of the free energy function, one has to incorporate the time rate of the right Cauchy-Green strain tensor $\dot{\mathbf{C}} = (\dot{\mathbf{F}}^T \cdot \mathbf{F} + \mathbf{F}^T \cdot \dot{\mathbf{F}})$, which allows to write the proper transformation of the gradient of the velocity vector $\nabla \mathbf{v} = \mathbf{F}^{-T} \cdot \frac{1}{2}\dot{\mathbf{C}} \cdot \mathbf{F}^{-1}$. Hence, the material rate of free energy function per unit mass $\dot{\psi}(\mathbf{C},\boldsymbol{\mathcal{E}})$ can be written as:

$$\rho_0 \dot{\psi} = \left(\mathbf{S} + \boldsymbol{\mathcal{P}}\boldsymbol{\mathcal{E}} \cdot \mathbf{C}^{-1}\right) \cdot\cdot \frac{1}{2}\dot{\mathbf{C}} - \boldsymbol{\mathcal{P}} \cdot \dot{\boldsymbol{\mathcal{E}}}. \tag{12.36}$$

In the material form of the problem the boundary conditions are obtained by transforming the second Piola Kirchoff stress tensor to its two-field tensor counterpart $\mathbf{P}^{tot} = \mathbf{F} \cdot \mathbf{S}^{tot}$,

$$\mathbf{N} \cdot [\![\mathbf{P}^{tot}]\!] = \mathbf{0}, \qquad \mathbf{N} \cdot [\![\boldsymbol{\mathcal{D}}]\!] = 0, \tag{12.37}$$

and by Faraday's law $\nabla_0 \times \boldsymbol{\mathcal{E}} = \mathbf{0}$,

$$\mathbf{N} \times [\![\boldsymbol{\mathcal{E}}]\!] = \mathbf{0}. \tag{12.38}$$

12.2.4 Constitutive Relations

In order to close the theory, the phenomenological properties of the material have to be taken into account. We consider only small gradients in the electric field and strains, which therefore allows to take the classical quadratic form of the generalized thermodynamic energy function, valid in electro-elastic bodies. The free energy function ψ is assumed to decompose additively into a mechanical part $\psi_{me}(\mathbf{C})$, and an electrical part $\psi_{el}(\mathbf{C},\boldsymbol{\mathcal{E}})$.

However, an additional term, called augmented free energy, motivated by the presence of ponderomotive forces in vacuum is additionally incorporated. This approach is suggested in Dorfmann and Ogden (2005). In order to make a clear distinction, we indicate the sum of all free energy functions with Ω,

$$\Omega = \psi + \psi_{aug} = \psi_{me}(\mathbf{C}) + \psi_{el}(\mathbf{C},\boldsymbol{\mathcal{E}}) + \psi_{aug}(\mathbf{C},\boldsymbol{\mathcal{E}}), \tag{12.39}$$

where the dependent variables of the augmentation term $\psi_{aug}(\mathbf{C},\boldsymbol{\mathcal{E}})$ are introduced in accordance to the electrical free energy. Writing the rate of the augmented free energy yields

$$\dot{\Omega} = \frac{\partial \psi_{me}}{\partial \mathbf{C}} \cdot\cdot \dot{\mathbf{C}} + \frac{\partial \psi_{el}}{\partial \mathbf{C}} \cdot\cdot \dot{\mathbf{C}} + \frac{\partial \psi_{aug}}{\partial \mathbf{C}} \cdot\cdot \dot{\mathbf{C}} + \frac{\partial \psi_{el}}{\partial \boldsymbol{\mathcal{E}}} \cdot \dot{\boldsymbol{\mathcal{E}}} + \frac{\partial \psi_{aug}}{\partial \boldsymbol{\mathcal{E}}} \cdot \dot{\boldsymbol{\mathcal{E}}}, \tag{12.40}$$

or after inserting Eq. (12.36)

$$\rho_0 \dot{\Omega} = \frac{1}{2}\left(\mathbf{S} + \boldsymbol{\mathcal{P}}\boldsymbol{\mathcal{E}} \cdot \mathbf{C}^{-1} + 2\rho_0 \frac{\partial \psi_{aug}}{\partial \mathbf{C}}\right) \cdot\cdot \dot{\mathbf{C}} - \left(\boldsymbol{\mathcal{P}} - \rho_0 \frac{\partial \psi_{aug}}{\partial \boldsymbol{\mathcal{E}}}\right) \cdot \dot{\boldsymbol{\mathcal{E}}}. \tag{12.41}$$

A comparison unveils

$$\frac{\partial \psi_{me}}{\partial \mathbf{C}} = \frac{1}{2\rho_0}\mathbf{S}, \qquad \frac{\partial \psi_{el}}{\partial \mathbf{C}} = \frac{1}{2\rho_0}\mathbf{S}^P, \qquad -\frac{\partial \psi_{el}}{\partial \boldsymbol{\mathcal{E}}} = \frac{1}{\rho_0}\boldsymbol{\mathcal{P}}, \tag{12.42}$$

$$\frac{\partial \psi_{aug}}{\partial \mathbf{C}} = \frac{1}{2\rho_0}\mathbf{S}^M, \quad -\frac{\partial \psi_{aug}}{\partial \boldsymbol{\mathcal{E}}} = \frac{1}{\rho_0}(\boldsymbol{\mathcal{D}} - \boldsymbol{\mathcal{P}}). \tag{12.43}$$

We obtain relations for the total electric displacement and the total stress expressed by simple addition:

$$\boldsymbol{\mathcal{D}} = -\rho_0 \frac{\partial \psi_{el}}{\partial \boldsymbol{\mathcal{E}}} - \rho_0 \frac{\partial \psi_{aug}}{\partial \boldsymbol{\mathcal{E}}} = -\rho_0 \frac{\partial \Omega}{\partial \boldsymbol{\mathcal{E}}}, \tag{12.44}$$

$$\mathbf{S}^{tot} = 2\rho_0 \frac{\partial \psi_{me}}{\partial \mathbf{C}} + 2\rho_0 \frac{\partial \psi_{el}}{\partial \mathbf{C}} + 2\rho_0 \frac{\partial \psi_{aug}}{\partial \mathbf{C}} = 2\rho_0 \frac{\partial \Omega}{\partial \mathbf{C}}. \tag{12.45}$$

Still the specific form of the energy functions is missing. For the mechanical part ψ_{me} any hyperelastic strain energy function can be used. In order to define the electrical energy function, we assume the energy to take a quadratic form in $\boldsymbol{\mathcal{E}}$, and start by transforming the heuristic relation $\boldsymbol{p} = \chi \boldsymbol{e}$ for the polarization vector given in the spatial framework into the material framework:

$$\boldsymbol{p} = \chi \boldsymbol{e}, \qquad \boldsymbol{\mathcal{P}} = J\chi \mathbf{C}^{-1} \cdot \boldsymbol{\mathcal{E}}, \tag{12.46}$$

$$\rho_0 \psi_{el} = -\frac{1}{2}\chi \varepsilon_0 \boldsymbol{\mathcal{E}} \cdot (\mathbf{C}^{-1} \cdot \boldsymbol{\mathcal{E}}), \tag{12.47}$$

where χ is the electric susceptibility. The augmentation term reads

$$\rho_0 \psi_{aug} = -\frac{1}{2}\varepsilon_0 J \boldsymbol{\mathcal{E}} \cdot (\mathbf{C}^{-1} \cdot \boldsymbol{\mathcal{E}}), \tag{12.48}$$

and by taking the derivative with respect to $\mathbf{C}$, while keeping in mind $\frac{\partial J}{\partial \mathbf{C}} = \frac{1}{2} J \mathbf{C}^{-1}$, we find

$$\frac{1}{2\rho_0}\mathbf{S}^P = \frac{\partial \psi_{el}}{\partial \mathbf{C}} = \frac{1}{2\rho_0}\chi \varepsilon_0 \mathbf{C}^{-1} \cdot \boldsymbol{\mathcal{E}}\boldsymbol{\mathcal{E}} \cdot \mathbf{C}^{-1}, \tag{12.49}$$

$$\frac{1}{2\rho_0}\mathbf{S}^M = \frac{\partial \psi_{aug}}{\partial \mathbf{C}} = \frac{1}{2\rho_0}\varepsilon_0 J \left(\mathbf{C}^{-1} \cdot \boldsymbol{\mathcal{E}}\boldsymbol{\mathcal{E}} - \frac{1}{2}\mathbf{I}(\boldsymbol{\mathcal{E}}\boldsymbol{\mathcal{E}} \cdot\cdot \mathbf{C}^{-1}) \right) \cdot \mathbf{C}^{-1}, \tag{12.50}$$

which are the constitutive relations for the polarization stress $\mathbf{S}^P$ and the material Maxwell stress tensor $\mathbf{S}^M$. Subsequently, we obtain the polarization vector by taking the derivative with respect to $\boldsymbol{\mathcal{E}}$,

$$\frac{1}{\rho_0}\boldsymbol{\mathcal{P}} = -\frac{\partial \psi_{el}}{\partial \boldsymbol{\mathcal{E}}} = \frac{1}{\rho_0}\chi \varepsilon_0 \mathbf{C}^{-1} \cdot \boldsymbol{\mathcal{E}}, \tag{12.51}$$

$$-\rho_0 \frac{\partial \psi_{aug}}{\partial \boldsymbol{\mathcal{E}}} = \varepsilon_0 J \mathbf{C}^{-1} \cdot \boldsymbol{\mathcal{E}}. \tag{12.52}$$

Here it should be noted, that the polarization stress $\mathbf{S}^P$ can directly be obtained by multiplication with $\boldsymbol{\mathcal{E}} \cdot \mathbf{C}^{-1}$. Gathering all terms one can find the constitutive relation

for the electric displacement vector $\boldsymbol{\mathcal{D}} = \boldsymbol{\mathcal{P}} + \varepsilon_0 J \mathbf{C}^{-1} \cdot \boldsymbol{\mathcal{E}}$ to

$$\rho_0 \dot{\Omega} = \left(\mathbf{S} + \boldsymbol{\mathcal{D}} \boldsymbol{\mathcal{E}} \cdot \mathbf{C}^{-1} - \frac{1}{2} \varepsilon_0 J \mathbf{C}^{-1} \cdot (\boldsymbol{\mathcal{E}} \boldsymbol{\mathcal{E}} \cdot \cdot \mathbf{C}^{-1}) \right) \cdot \cdot \frac{1}{2} \dot{\mathbf{C}} - \boldsymbol{\mathcal{D}} \cdot \dot{\boldsymbol{\mathcal{E}}}. \tag{12.53}$$

The bracketed term is the total second Piola-Krichoff stress tensor $\mathbf{S}^{tot}$, which is composed of the unsymmetric mechanical second Piola-Kirchoff stress tensor $\mathbf{S}$, the polarization stress $\mathbf{S}^P$ and the symmetric Maxwell stress $\mathbf{S}^M$.

This renders the classical version of electro-mechanical coupling most commonly used in the field of dielectric elastomers, where the major driving mechanism is given by the electrostatic force. In case of incompressible dielectric elastomers, the deformation gradient is oblige to $\det \mathbf{F} = J = 1$, hence $\dot{J} = 0$. Therefore, the constitutive relation for the total second Piola-Kirchoff stress tensor is constrained, and we account for the constraint by introducing a Lagrange multiplier p,

$$\mathbf{S}^{tot} = 2\rho_0 \frac{\partial \Omega}{\partial \mathbf{C}} + p \mathbf{C}^{-1}, \qquad \boldsymbol{\mathcal{D}} = -\rho_0 \frac{\partial \Omega}{\partial \boldsymbol{\mathcal{E}}} \tag{12.54}$$

and refer to Dorfmann and Ogden (2005) as well as to Wissler and Mazza (2005) when making the interpretation of p as taking the role of a hydrostatic pressure, which can be identified as taking a mechanical part as well as an electric part which corresponds to the electrostatic force acting on the dielectric material, in case of plane stress.

12.3 Electromechanical Coupling Using a Multiplicative Decomposition of the Deformation Gradient Tensor

In order to broaden the constitutive model to nonlinear effects involving electro-mechanical coupling on the constitutive level, we make use of the multiplicative decomposition of the deformation gradient tensor. This idea is adopted from the fields of thermo-elasticity and plasticity. Using this technique allows for incorporating different or even multiple phenomena and cross effects, e.g. piezoelectricity or electrostriction.

Obviously when dealing with electro-mechanical coupling, the deformation gradient is naturally decomposed into an elastic part $\mathbf{F}_{me}$, called the mechanical and an electric part $\mathbf{F}_{el}$ called the electric deformation gradient tensor,

$$\mathbf{F} = \mathbf{F}_{me} \cdot \mathbf{F}_{el}, \tag{12.55}$$

where $\mathbf{F}_{el} = \mathbf{F}_{el}(\boldsymbol{\mathcal{E}})$ is assumed to solely depend on the material electric field vector $\boldsymbol{\mathcal{E}}$.

We apply the right (Lee-type; Lee, 1969) decomposition proposed by Skatulla et al (2012) for dielectric elastomers, where the right Cauchy-Green tensor can then be expressed by $\mathbf{C} = \mathbf{F}_{el}^T \cdot \mathbf{C}_{me} \cdot \mathbf{F}_{el}$ with the mechanical part being $\mathbf{C}_{me} = \mathbf{F}_{me}^T \cdot \mathbf{F}_{me}$.

It is clear, that the decomposition order puts already restrictions on the choice of dependent variables within the free energy function, resulting in a rather specific theory. Therefore, switching the order of the multiplicative decomposition to a right (Clifton-type; Clifton, 1972) decomposition, necessitates a completely different modeling approach from the very beginning.

We start the derivation of the constitutive relations analogously to the previous section 12.2, however, we drop the augmentation term temporary, the free energy function now reads $\psi = \psi_{me}(\mathbf{C}_{me}) + \psi_{el}(\mathbf{C}, \boldsymbol{\mathcal{E}})$, while its rate computes to:

$$\dot{\psi} = \frac{\partial \psi_{me}}{\partial \mathbf{C}_{me}} \cdot\cdot \dot{\mathbf{C}}_{me} + \frac{\partial \psi_{el}}{\partial \mathbf{C}} \cdot\cdot \dot{\mathbf{C}} + \frac{\partial \psi_{el}}{\partial \boldsymbol{\mathcal{E}}} \cdot \dot{\boldsymbol{\mathcal{E}}}. \tag{12.56}$$

Inserting $\mathbf{C} = \mathbf{F}_{el}^T \cdot \mathbf{C}_{me} \cdot \mathbf{F}_{el}$ the time rate of the mechanical right Cauchy-Green tensor is:

$$\dot{\mathbf{C}}_{me} = \mathbf{F}_{el}^{-T} \cdot \dot{\mathbf{C}} \cdot \mathbf{F}_{el}^{-1} - 2\mathrm{sym}\left(\mathbf{C}_{me} \cdot \dot{\mathbf{F}}_{el} \cdot \mathbf{F}_{el}^{-1}\right). \tag{12.57}$$

Therefore, upon applying the symmetry and cyclic permutation property of the double dot product, and after inserting $\dot{\mathbf{F}}_{el} = \frac{\partial \mathbf{F}_{el}}{\partial \boldsymbol{\mathcal{E}}} \cdot \dot{\boldsymbol{\mathcal{E}}}$, the first, mechanical, part of the free energy, gets:

$$\frac{\partial \psi_{me}}{\partial \mathbf{C}_{me}} \cdot\cdot \dot{\mathbf{C}}_{me} = \mathbf{F}_{el}^{-1} \cdot \frac{\partial \psi_{me}}{\partial \mathbf{C}_{me}} \cdot \mathbf{F}_{el}^{-T} \cdot\cdot \dot{\mathbf{C}} - \left(2\mathbf{F}_{el}^{-1} \cdot \frac{\partial \psi_{me}}{\partial \mathbf{C}_{me}} \cdot \mathbf{C}_{me} \cdot\cdot \frac{\partial \mathbf{F}_{el}}{\partial \boldsymbol{\mathcal{E}}}\right) \cdot \dot{\boldsymbol{\mathcal{E}}}. \tag{12.58}$$

This allows to rewrite the rate of the free energy Eq. (12.56), such that the global form with regard to the double dot product can be restored:

$$\dot{\psi} = \left(\mathbf{F}_{el}^{-1} \cdot \frac{\partial \psi_{me}}{\partial \mathbf{C}_{me}} \cdot \mathbf{F}_{el}^{-T} + \frac{\partial \psi_{el}}{\partial \mathbf{C}}\right) \cdot\cdot \dot{\mathbf{C}} - \left(2\mathbf{F}_{el}^{-1} \cdot \frac{\partial \psi_{me}}{\partial \mathbf{C}_{me}} \cdot\cdot \frac{\partial \mathbf{F}_{el}}{\partial \boldsymbol{\mathcal{E}}} - \frac{\partial \psi_{el}}{\partial \boldsymbol{\mathcal{E}}}\right) \cdot \dot{\boldsymbol{\mathcal{E}}}. \tag{12.59}$$

Comparing now the coefficients of this relation to the material rate of free energy from Eq. (12.36), the constitutive relations for the symmetric second Piola-Kirchoff stress tensor as well as for the polarization vector can be obtained:

$$\mathbf{S}^S = \mathbf{S} + \boldsymbol{\mathcal{P}}\boldsymbol{\mathcal{E}} \cdot \mathbf{C}^{-1} = 2\rho_0 \mathbf{F}_{el}^{-1} \cdot \frac{\partial \psi_{me}}{\partial \mathbf{C}_{me}} \cdot \mathbf{F}_{el}^{-T} + 2\rho_0 \frac{\partial \psi_{el}}{\partial \mathbf{C}}, \tag{12.60}$$

$$\boldsymbol{\mathcal{P}} = 2\rho_0 \mathbf{F}_{el}^{-1} \cdot \frac{\partial \psi_{me}}{\partial \mathbf{C}_{me}} \cdot \mathbf{C}_{me} \cdot\cdot \frac{\partial \mathbf{F}_{el}}{\partial \boldsymbol{\mathcal{E}}} - \rho_0 \frac{\partial \psi_{el}}{\partial \boldsymbol{\mathcal{E}}}. \tag{12.61}$$

Here the pronounced coupling nature of the multiplicative decomposition becomes clear, while the second Piola-Kirchoff stress tensor experiences a transformation by the electrical deformation gradient, the polarization vector $\boldsymbol{\mathcal{P}}$ is now composed of two parts, an electrical one $\boldsymbol{\mathcal{P}}_{el}$ and a coupled electro-mechanical part $\boldsymbol{\mathcal{P}}_{coup}$.

$$\boldsymbol{\mathcal{P}}_{el} = -\rho_0 \frac{\partial \psi_{el}}{\partial \boldsymbol{\mathcal{E}}}, \qquad \boldsymbol{\mathcal{P}}_{coup} = 2\rho_0 \mathbf{F}_{el}^{-1} \cdot \frac{\partial \psi_{me}}{\partial \mathbf{C}_{me}} \cdot \mathbf{C}_{me} \cdot\cdot \frac{\mathbf{F}_{el}}{\partial \boldsymbol{\mathcal{E}}}. \tag{12.62}$$

Moreover, the corresponding polarization stress $\mathbf{S}^P = \mathbf{S}^{pol,el} + \mathbf{S}^{pol,coup}$ is again obtained by multiplication with $\boldsymbol{\mathcal{E}} \cdot \mathbf{C}^{-1}$:

$$\mathbf{S}^{pol,el} = \boldsymbol{\mathcal{P}}_{el} \boldsymbol{\mathcal{E}} \cdot \mathbf{C}^{-1} = -\rho_0 \left(\frac{\partial \psi_{el}}{\partial \boldsymbol{\mathcal{E}}} \right) \boldsymbol{\mathcal{E}} \cdot \mathbf{C}^{-1}, \tag{12.63}$$

$$\mathbf{S}^{pol,coup} = \boldsymbol{\mathcal{P}}_{coup} \boldsymbol{\mathcal{E}} \cdot \mathbf{C}^{-1} = \rho_0 \left(2\mathbf{F}_{el}^{-1} \cdot \frac{\partial \psi_{me}}{\partial \mathbf{C}_{me}} \cdot \mathbf{C}_{me} \cdot\cdot \frac{\partial \mathbf{F}_{el}}{\partial \boldsymbol{\mathcal{E}}} \right) \boldsymbol{\mathcal{E}} \cdot \mathbf{C}^{-1}. \tag{12.64}$$

The symmetric Piola-Kirchoff stress tensor composes now of three parts $\mathbf{S}^S = \mathbf{S} + \mathbf{S}^{pol,coup} + \mathbf{S}^{pol,el}$. The term $\mathbf{S}^{pol,el}$, introduces the electrostatic force into the theory, since it has to fulfill the restriction

$$-\frac{\partial \psi_{el}}{\partial \boldsymbol{\mathcal{E}}} \boldsymbol{\mathcal{E}} \cdot \mathbf{C}^{-1} = 2 \frac{\partial \psi_{el}}{\partial \mathbf{C}}. \tag{12.65}$$

Due to simplicity, we choose the same free energy $\rho_0 \psi_{el} = -\frac{1}{2} \chi \varepsilon_0 \boldsymbol{\mathcal{E}} \cdot (\mathbf{C}^{-1} \cdot \boldsymbol{\mathcal{E}})$ from Sect. 12.2, since it has already been shown to be a proper choice. The variables ε_0 and χ are the permittivity in vacuum and the electric susceptibility respectively.

The identification of the electrostatic force motivates further statements on the analysis of the electro-elastic coupling nature. As the responsible term for the electrostatic force can now be excluded, still a symmetric electro-mechanical stress tensor $\mathbf{S}^{em} = \mathbf{S} + \boldsymbol{\mathcal{P}}_{coup} \boldsymbol{\mathcal{E}} \cdot \mathbf{C}^{-1}$ can be obtained, which upon comparison to Eq. (12.60) reads:

$$\mathbf{S}^{em} = 2\rho_0 \mathbf{F}_{el}^{-1} \cdot \frac{\partial \psi_{me}}{\partial \mathbf{C}_{me}} \cdot \mathbf{F}_{el}^{-T}. \tag{12.66}$$

By using $\mathbf{C}_{me} = \mathbf{F}_{el}^{-T} \cdot \mathbf{C} \cdot \mathbf{F}_{el}^{-1}$, the coupling polarization can be written as:

$$\boldsymbol{\mathcal{P}}_{coup} = \mathbf{S}^{em} \cdot \mathbf{C} \cdot \mathbf{F}_{el}^{-1} \cdot\cdot \frac{\partial \mathbf{F}_{el}}{\partial \boldsymbol{\mathcal{E}}}, \tag{12.67}$$

which after comparison to the balance of energy yields the symmetric second Piola-Kirchoff stress tensor being composed of an electromechanical part $\mathbf{S}^{em}$ and the electrical part $\mathbf{S}^{pol,el}$.

$$\mathbf{S}^S = \mathbf{S} + \boldsymbol{\mathcal{P}} \boldsymbol{\mathcal{E}} \cdot \mathbf{C}^{-1} = \mathbf{S}^{em} + \mathbf{S}^{pol,el}, \qquad \boldsymbol{\mathcal{P}} = \boldsymbol{\mathcal{P}}_{el} + \boldsymbol{\mathcal{P}}_{coup} \tag{12.68}$$

with the specific constitutive relations:

$$\mathbf{S}^{em} = 2\rho_0 \mathbf{F}_{el}^{-1} \cdot \frac{\partial \psi_{me}}{\partial \mathbf{C}_{me}} \cdot \mathbf{F}_{el}^{-T} \quad \text{and} \quad \mathbf{S}^{pol,el} = 2\rho_0 \frac{\partial \psi_{el}}{\partial \mathbf{C}}, \tag{12.69}$$

$$\boldsymbol{\mathcal{P}}_{el} = -\rho_0 \frac{\partial \psi_{el}}{\partial \boldsymbol{\mathcal{E}}} \quad \text{and} \quad \boldsymbol{\mathcal{P}}_{coup} = \mathbf{S}^{em} \cdot \mathbf{C} \cdot \mathbf{F}_{el}^{-1} \cdot\cdot \frac{\partial \mathbf{F}_{el}}{\partial \boldsymbol{\mathcal{E}}}. \tag{12.70}$$

In view of a proper thermodynamic presentation of the rate of the free energy, one can finally write:

$$\rho_0\dot{\psi} = \underbrace{\mathbf{F}_{el}\cdot\mathbf{S}^{em}\cdot\mathbf{F}_{el}^T\cdot\cdot\frac{1}{2}\dot{\mathbf{C}}_{me}}_{=\rho_0\dot{\psi}_{me}(\mathbf{C}_{me})} + \underbrace{\mathbf{S}^{pol,el}\cdot\cdot\frac{1}{2}\dot{\mathbf{C}}-\boldsymbol{\mathcal{P}}_{el}\cdot\dot{\boldsymbol{\mathcal{E}}}}_{=\rho_0\dot{\psi}_{el}}. \tag{12.71}$$

Consider the case $\mathbf{F}_{el}=\mathbf{I}$; then $\boldsymbol{\mathcal{P}}_{coup}=\mathbf{0}, \mathbf{F}_{me}=\mathbf{F}, \mathbf{C}_{me}=\mathbf{C}$ and $\mathbf{S}^{em}=\mathbf{S}$ hold, and the previous constitutive model without constitutive coupling is found:

$$\mathbf{S}=2\rho_0\cdot\frac{\partial\psi_{me}}{\partial\mathbf{C}}, \quad \mathbf{S}^{pol}=\boldsymbol{\mathcal{P}}\boldsymbol{\mathcal{E}}\cdot\mathbf{C}^{-1}=2\rho_0\frac{\partial\psi_{el}}{\partial\mathbf{C}} \quad \text{and} \quad \boldsymbol{\mathcal{P}}=-\rho_0\frac{\partial\psi_{el}}{\partial\boldsymbol{\mathcal{E}}}. \tag{12.72}$$

12.3.1 Total Stress

It remains to incorporate the contribution of the electric field in vacuum to the constitutive model. Starting at the definition for the free energy function, the objective we want to achieve is to find some global relationships which hold when incorporating the multiplicative decomposed deformation gradient. We make use of the augmentation term in Sect. 12.2 and write

$$\rho_0\Omega=\rho_0\psi-\frac{1}{2}\varepsilon_0 J\boldsymbol{\mathcal{E}}\cdot(\mathbf{C}^{-1}\cdot\boldsymbol{\mathcal{E}})=\rho_0\psi+\rho_0\psi_{aug}. \tag{12.73}$$

Hence, in analogy to the electric free energy, also for the augmentation term $\psi_{aug}=\psi_{aug}(\mathbf{C},\boldsymbol{\mathcal{E}})$ holds, and the rate of the augmented free energy can be expressed as:

$$\rho_0\dot{\Omega}=\rho_0\dot{\psi}+\rho_0\frac{\partial\psi_{aug}}{\partial\mathbf{C}}\cdot\cdot\dot{\mathbf{C}}+\rho_0\frac{\partial\psi_{aug}}{\partial\boldsymbol{\mathcal{E}}}\cdot\dot{\boldsymbol{\mathcal{E}}}. \tag{12.74}$$

However, in few of a uniform presentation, one might be interested to further develop $\dot{\psi}$ given in Eq. (12.59). As $\psi_{me}=\psi_{me}(\mathbf{C}_{me})=\psi_{me}(\mathbf{C}_{me}(\mathbf{C},\boldsymbol{\mathcal{E}}))$ holds, and by using $\mathbf{C}_{me}=\mathbf{F}_{el}^{-T}\cdot\mathbf{C}\cdot\mathbf{F}_{el}^{-1}$ the derivative of the mechanical free energy can be expressed by

$$\begin{aligned}\frac{\partial\psi_{me}}{\partial\boldsymbol{\mathcal{E}}} &= \frac{\partial\psi_{me}}{\partial\mathbf{C}_{me}}\cdot\cdot\frac{\partial\mathbf{C}_{me}}{\partial\boldsymbol{\mathcal{E}}}=\frac{\partial\psi_{me}}{\partial\mathbf{C}_{me}}\cdot\cdot\frac{\partial(\mathbf{F}_{el}^{-T}\cdot\mathbf{C}\cdot\mathbf{F}_{el}^{-1})}{\partial\boldsymbol{\mathcal{E}}}\\ &= -2\mathbf{F}_{el}^{-1}\cdot\frac{\partial\psi_{me}}{\partial\mathbf{C}_{me}}\cdot\left(\mathbf{C}_{me}\cdot\frac{\partial\mathbf{F}_{el}}{\partial\boldsymbol{\mathcal{E}}}\cdot\mathbf{F}_{el}^{-1}\right)=-2\mathbf{F}_{el}^{-1}\frac{\partial\psi_{me}}{\partial\mathbf{C}_{me}}\cdot\mathbf{C}_{me}\cdot\cdot\frac{\partial\mathbf{F}_{el}}{\partial\boldsymbol{\mathcal{E}}},\end{aligned} \tag{12.75}$$

and furthermore,

$$\frac{\partial\psi_{me}}{\partial\mathbf{C}}=\frac{\partial\psi_{me}}{\partial\mathbf{C}_{me}}\cdot\cdot\frac{\partial\mathbf{C}_{me}}{\partial\mathbf{C}}=\frac{\partial\psi_{me}}{\partial\mathbf{C}_{me}}\cdot\cdot\frac{\partial(\mathbf{F}_{el}^{-T}\cdot\mathbf{C}\cdot\mathbf{F}_{el}^{-1})}{\partial\mathbf{C}}=\mathbf{F}_{el}^{-1}\cdot\frac{\partial\psi_{me}}{\partial\mathbf{C}_{me}}\cdot\cdot\mathbf{F}_{el}^{-T} \tag{12.76}$$

which finally gives

$$\boldsymbol{\mathcal{D}} = -\rho_0 \frac{\partial \psi_{me}}{\partial \boldsymbol{\mathcal{E}}} - \rho_0 \frac{\partial \psi_{el}}{\partial \boldsymbol{\mathcal{E}}} - \rho_0 \frac{\partial \psi_{aug}}{\partial \boldsymbol{\mathcal{E}}} = -\rho_0 \frac{\partial \Omega}{\partial \boldsymbol{\mathcal{E}}}, \tag{12.77}$$

$$\mathbf{S}^{tot} = 2\rho_0 \frac{\partial \psi_{me}}{\partial \mathbf{C}} + 2\rho_0 \frac{\partial \psi_{el}}{\partial \mathbf{C}} + 2\rho_0 \frac{\partial \psi_{aug}}{\partial \mathbf{C}} = 2\rho_0 \frac{\partial \Omega}{\partial \mathbf{C}}. \tag{12.78}$$

Note, that in case of multiplicative decomposition, the electric displacement vector contains the material derivative of the mechanical free energy with respect to the electric field, this was not the case in the representation of the constitutive equations in section 12.2. Inserting the quantities into the rate of augmented free energy finally yields

$$\rho_0 \dot{\Omega} = \mathbf{S}^{tot} \cdot\cdot \frac{1}{2} \dot{\mathbf{C}} - \boldsymbol{\mathcal{D}} \cdot \dot{\boldsymbol{\mathcal{E}}}. \tag{12.79}$$

12.3.2 Intermediate Configuration

By using a multiplicative decomposition, one equivalently introduces an intermediate configuration, see Fig. 12.3 into the deformation path, which is a result of the right Lee-type decomposition. In our case, the intermediate configuration might be adopted in case when no mechanical loads are applied, hence $\mathbf{C}_{me} = \mathbf{I}$, and the electro-mechanical stress, given in Eq. (12.68)

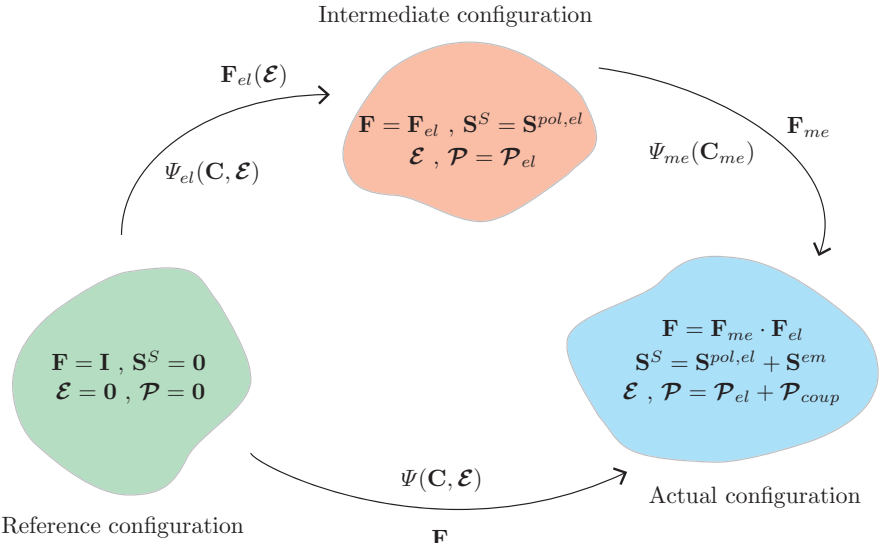

Fig. 12.3: The intermediate configuration is entered by instantaneously taking all mechanical loads yielding to an appealing, yet unphysical state of pure electrical origin.

$$\mathbf{S}^{em} = \mathbf{S} + \boldsymbol{\mathcal{P}}_{coup}\boldsymbol{\mathcal{E}} \cdot \mathbf{C}^{-1} = 2\rho_0 \mathbf{F}_{el}^{-1} \cdot \frac{\partial \psi_{me}}{\partial \mathbf{C}_{me}} \cdot \mathbf{F}_{el}^{-T}, \tag{12.80}$$

has to vanish. This is true, because the mechanical part of the free energy $\psi_{me}(\mathbf{C}_{me})$ per definition depends solely on the mechanical right Cauchy-Green tensor. Hence, the symmetric stress tensor $\mathbf{S}^S$ for the intermediate configuration must be equal to the electrical polarization stress tensor $\mathbf{S}^S = \mathbf{S}^P = \mathbf{S}^{pol,el}$. Moreover, as the total stress tensor is $\mathbf{S}^{tot} = \mathbf{S}^S - \mathbf{S}^P + \mathbf{S}^E = \mathbf{S}^E$ yields to $\mathbf{S}^{tot} = \mathbf{S}^P + \mathbf{S}^M$, hence, there exists a total stress tensor in the intermediate configuration, composed of the symmetric Maxwell stress and the unsymmetric electrical polarization stress.

Moreover, as the electro-mechanical stress tensor is composed of the second (mechanical) Piola-Kirchoff stress and the coupling polarization stress tensor $\mathbf{S}^{em} = \mathbf{S} + \mathbf{S}^{pol,coup} = \mathbf{0}$ yields in consequence that either $\mathbf{S} = -\mathbf{S}^{pol,coup}$ or both $\mathbf{S}^{pol,coup} = \mathbf{0}$ and $\mathbf{S} = \mathbf{0}$ vanish. However, in either way, the total stress tensor remains apparent because of the Maxwell stress tensor, hence the intermediate configuration cannot be a "*stress-free*" configuration.

This concept of different deformation path yields to three possible configurations:

1. The unloaded reference configuration, here both the electric field vector and the polarization vector have to vanish $\boldsymbol{\mathcal{E}} = \boldsymbol{\mathcal{P}} = \mathbf{0}$, while at the same time $\mathbf{F} = \mathbf{I}$ is prescribed, hence this configuration is stress free $\mathbf{S}^{tot} = \mathbf{S}^S = \mathbf{0}$.
2. Allowing a pure electric polarization $\boldsymbol{\mathcal{P}} = \boldsymbol{\mathcal{P}}_{el}$ accompanied with an electric field, yields to a configuration where stress fields $\mathbf{S}^{tot} = \mathbf{S}^{pol,coup} + \mathbf{S}^M$ are present yielding to a deformation field characterized by $\mathbf{F} = \mathbf{F}_{el}$.
3. Within the actual configuration, all physical possible combination of electric and mechanic sources are present, allowing now the the symmetric electro-mechanical stress tensor $\mathbf{S}^{em}$ to emerge. When starting in the intermediate configuration, the actual configuration is attained through the mechanical energy field $\psi_{me}(\mathbf{C}_{me})$. When going this path one has to keep the electric field $\boldsymbol{\mathcal{E}}$ constant and add the coupled term to the polarization vector field $\boldsymbol{\mathcal{P}} = \boldsymbol{\mathcal{P}}_{el} + \boldsymbol{\mathcal{P}}_{coup}$.

12.4 Electrostriction

Phenomenologically speaking, electrostriction is the quadratic response in the strain field, upon application of an electric field; hence, we write a series expansion

$$\varepsilon \approx \boldsymbol{\mathcal{E}} \cdot {}^3\mathbf{e} + \boldsymbol{\mathcal{E}}\boldsymbol{\mathcal{E}} \cdot\cdot {}^4\mathbf{D} + \ldots \tag{12.81}$$

for the infinitesimal strain tensor ε, where ${}^3\mathbf{e}$ is a third rank piezoelectric coefficient tensor, and ${}^4\mathbf{D}$ a fourth rank electrostrictive parameter tensor. Therefore, electrostriction renders a higher order effect, which is typically considered as negligible. However, experimental results show, that among the linear electrostatic relation, also higher order effects contribute significantly to the strain field. As piezoelectricity is a special topic, in this work we restrict ourselves to electrostriction, which also depends

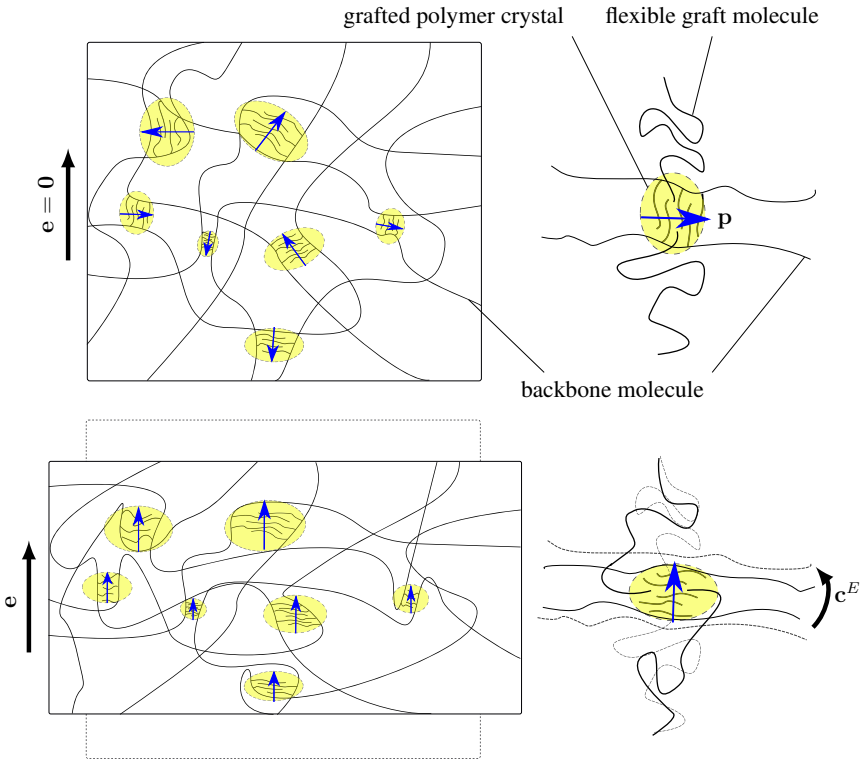

Fig. 12.4: Electrostrictive graft polymer, left deformation of the whole specimen, right reorientation of the crystal unit upon application of an electric field.

highly on the materials properties. In order to explore the origin of this effect one has to look inside the micro-structure. Materials which exhibit a distinct electrostrictive behavior carry typically polarized cells or crystalline groups within a matrix of long chained elastomer molecules. In 1998 NASA (Su et al, 1999) published results on their improved EAP material, with especially improved electrostrictive properties, called electrostrictive graft elastomer. The key ingredients are crystalline groups which are solvents in a flexible backbone polymer matrix. Upon application of an external electric field, the dipole moments within the crystal cells have to reorient, according to the induced dipole couple $\mathbf{c}^E = \mathbf{p} \times \mathbf{e}$. This effect is shown in Fig. 12.4. If the sign of the external field is changed, the dipole couple reorients in the other direction; hence, the cells are turned around such, that the same net deformation can be measured. As every material carries imperfections, which lead to polarized cells within the structure, every material is capable of undergoing an electrostrictive behavior. However, e.g. in silicon rubber one can barely find such micro defects,

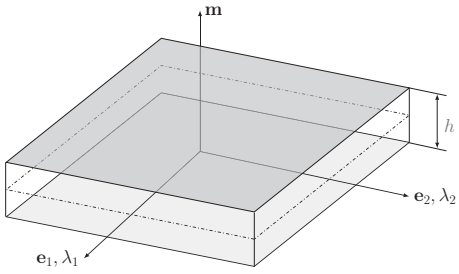

Fig. 12.5: Homogeneous plate in cartesian coordinates, the unit vector in thickness direction is denoted with **m**.

which is why the electrostrictive effect of silicon rubber (3M VHB4910) is rather small. Other materials which have drawn attention due to their distinct ability towards electrostriction are polyurethane elastomers, see experimental results in Diaconu et al (2006). We complete this introductory part by mentioning that also combinations of electrostrictive EAPs and piezoelectric copolymers exist (so called ferroelectric-electrostrictive materials), which allow the use of the piezoelectric polymer for sensing and the electrostrictive one for actuation.

In order to incorporate electrostriction into the constitutive relations in the present paper, we make use of the yet undefined electric deformation gradient tensor $\mathbf{F}_{el}$, which in case of electrostriction might e.g. take the form of an exponential function suggested by Skatulla et al (2012):

$$\mathbf{F}_{el} = \exp\mathbf{D}, \tag{12.82}$$

where $\mathbf{D}$ is a proper second rank tensor, which in turn has to satisfy $\mathbf{D} = \ln\mathbf{F}_{el}$, such that by choosing $\mathbf{D}$ carefully, the electric deformation gradient might become identical to the electric right stretch tensor $\mathbf{F}_{el} = \mathbf{R}_{el}\mathbf{U}_{el} := \mathbf{U}_{el}$, hence $\mathbf{D}$ becomes an electrical logarithmic strain tensor $\mathbf{E}^{el}$

$$\mathbf{D} = \ln\mathbf{U}_{el} = \mathbf{E}^{el}, \tag{12.83}$$

which then can be chosen quadratic in the material electric field vector $\boldsymbol{\mathcal{E}}$.

12.4.1 Homogeneously Deformed Plate

We turn now to the case of homogeneous in plane deformation of a plate, see Fig. 12.5, in which the coordinate system is embedded such, that one unit vector $\mathbf{m}$ is aligned with the electric field vector $\boldsymbol{\mathcal{E}} = \mathcal{E}_3\mathbf{m}$, acting in thickness direction. Hence the electric deformation gradient takes the form $\mathbf{F}_{el} = \mathbf{U}_{el} = \lambda_{el}(\mathbf{I} - \mathbf{mm}) + \lambda_{el,3}\mathbf{mm}$.

Therefore, the tensor $\mathbf{D}$ is taken in the form of a diagonal tensor:

$$\mathbf{D} = c_1(\boldsymbol{\mathcal{E}}\cdot\boldsymbol{\mathcal{E}})\mathbf{mm} + c_2(\boldsymbol{\mathcal{E}}\cdot\boldsymbol{\mathcal{E}})(\mathbf{I}-\mathbf{mm}), \tag{12.84}$$

where the two parameter c_1 and c_2 are electrostrictive material parameter, which allow to write the electrical stretches in the form:

$$\lambda_{el} = \exp(c_2\boldsymbol{\mathcal{E}}\cdot\boldsymbol{\mathcal{E}}) \quad , \qquad \lambda_{el,3} = \exp(c_1\boldsymbol{\mathcal{E}}\cdot\boldsymbol{\mathcal{E}}). \tag{12.85}$$

It remains now to recall and specify the augmented free energy. Starting with the mechanical free energy, we are using a neo-Hookean hyperelastic strain energy function,

$$\rho_0\psi_{me}(\mathbf{C}_{me}) = \rho_0\psi_{me}(I_{\mathbf{C}_{me}}, II_{\mathbf{C}_{me}}, III_{\mathbf{C}_{me}}) = \frac{\mu}{2}(I_{\mathbf{C}_{me}} - 3 - 2\ln J_{me}) + K(\ln J_{me})^2; \tag{12.86}$$

the electrical and the augmented free energy are respectively:

$$\rho_0\psi_{el} = -\frac{1}{2}\chi\varepsilon_0\boldsymbol{\mathcal{E}}\cdot(\mathbf{C}^{-1}\cdot\boldsymbol{\mathcal{E}}) \tag{12.87}$$

$$\rho_0\psi_{aug} = -\frac{1}{2}J\varepsilon_0\boldsymbol{\mathcal{E}}\cdot(\mathbf{C}^{-1}\cdot\boldsymbol{\mathcal{E}}). \tag{12.88}$$

12.4.1.1 Plane Stress

For the total stress tensor we write $\mathbf{S}^{tot} = \mathbf{S}_2 + \boldsymbol{\tau}\mathbf{m} + \mathbf{m}\boldsymbol{\tau} + S_{33}\mathbf{mm}$, where $\mathbf{S}_2$ is its plane part, $\boldsymbol{\tau}$ is the total transverse shear stress vector and S_{33} the magnitude of the total stress in direction of the unit normal vector $\mathbf{m}$. We use $\mathbf{I}_2 = \mathbf{I} - \mathbf{mm}$ for the plane identity tensor. The restriction to plane stress allows to set all parts in $\mathbf{m}$ direction zero, hence $\mathbf{S}^{tot} = \mathbf{S}_2$. As a consequence, shear components of the right Cauchy-Green tensor vanish; hence, it can be decomposed into an in plane part $\mathbf{C}_2$, and an out of plane tensor $C_{33}\mathbf{mm}$ such that $\mathbf{C} = \mathbf{C}_2 + C_{33}\mathbf{mm}$. In accordance to the total right Cauchy-Green tensor, also the mechanical right Cauchy-Green tensor turns into $\mathbf{C}_{me} = \mathbf{C}_{2,me} + C_{33,me}\mathbf{mm}$. By using the transformation rule $\mathbf{C}_{me} = \mathbf{F}_{el}^{-T}\cdot\mathbf{C}\cdot\mathbf{F}_{el}^{-1}$, the invariants of $\mathbf{C}_{me}$ turn into functions of the total right Cauchy-Green tensor, and the electric stretches.

$$I_{\mathbf{C}_{me}} = \mathrm{tr}\mathbf{C}_{me} = \lambda_{el}^{-2}\mathrm{tr}\mathbf{C}_2 + \lambda_{el,3}^{-2}C_{33}, \tag{12.89}$$

$$II_{\mathbf{C}_{me}} = \mathbf{C}_{me}\cdot\cdot\,\mathbf{C}_{me} = \lambda_{el,3}^{-4}C_{33}^2 + \lambda_{el}^{-4}\mathbf{C}_2\cdot\cdot\,\mathbf{C}_2, \tag{12.90}$$

$$III_{\mathbf{C}_{me}} = \det\mathbf{C}_{me} = \lambda_{el}^{-4}\lambda_{el,3}^{-2}C_{33}\det\mathbf{C}_2. \tag{12.91}$$

12.4.1.2 Incompressibility

As dielectric elastomers are often considered incompressible, $J = \det\mathbf{F} := 1$, we have:

- $\det\mathbf{F} = \det\mathbf{F}_{el} = 1$,
- $\det\mathbf{C} = 1 \rightarrow C_{33} = \det\mathbf{C}_2^{-1} = III_{\mathbf{C}_2}^{-1}$.

Turning to the electric deformation gradient tensor, the incompressibility condition already defines one of the electrostrictive material parameter c_1 in terms of the second one c_2:

$$\det\mathbf{F}_{el} = 1 = \lambda_{el}^2\lambda_{el,3} = \exp((c_1 + 2c_2)\mathcal{E}_3^2) = 1 \quad \rightarrow \quad c_1 = -2c_2. \tag{12.92}$$

Additionally the electric in plane stretch is a direct result of the out of plane stretch: $\lambda_{el}^{-2} = \lambda_{el,3} = \exp(c_1\mathcal{E}_3^2)$. For the electric free energy function the inverse total right Cauchy-Green tensor is needed, as $\mathbf{C}^{-1} = \mathbf{C}_2^{-1} + C_{33}^{-1}\mathbf{mm}$ holds. Upon inserting $\boldsymbol{\mathcal{E}} = \mathcal{E}_3\mathbf{m}$ and using $J = 1$ the electric free energy and the augmented free energy are:

$$\rho_0(\psi_{el} + \psi_{aug}) = -\frac{1}{2}\varepsilon_0(1+\chi)\frac{\mathcal{E}_3^2}{C_{33}}. \tag{12.93}$$

For the mechanical free energy function the trace of the mechanical right Cauchy-Green tensor is needed. If we make use of $\lambda_{el}^{-2} = \lambda_{el,3}$ and $C_{33} = \det\mathbf{C}_2^{-1} = III_{\mathbf{C}_2}^{-1}$, the invariants read:

$$I_{\mathbf{C}_{me}} = \lambda_{el}^{-2}\mathrm{tr}\mathbf{C}_2 + \lambda_{el}^4 C_{33}, \tag{12.94}$$

$$II_{\mathbf{C}_{me}} = \lambda_{el}^8 C_{33}^2 + \lambda_{el}^{-4}\mathbf{C}_2 \cdot\cdot \mathbf{C}_2, \tag{12.95}$$

$$III_{\mathbf{C}_{me}} = \lambda_{el}^{-4}\lambda_{el}^4 III_{\mathbf{C}_2}^{-1}\det\mathbf{C}_2 = 1. \tag{12.96}$$

We can now write the augmented free energy for the incompressible neo-Hookean material and the plane stress case:

$$\rho_0\Omega_2 = \frac{\mu}{2}\left(\lambda_{el}^{-2}\mathrm{tr}\mathbf{C}_2 + \lambda_{el}^4 III_{\mathbf{C}_2}^{-1} - 3\right) - \frac{1}{2}\varepsilon III_{\mathbf{C}_2}\mathcal{E}_3^2, \tag{12.97}$$

where we used the permittivity $\varepsilon = \varepsilon_0(\chi + 1) = \varepsilon_r\varepsilon_0$, with the relative permittivity $\varepsilon_r = \chi + 1$, and the electrical stretch $\lambda_{el} = \exp(c_2\mathcal{E}_3^2) = \exp((-c_1/2)\mathcal{E}_3^2)$.

12.4.1.3 Electrostatic Force

We already reduced the augmented free energy to its plane counterpart, however, in order to ensure incompressibility when using the constitutive relations for the stresses one has to enforce the condition by using a Lagrange multiplier p:

$$\mathbf{S}_2 = 2\rho_0\frac{\partial\Omega_2}{\partial\mathbf{C}_2} + p\mathbf{C}_2^{-1}, \tag{12.98}$$

where p can be obtained by making use of the plane stress condition in the three dimensional problem $S_{33} = 0$, keeping $C_{33} = III_{\mathbf{C}_2}^{-1}$ in mind yields

$$S_{33} = 2\rho_0\frac{\partial\Omega_2}{\partial C_{33}} + pC_{33}^{-1} = 0 \quad \rightarrow \qquad p = -2C_{33}\rho_0\frac{\partial\Omega_2}{\partial C_{33}}. \tag{12.99}$$

Inserting p into the plane part of the total stress tensor finds

$$\mathbf{S}_2 = 2\rho_0 \frac{\partial \Omega_2}{\partial \mathbf{C}_2} - 2C_{33}\rho_0 \frac{\partial \Omega_2}{\partial C_{33}} \mathbf{C}_2^{-1}, \tag{12.100}$$

which upon using $C_{33} = III_{\mathbf{C}_2}^{-1}$ and the identity $\frac{\partial III_{\mathbf{C}_2}^{-1}}{\partial \mathbf{C}_2} = -III_{\mathbf{C}_2}^{-1}\mathbf{C}_2^{-1}$ finally becomes

$$\begin{aligned} \mathbf{S}_2 &= 2\rho_0 \left(\left. \frac{\partial \Omega_2}{\partial \mathbf{C}_2} \right|_{C_{33}=III_{\mathbf{C}_2}^{-1}} - \left. \frac{\partial \Omega_2}{\partial C_{33}} \right|_{C_{33}=III_{\mathbf{C}_2}^{-1}} III_{\mathbf{C}_2}^{-1}\mathbf{C}_2^{-1} \right) \\ &= 2\rho_0 \left(\frac{\partial \Omega_2}{\partial \mathbf{C}_2} + \frac{\partial \Omega_2}{\partial C_{33}} \frac{\partial C_{33}}{\partial \mathbf{C}_2} \right) \Bigg|_{C_{33}=III_{\mathbf{C}_2}^{-1}} . \end{aligned} \tag{12.101}$$

This is however identical to

$$\mathbf{S}_2 = 2\rho_0 \frac{\partial \Omega_2}{\partial \mathbf{C}_2}, \tag{12.102}$$

with the plane part of the augmented free energy. Hence, for the case of plane stress, incompressibility is ensured by application of a pressure p. The externally applied electric field acting in thickness direction, however yields to a stress resultant in thickness direction whose contribution has to be balanced by the Lagrange multiplier

$$p = -2C_{33}\rho_0 \frac{\partial \Omega_2}{\partial C_{33}}, \tag{12.103}$$

which takes part of a mixed mechanical portion on the one hand, and an electrostatic Coulomb force resultant on the other hand.

In summery, the constitutive model for plane stress case yields the total stress tensor $\mathbf{S}^{tot} = \mathbf{S}_2^{tot}$. Because of $\boldsymbol{\mathcal{E}} = \mathcal{E}_3\mathbf{m}$ also the electric displacement vector has only a component in the thickness direction $\mathcal{D}_3$; hence, the non-vanishing components read:

$$\mathbf{S}_2^{tot} = 2\rho_0 \frac{\partial \Omega_2}{\partial \mathbf{C}_2} \quad , \qquad \mathcal{D}_3 = -\rho_0 \frac{\partial \Omega_2}{\partial \mathcal{E}_3} = \varepsilon III_{\mathbf{C}_2}\mathcal{E}_3 - \rho_0 \frac{\partial \Omega_2}{\partial \lambda_{el}} \frac{\partial \lambda_{el}}{\partial \mathcal{E}_3}. \tag{12.104}$$

12.4.1.4 Traction Boundary Condition

Within a conducting material such as the electrodes attached on top and bottom of the dielectric layer, the electric field has to vanish. Hence, there is no contribution from the Maxwell stress in thickness direction, however, at the vertical boundaries where the film and the exterior (vacuum) field share a surface, continuity in the stress field must be ensured. Denoting with subscript 2 the plane components, for the plane first Piola-Kirchoff $[\![\mathbf{P}_2^{tot}]\!] \cdot \mathbf{N}_2 = \mathbf{0}$ must hold, where $\mathbf{N}_2$ is the in plane unit normal vector at the vertical edges. In addition, we claim also continuity of $\mathbf{F}_2$ as well as $\mathbf{S}_2^{tot}$ at the interface. For this reason, we can demand the continuity condition equivalently by taking the second Piola-Kirchoff stress, and its Maxwell stress version in air $\mathbf{S}_2^M$.

Let us discuss the jump condition in more detail, for the dielectric material the full contribution of the augmented free energy applies, while in the exterior field, all constituents arising from the presence of material vanish, hence the only remaining term is the augmentation term:

$$\rho_0\Omega_{Dielectric} = \rho_0\Omega_2, \qquad \rho_0\Omega_{exterior\ field} = \rho_0\psi_{aug}. \tag{12.105}$$

The Maxwell stress tensor is obtained by differentiating the augmented free energy, and using $\mathbf{C}^{-1} = \mathbf{C}_2^{-1} + C_{33}^{-1}\mathbf{mm}$ with $\boldsymbol{\mathcal{E}}\cdot\boldsymbol{\mathcal{E}} = \mathcal{E}_3^2$ and $\boldsymbol{\mathcal{E}}\boldsymbol{\mathcal{E}} = \mathcal{E}_3^2\mathbf{mm}$:

$$\begin{aligned}\mathbf{S}^M &= 2\frac{\partial\psi_{aug}}{\partial\mathbf{C}} = \frac{1}{2}\varepsilon_0 J\mathbf{C}^{-1}\cdot\mathcal{E}_3\mathcal{E}_3\mathbf{mm}\cdot\mathbf{C}^{-1} - \frac{1}{2}\varepsilon_0 J\mathbf{C}^{-1}\cdot\mathcal{E}_3^2\cdot\mathbf{C}^{-1}\\ &= \frac{1}{2}\varepsilon_0 J C_{33}^{-2}\mathcal{E}_3^2\mathbf{mm} - \frac{1}{2}\varepsilon_0 J\mathcal{E}_3^2\mathbf{C}_2^{-2},\end{aligned} \tag{12.106}$$

such that the plane part is $\mathbf{S}_2^M = \mathbf{S}^M\cdot\mathbf{I}_2$.

We use $\mathbf{P}_2^{tot} = \mathbf{F}_2\cdot\mathbf{S}_2^{tot}$ in the dielectric elastomer, and by writing the equilibrium at the surface with the plane Maxwell stress tensor $\mathbf{P}_2^M = \mathbf{F}_2\cdot\mathbf{S}_2^M$ the traction boundary condition is found to:

$$\mathbf{F}_2\cdot\mathbf{S}_2^{tot}\cdot\mathbf{N}_2 = \mathbf{F}_2\cdot\mathbf{S}_2^M\cdot\mathbf{N}_2 = -\mathbf{F}_2\cdot\frac{1}{2}\varepsilon_0 J\mathcal{E}_3^2\mathbf{C}_2^{-2}\cdot\mathbf{N}_2. \tag{12.107}$$

12.5 Electromechanical Stability

Still, the electrostrictive material parameter c_1 remains unknown, in order to define c_1 one has to rely on experimental measurements. However, proper identification of a coefficient associated with electrostriction is difficult as also the strain coming from Maxwell effect is quadratic in the electric field. Hence, a direct comparison to experimental data is not possible, and a proper conversion from the measured value to the electrostrictive parameter c_1 has to be applied. Diaconu and Dorohoi (2005) used a parameter M to relate the experimental dependence of the measured strain on the applied electric field. We will make use of the data set obtained there and proceed by specifying the mathematical model to the problem from the measurements.

We consider a thin plate, which is free to deform in plane upon application of an external electric field in thickness direction $\mathcal{E}_3 = V/h$, where V is the magnitude of the applied voltage, and h the thickness of the plate. For this problem, a spherical right Cauchy-Green tensor, with the same in plane stretches due to the homogeneous deformation, applies, hence $\mathbf{C}_2 = C\mathbf{I}_2$ with $C = \lambda^2$. Where $\lambda = \lambda_1 = \lambda_2$ denotes the principal stretch in both in-plane directions. We use $\mathbf{I}_2 = \mathbf{I} - \mathbf{mm}$ the plane identity tensor, and write the invariants for this problem,

$$I_{\mathbf{C}_2} = 2C = 2\lambda^2 \quad , \qquad III_{\mathbf{C}_2} = C^2 = \lambda^4. \tag{12.108}$$

By using the neo-Hookean strain energy function, specified for incompressible materials, the expression for the augmented free energy gets,

$$\rho_0 \Omega_2 = \frac{\mu}{2}(2\lambda_{el}^{-2}\lambda^2 + \lambda_{el}^4\lambda^{-4} - 3) - \frac{1}{2}\varepsilon\lambda^4\mathcal{E}_3^2, \tag{12.109}$$

where $\varepsilon = \varepsilon_0(1+\chi) = \varepsilon_0\varepsilon_r$. The plane total second Piola-Kirchoff stress tensor is obtained by using implicit differentiation, and the relation $\partial\mathbf{C}_2/\partial\lambda = 2\lambda\mathbf{I}_2$:

$$\mathbf{S}_2^{tot} = 2\rho_0\frac{\partial\Omega_2}{\partial\mathbf{C}_2} = 2\rho_0\frac{\partial\Omega_2}{\partial\lambda}\frac{\partial\lambda}{\partial\mathbf{C}_2} = \rho_0\frac{1}{\lambda}\frac{\partial\Omega_2}{\partial\lambda}\mathbf{I}_2. \tag{12.110}$$

Next, the contributing stress from the exterior field needs to be incorporated, by making use of the traction boundary condition $\mathbf{F}_2\cdot\mathbf{S}_2^{tot}\cdot\mathbf{N}_2 = \mathbf{F}_2\cdot\mathbf{S}_2^{M}\cdot\mathbf{N}_2$ with $\mathbf{F}_2 = \lambda\mathbf{I}_2$ and $\mathbf{S}_2^M = -\frac{1}{2}\varepsilon_0\mathcal{E}_3^2\lambda^{-4}\mathbf{I}_2$:

$$\rho_0\frac{\partial\Omega_2}{\partial\lambda}\mathbf{N}_2 = -\frac{1}{2}\varepsilon_0\mathcal{E}_3^2\lambda^{-3}\mathbf{N}_2 \tag{12.111}$$

Integration yields the augmentation energy $\rho_0\Omega_2^{aug} = \frac{1}{4}\varepsilon_0\lambda^{-2}\mathcal{E}_3^2$, such that the traction boundary condition can be expressed by an overall plane energy function $\bar{\Omega}_2$

$$\rho_0\bar{\Omega}_2 = \rho_0\Omega_2 - \rho_0\Omega_2^{aug} = \rho_0\Omega_2 - \frac{1}{4}\varepsilon_0\lambda^{-2}\mathcal{E}_3^2. \tag{12.112}$$

Moreover, the traction boundary condition can be equally written in terms of a new overall second Piola-Kirchoff type stress tensor $\bar{\mathbf{S}}_2\cdot\mathbf{N}_2 = 0$, where

$$\bar{\mathbf{S}}_2 = \frac{1}{\lambda}\left(\frac{\partial\Omega_2}{\partial\lambda} + \frac{1}{2}\varepsilon_0\lambda^{-3}\mathcal{E}_3^2\right)\mathbf{I}_2 = \frac{1}{\lambda}\frac{\partial\bar{\Omega}_2}{\partial\lambda}\mathbf{I}_2 \tag{12.113}$$

has to vanish in the whole plate. We can now proceed to the stability analyses by writing the equilibrium condition in form of the Principle of Gibbs (Ziegler, 1998)

$$\frac{\partial\bar{\Omega}_2}{\partial\lambda} = 0, \tag{12.114}$$

yielding the equilibrium stretches $\lambda = \lambda_0$. Furthermore, one can judge on the stability of these stretches by making use of the Dirichlet stability criterion for conservative problems,

$$\left.\frac{\partial^2\bar{\Omega}_2}{\partial\lambda^2}\right|_{\lambda_0} > 0. \tag{12.115}$$

We proceed by specifying the overall free energy for the incompressible neo-Hookean material and write the equilibrium condition:

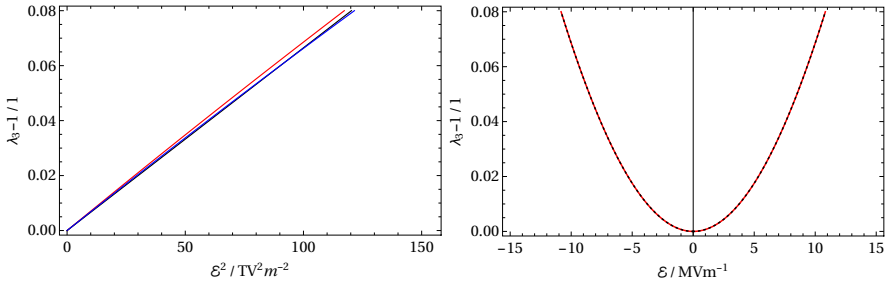

Fig. 12.6: Equilibrium Biot strain for Voltage driven actuation, left the linear course of the strain upon the square electric field, right the quadratic course upon the linear electric field. The red line corresponds to the full model, black dotted neglects Maxwell effect, black solid line linearized problem, blue line electric stretch.

$$\rho_0 \bar{\Omega}_2 = \frac{\mu}{2}(2\lambda_{el}^{-2}\lambda^2 + \lambda_{el}^4\lambda^{-4} - 3) - \frac{1}{2}\varepsilon\lambda^4\mathcal{E}_3^2 - \frac{1}{4}\varepsilon_0\mathcal{E}_3^2\lambda^{-2}, \tag{12.116}$$

$$(\lambda_{el}^{-2}\lambda - \lambda_{el}^4\lambda^{-5}) - \lambda^3\left(1 - \frac{1}{4\varepsilon_r}\lambda^{-6}\right)\frac{\varepsilon}{\mu}\mathcal{E}_3^2 = 0 \tag{12.117}$$

where we have used $\varepsilon = \varepsilon_0(1+\chi) = \varepsilon_0\varepsilon_r$. Finally, we close the theoretical part by specifying the material parameter using the same polyurethane elastomer reported in Diaconu and Dorohoi (2005), with $\varepsilon_r = 8.8, Y = 3\mu$ and $Y = 3.6$MPa. Multiplication of Eq. (12.117) with λ and using $\lambda^2 = \lambda_3^{-1}$, which was obtained from the incompressibility condition, yields:

$$(\lambda_{el}^{-2}\lambda_3^{-1} - \lambda_{el}^4\lambda_3^2) - \lambda_3^{-2}\left(1 - \frac{1}{4\varepsilon_r}\lambda_3^3\right)\frac{\varepsilon}{\mu}\mathcal{E}_3^2 = F(\lambda_3, \mathcal{E}_3^2) = 0. \tag{12.118}$$

In the reference, quadratic dependence of the strains was observed at low electric field strength. To ensure comparability, we approximate the equilibrium condition $F(\lambda_3, \mathcal{E}_3^2)$ in the vicinity of $\lambda_3 \approx 1$ and $\mathcal{E}_3^2 \approx 0$, leading to the linear relation

$$\varepsilon_3 = -\left(\frac{\varepsilon(1 - \frac{1}{4\varepsilon_r})}{3\mu} - c_1\right)\mathcal{E}_3^2 = -M\mathcal{E}_3^2, \tag{12.119}$$

which allows for incorporating the measured value M. Diaconu and Dorohoi (2005) measured a value of $M = 7.07 \times 10^{-16}\text{m}^2\text{V}^{-2}$, which is a parameter, that still carries the contribution from the Maxwell effect. Here, we have introduced the Biot strain measure $\varepsilon_3 = \lambda_3 - 1$. By solving the linearized system the electrostrictive parameter $c_1 = -6.86 \times 10^{-16}\text{m}^2\text{V}^{-2}$ can be found. Hence, the contribution of the Maxwell effect is 3.066%, which agrees well with the value 3.07% given in Diaconu and Dorohoi (2005). In Fig. 12.6, the equilibrium Biot strain against the square of the electric field (left), and right the equilibrium Biot strain against the linear electric

field is shown. Clearly, the electrostrictive behavior can be observed. On the left side, the solid red line corresponds to the strain response of the whole problem, the black line shows the linearized system giveni Eq. (12.119), and the blue line corresponds only to the electrical Biot strain $\varepsilon_{el,3} = \lambda_{el}^{-2} - 1 = \exp(c_1 \mathcal{E}_3^2) - 1$. However, for small strains, almost no difference is visible. In the right figure, the red line corresponds to the problem discussed in this paper, accounting for the Maxwell effect, while the black dotted line shows the solution of the problem if the Maxwell stress arising from the traction boundary is neglected. Results show, there is almost no difference, due to the prevailing electrostrictive effect.

Increasing the field strength and allowing also higher strains, the different curves deviate from each other, unveiling the nature of each contributor, see Fig. 12.7. In the left figure, again the red as well as the black dotted line are almost equal, as the Maxwell effect from the boundary condition is negligibly small. The black solid line is again the linear solution. Attention should be taken to the blue line, which shows the electric stretch $\lambda_{el,3} - 1$ in thickness direction; its nature unveils the impact of the electrostrictive effect, which for the polyurethane material under consideration seems to be of great importance.

Strains of such magnitude cannot be obtained in nature, as at some point the system looses its stability. On the right hand side in Fig. 12.7 the equilibrium strains (red, black dotted and brown) and the stability margin in black are presented. The theoretical bounds of equilibrium stretches, and the corresponding field strength can be obtained by using the Dirichlet criteria and are acquired for several cases:

- Considering both Maxwell effect and electrostriction, with the following equilibrium condition, and Dirichlet stability criterion

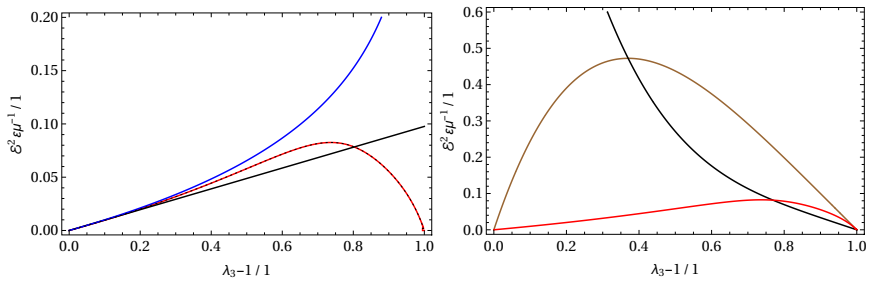

Fig. 12.7: Equilibrium Biot strain considering higher electric fields, up to Biot strain =1. On the right side, the red line corresponds to the case with electrostriction, brown line only electrostatic forces, black line stability margin. Left: red and black dotted line, equilibrium strain accounting for electrostriction with and without Maxwell boundary term, black solid line linearized strain, blue line electrical part of the Biot strain $\varepsilon_{el,3}$.

$$(\lambda\lambda_{el}^{-2} - \lambda^{-5}\lambda_{el}^{4}) - \lambda^{3}\left(1 - \frac{1}{4\varepsilon_r}\lambda^{-6}\right)\frac{\varepsilon}{\mu}\mathcal{E}_3^2 = 0, \tag{12.120}$$

$$(\lambda_{el}^{-2} + 5\lambda^{-6}\lambda_{el}^{4}) - 3\lambda^{2}\left(1 + \frac{1}{4\varepsilon_r}\lambda^{-6}\right)\frac{\varepsilon}{\mu}\mathcal{E}_3^2 = 0. \tag{12.121}$$

- Neglecting the Maxwell effect from the boundary conditions, which follows by dropping the terms with the relative permittivity.

$$(\lambda\lambda_{el}^{-2} - \lambda^{-5}\lambda_{el}^{4}) - \lambda^{3}\frac{\varepsilon}{\mu}\mathcal{E}_3^2 = 0, \tag{12.122}$$

$$(\lambda_{el}^{-2} + 5\lambda^{-6}\lambda_{el}^{4}) - 3\lambda^{2}\frac{\varepsilon}{\mu}\mathcal{E}_3^2 = 0. \tag{12.123}$$

- In order to draw a comparison to the impact of the electrostrictive effect the case for pure electrostatic force, shown in Krommer et al (2016):

$$(\lambda - \lambda^{-5}) - \lambda^{3}\frac{\varepsilon}{\mu}\mathcal{E}_3^2 = 0, \tag{12.124}$$

$$(1 + 5\lambda^{-6}) - 3\lambda^{2}\frac{\varepsilon}{\mu}\mathcal{E}_3^2 = 0. \tag{12.125}$$

The critical stretch in thickness direction is obtained by inserting $\lambda_3 = \lambda^{-2}$, and solving the equilibrium condition. Using this critical value thereafter in the Dirichlet criterion yields the critical electric field. For the last two discussed cases, the critical stretch λ_{crit} along with the critical electric field $\mathcal{E}_{3,crit}$ are

$$\lambda_{crit,ES\,Force}^{-6} = \frac{1}{4} \rightarrow \sqrt{\frac{\varepsilon}{\mu}}\mathcal{E}_{3,crit,ES\,Force} = \frac{\sqrt{3}}{4^{\frac{2}{3}}} = 0.687, \tag{12.126}$$

$$\left(\frac{\lambda_{crit,DE}}{\lambda_{e,crit,DE}}\right)^{-6} = \frac{1}{4} \rightarrow \sqrt{\frac{\varepsilon}{\mu}}(\exp(-c_1\mathcal{E}_{3,crit,DE}^2)\mathcal{E}_{3,crit,DE}) = \frac{\sqrt{3}}{4^{\frac{2}{3}}} = 0.687. \tag{12.127}$$

First the critical stretch and electric field for the electrostatic force is presented in Eq.(12.126). For the problem accounting for electrostriction but dropping the Maxwell effect, a very similar correlation can be found, see Eq.(12.127). However, if considering both electrostriction and Maxwell effect, the limiting criteria turn out to be more complicated

$$\lambda_{crit}^{6} = \left(-1 + 8\lambda_{el,crit}^{6}\varepsilon_r + \sqrt{1 - 20\lambda_{el,crit}^{6}\varepsilon_r + 64\lambda_{el,crit}^{12}\varepsilon_r^2}\right)(4\varepsilon_r)^{-1}, \tag{12.128}$$

which upon using the abbreviations $k = (-1 + 8\varepsilon_r\lambda_{el,crit}^{6})$ and $s = \sqrt{k^2 - 4\varepsilon_r\lambda_{el,crit}^{6}}$ yields

$$\lambda_{crit}^{6} = (k + s)(4\varepsilon_r)^{-1}, \tag{12.129}$$

and

$$\sqrt{\frac{\varepsilon}{\mu}}\mathcal{E}_{crit} = \sqrt{\frac{2^{\frac{2}{3}}(\lambda_{crit}^6 - \lambda_{el,crit}^6)4\varepsilon_r}{\lambda_{el,crit}^2(k-1+s)(k\varepsilon_r^{-1}+s\varepsilon_r^{-1})^{\frac{1}{3}}}}. \tag{12.130}$$

However, as it has turned out that the Maxwell effect seems to have minimal effect on the solution, Fig. 12.7 indicates almost the same critical values, as for the case where Maxwell effect is dropped. This can be judged, as the black line giving the stability margin crosses the equilibrium lines where the tangent gets horizontally, in both cases.

12.5.1 Stiffening Effect of Electrodes

Up to now, the investigations presented above, were made under the assumption, that electrodes attached to the EAP film do not interact with the EAP other than supplying an electric field. As this assumption is fairly crude, the last part is devoted to the problem of a homogeneously deformed plate with electrodes attached on top and bottom, see Fig. 12.8.

We investigate the case of compliant electrodes made of steel, hence we use a St. Vernant-Kirchoff material model for the constitutive part of the electrodes. Due to the in plane deformation a membrane state of stress prevails. We use $\varepsilon_3 = \lambda_3 - 1$, the strain component in thickness direction as it correlates to the in plane stretches by the relation $\lambda^2 = \lambda_3^{-1}$.

$$u_{m,2} = \frac{1}{2}(A(\mathrm{tr}\varepsilon_3\ \mathrm{tr}\varepsilon_3 - 2(1-\nu)\mathrm{det}\varepsilon_3)) \quad , \qquad A = \int_{-H/2}^{-h/2} \frac{Y}{(1+\nu)(1-\nu)}dz \tag{12.131}$$

A denotes the tension stiffness, which is given for the lower electrode, but correspondingly applies to the upper electrode by changing the sign of the bounds. $Y = 210\times10^9\mathrm{Nm}^{-2}$ denotes the elastic modulus, and $\nu = 0.33$ the Poisson's ratio of steel. We incorporate the electrodes into our constitutive model of the dielectric by simply adding the strain energy $u_{m,2}$ to the augmented free energy function Ω_2 and

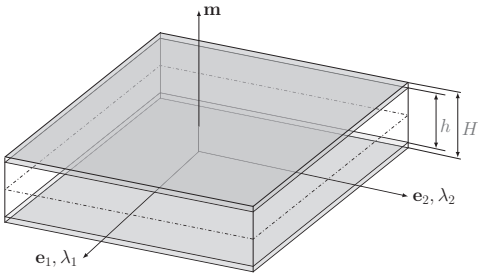

Fig. 12.8 Electro-active polyurethan plate with electrodes at the height $\frac{1}{2}(H-h)$ attached on top and bottom.

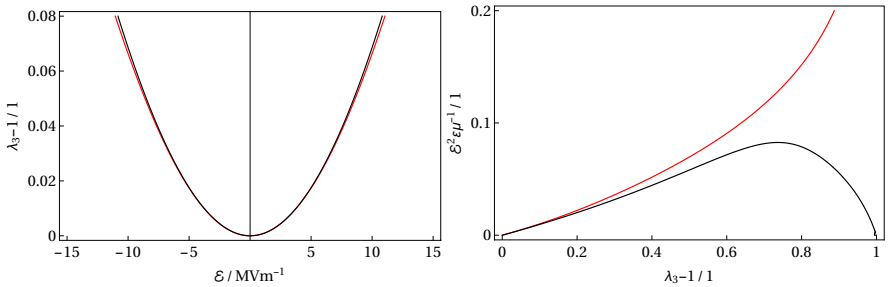

Fig. 12.9: In red, the equilibrium Biot strain for the plate with electrodes, black plate problem without electrodes.

find

$$\Omega_{2,ges} = \frac{1}{2}h\Big(-\mathcal{E}_3^2\varepsilon_0\varepsilon_r\lambda^4 + (2\lambda^2\lambda_{el}^{-2} + \lambda^{-4}\lambda_{el}^4 - 3)\mu\Big) + \frac{(\frac{H}{2} - \frac{h}{2})Y(\lambda^2-1)^2}{(1+\nu)(1-\nu)}. \quad (12.132)$$

As the Maxwell effect coming from the vertical boundaries has been shown to be negligible, we dropped the term, and proceed by determining the electrostrictive coefficient c_1. Again the correlation of the experimentally known data for M to our parameter c_1 is found in a linear approximation of the equilibrium equation $\frac{\partial\Omega_{2,ges}}{\partial\lambda} = 0$, in the vicinity of $\lambda_3 \approx 1$ and $\mathcal{E}_3 \approx 0$ to

$$\varepsilon_3 = -h(\nu^2-1)\left(\frac{(\varepsilon + 3c_1\mu)}{h(\mathrm{Y} + 3\mu(\nu^2-1)) - HY}\right)\mathcal{E}_3^2 = -M\mathcal{E}_3^2. \quad (12.133)$$

At first, the difference in the electrostrictive behavior and the stable stretch configuration is investigated. As for the experimental value $M = 7.07\times10^{-16}\mathrm{m}^2\mathrm{V}^{-2}$ no thickness of the sample was reported, we assumed a value of 1μm. The thickness of the electrodes is derived by using the relation $H = 1{,}00002h$, which results in an electrostrictive coefficient of $c_1 = -9.94\times10^{-15}\mathrm{m}^2\mathrm{V}^{-2}$. Figure Fig. 12.9 shows the resulting equilibrium Biot strain, the electrostrictive behavior upon the linear electric field is still visible, furthermore, the stiffening effect of the electrodes tends to stabilize the problem (red line) as no horizontal tangent is present any more. Additionally to the single parameter set we are using so far, Diaconu et al (2006) published experimental results for another set of fife different materials, given in Tab. 12.1. Again only results for small electric fields and strains are presented in Diaconu et al (2006). At this regime the results given in Fig. 12.10, upon the square electric field agree very well, however at higher fields, experimental results tend to deviate nonlinearly, which is not visible from the results of our model. The black line in Fig. 12.10 corresponds to the pure EAP model without electrodes. Finally, the results upon increasing the thickness of the electrodes by the factor 10 are presented in Fig.12.11. The electrostictive behavior remains for a broadened electric field.

Table 12.1: Material parameter taken from Diaconu et al (2006), at an electric field of $\mathcal{E}_3 = 4.5\mathrm{MVm}^{-1}$

Sample	Thickness [μm]	Y [MPa]	$M \times 10^{16}$ [$\mathrm{m}^2\mathrm{V}^{-2}$]	$c_1 \times 10^{16}$	Color
1	15	6.19	3.00	2.87	Red
2	27	25.46	5.00	4.97	Green
3	33	51.03	8.00	7.89	Blue
4	49	52.39	12.28	12.27	Brown
5	110	59.5	8.92	8.91	Magenta

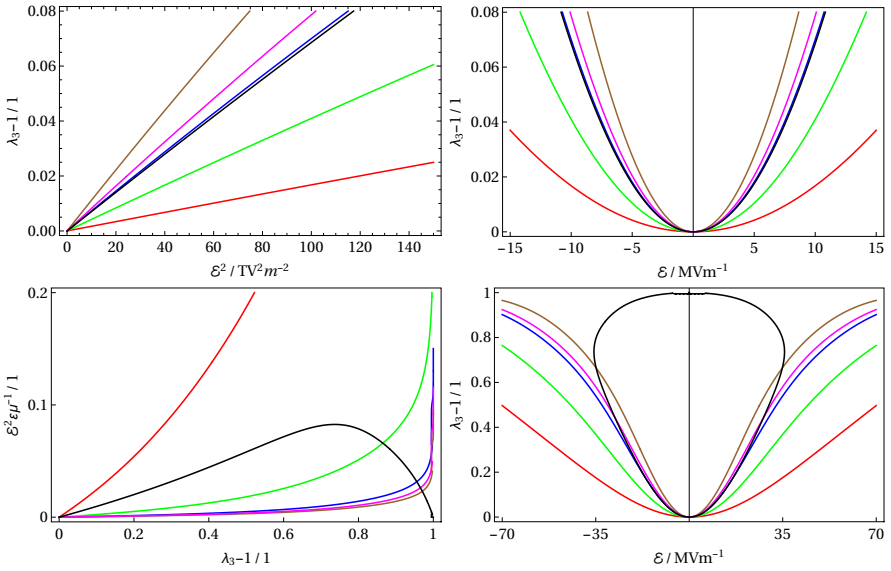

Fig. 12.10: Response at small electric fields and strains above, and at large fields bottom.

While if the thickness is decreased, the curves tend to the solution without electrodes, presented in black.

12.6 Conclusion and Outlook

The constitutive modeling framework in the field of nonlinear electro-elasticity has been presented in details, starting by introducing the electrostatic field quantities, the macroscopic balance equations were derived and with clear focus on the multiplicative decomposition of the deformation gradient applied to the effect of electrostriction.

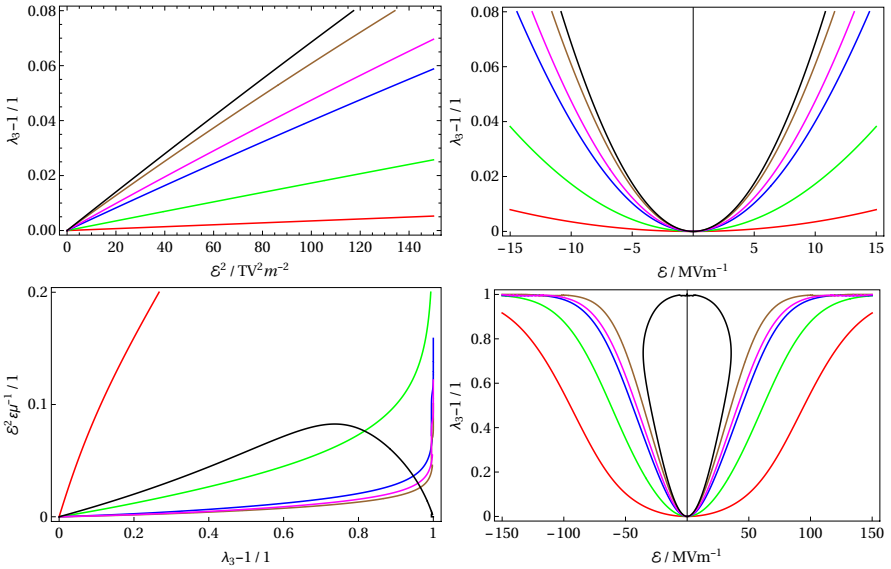

Fig. 12.11: Increasing the thickness of the electrodes by factor 10.

The application on a simple homogeneously deformed plate unveiled, that the electrostrictive effect has a considerable effect on the actuation behavior of electroactive polymers, and can be enhanced by the choice of dielectric material. In the near future, the presented model is extended to geometrically nonlinear shells and the implementation into a geometrically nonlinear finite element code, in order to apply the model to more general problems. Special attention will be paid on the choice of the specific constitutive law for the electrical part of the deformation gradient, as this component offers access to easily incorporate different models and requires some more investigations.

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