THEORY OF DIELECTRIC ELASTOMERS⋆⋆

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ABSTRACT In response to a stimulus, a soft material deforms, and the deformation provides
a function. We call such a material a soft active material (SAM). This review focuses on one
class of soft active materials: dielectric elastomers. When a membrane of a dielectric elastomer
is subject to a voltage through its thickness, the membrane reduces thickness and expands area,
possibly straining over 100%. The dielectric elastomers are being developed as transducers for
broad applications, including soft robots, adaptive optics, Braille displays, and electric generators.
This paper reviews the theory of dielectric elastomers, developed within continuum mechanics
and thermodynamics, and motivated by molecular pictures and empirical observations. The the-
ory couples large deformation and electric potential, and describes nonlinear and nonequilibrium
behavior, such as electromechanical instability and viscoelasticity. The theory enables the finite
element method to simulate transducers of realistic configurations, predicts the efficiency of elec-
tromechanical energy conversion, and suggests alternative routes to achieve giant voltage-induced
deformation. It is hoped that the theory will aid in the creation of materials and devices.

KEY WORDS soft active material, dielectric elastomer, electromechanical instability, large de-
formation, transducer

I. INTRODUCTION

1.1. Soft Active Materials for Soft Machines

The convergence of parts of biology and engineering has created exciting opportunities of discovery,
invention and commercialization. The overarching themes include using engineering methods to advance
biology, combining biology and engineering to invent medical procedures, and mimicking biology to
create engineering devices.

Machines in engineering use mostly hard materials, while machines in nature are often soft. What
does softness impart to the life of animals and plants? A conspicuous feature of life is to receive and
process information from the environment, and then move. The movements are responsible for diverse
functions, far beyond the function of going from place to place. For example, an octopus can change
its color at an astonishing speed, for camouflage and signaling. This rapid change in color is mediated
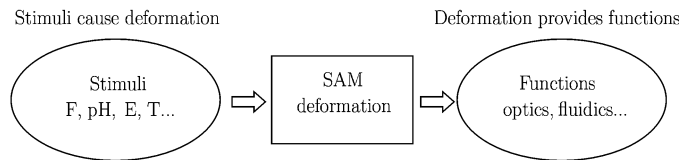
Fig. 1. The environment affects a material through diverse stimuli, such as a force, an electric field, a change in pH, and a change in temperature. In response to a stimulus, a soft active material (SAM) deforms. The deformation provides a function, such as a change in color and a change in flow rate.

by thousands of pigment-containing sacs. Attached to the periphery of each sac are dozens of radial muscles. By contracting or relaxing the muscles, the sac increases or decreases in area in less than a second. An expanded sac may be up to about 1 mm in diameter, showing the color. A retracted sac may be down to about 0.1 mm in diameter, barely visible to the naked eye[1].

As another example, in response to a change in the concentration of salt, a plant can change the rate of water flowing through the xylem. This regulation of flow is thought to be mediated by pectins, polysaccharides that are used to make jellies and jams. Pectins are long polymers, crosslinked into a network. The network can imbibe a large amount of water and swell many times its own volume, resulting in a hydrogel. The amount of swelling changes in response to a change in the concentration of salt. The change in the volume of the hydrogel alters the size of the microchannels in the xylem, regulating the rate of flow[2].

The above examples concerning animals and plants are intriguing. But many more examples are everywhere around and inside us. Consider the accommodation of the eye, the beating of the heart, the sound shaped by the vocal folds, and the sound in the ear. Abstracting these biological soft machines, we may say that a stimulus causes a material to deform, and the deformation provides a function (Fig.1). Connecting the stimulus and the function is the material capable of large deformation in response to a stimulus. We call such a material a soft active material (SAM).

An exciting field of engineering is emerging that uses soft active materials to create soft machines. Soft active materials in engineering are indeed apt in mimicking the salient feature of life: movements in response to stimuli. An electric field can cause an elastomer to stretch several times its length. A change in pH can cause a hydrogel to swell many times its volume. These soft active materials are being developed for diverse applications, including soft robots, adaptive optics, self-regulating fluidics, programmable haptic surfaces, electric generators, and oilfield management[3–8].

Research in soft active materials has once again brought mechanics to the forefront of human creativity. The familiar language finds new expressions, and deep thoughts are stimulated by new experience. To participate in advancing the field of soft active materials and soft machines effectively, mechanicians must retool our laboratories and our software, as well as adapt our theories.

The biological phenomena, as well as the tantalizing engineering applications, have motivated the development of theories of diverse soft active materials, including dielectric elastomers[9–13], elastomeric gels[14–19], polyelectrolytes[20, 21], pH-sensitive hydrogels[22–24], and temperature-sensitive hydrogels[25]. The theories attempt to answer commonly asked questions. How do mechanics, chemistry, and electrostatics work together to generate large deformation? What characteristics of the materials optimize their functions? How do molecular processes affect macroscopic behavior? How efficiently can a material convert energy from one form to another? The theories are being implemented in software, so that they can become broadly useful in the creation of materials and devices.

1.2. Dielectric Elastomers

This review will focus on a class of soft active materials: dielectric elastomers. All materials contain electrons and ions—charged particles that move in response to an applied voltage. In a conductor, electrons or ions can move over macroscopic distances. By contrast, in a dielectric, the charged particles move relative to one another by small distances. In the dielectric, the two processes—deformation and polarization—are inherently coupled. All dielectrics are electroactive.

Figure 2 illustrates the principle of operation of a dielectric elastomer transducer. A membrane of a dielectric elastomer is sandwiched between two electrodes. For the dielectric elastomer to deform
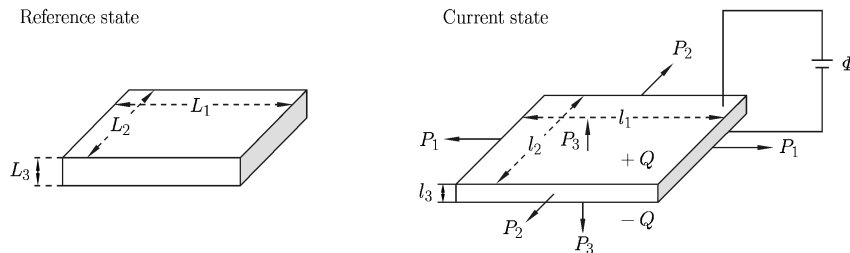
Fig. 2. A dielectric elastomer in the reference state and in a current state.

substantially, the electrodes are made of an even softer substance, with mechanical stiffness lower than that of the dielectric elastomer. A commonly used substance for electrodes is carbon grease. When the transducer is subject to a voltage, charge flows through an external conducting wire from one electrode to the other. The charges of the opposite signs on the two electrodes cause the membrane to deform.

It was discovered a decade ago that an applied voltage may cause dielectric elastomers to strain over 100%[3]. Because of this large strain, dielectric elastomers are often called artificial muscles. In addition to large voltage-induced strains, other desirable attributes of dielectric elastomers include fast response, no noise, light weight, and low cost. The discovery has inspired intense development of dielectric elastomers as transducers for diverse applications[26–28]. The discovery has also inspired renewed interest in the theory of coupled large deformation and electric field[9–13].

This paper reviews the theory of dielectric elastomers. §II describes the thermodynamics of a transducer of two independent variations. Emphasis is placed on basic ideas: states of the transducer, cyclic operation of the transducer, region of allowable states, equations of state, stability of a state, and nonconvex free-energy function. These ideas are described in both analytical and geometrical terms. §III develops the theory of homogeneous fields. After setting up a thermodynamic framework for electromechanical coupling, we consider several specific material models: a vacuum as an elastic dielectric of vanishing stiffness, incompressible materials, ideal dielectric elastomers, electrostrictive materials, and nonlinear dielectrics. §IV applies nonequilibrium thermodynamics to dissipative processes, such as viscoelasticity, dielectric relaxation, and electrical conduction. §V discusses electromechanical instability, both as a mode of failure and as a means to achieve giant voltage-induced deformation. §VI outlines the theory of inhomogeneous fields. The condition for thermodynamic equilibrium is formulated in terms of a variational statement, as well as in terms of partial differential equations. The model of ideal dielectric elastomers is described. Also described is a finite element method for analyzing elastic dielectric membranes of arbitrary shapes. We perturb a state of static equilibrium to analyze oscillation and bifurcation. We examine the conditions of equilibrium for coexistent phases.

II. THERMODYANMICS OF A TRANSDUCER

It is well appreciated that most fundamental concepts of thermodynamics can be illustrated by using a fluid—a system capable of two independent variations (thermal and mechanical). The fluid can serve as the working substance of many devices for thermomechanical energy conversion, including engines and refrigerators. By analogy, the same concepts of thermodynamics are summarized here for electromechanical transducers. We are mostly concerned with transducers made of elastomers, which are highly entropic. To describe such a transducer with two independent variations (electrical and mechanical), here we limit ourselves to isothermal processes, removing temperature from explicit consideration.

2.1. States of a Transducer

Figure 3 illustrates a transducer, consisting of a dielectric that separates two electrodes. The transducer is subject to a force P, represented by a weight. The two electrodes are connected through a conducting wire to a voltage Φ, represented by a battery. The weight moves by distance l, and the battery pumps charge Q to flow through the conducting wire from one electrode to the other.

Because the force P and the voltage Φ can be applied independently, the transducer is capable of two independent variations. Consequently, the states of the transducer can be represented graphically on a plane. The two coordinates of the plane may be chosen from variables such as P, Φ, l and Q. Figure 4, for
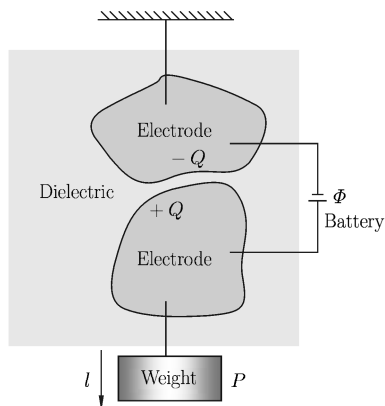
Fig. 3. A transducer consists of a dielectric separating two electrodes. The transducer is subject to a force, represented by a weight P. The two electrodes are connected through a conducting wire to a battery of voltage Φ. The weight moves by distance l, and the battery pumps charge Q from one electrode to the other.
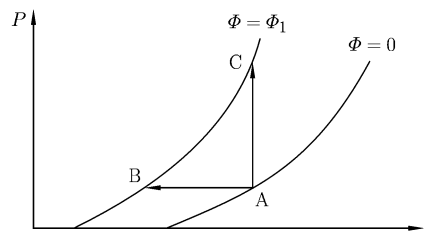
Fig. 4. In a plane with force and displacement as coordinates, a point represents a state of a transducer. A curve of a constant voltage is the force-displacement curve measured when the transducer is subject to a constant voltage.

example, illustrates a plane with the force P and the displacement l as the coordinates. A point in the plane represents a state of the transducer. Also plotted in the plane are two force-displacement curves of the transducer, each curve being measured as the transducer deforms under a constant voltage.

When the transducer is subject to a constant weight, but the voltage is changed from 0 to Φ_1, the transducer changes from state A to state B, lifting the weight. When the displacement l is held constant, a change in the voltage causes the transducer to change from state A to state C, with accompanying change in the force.

We could have also plotted on the (l, P) plane curves of constant values of charge. Each curve of a constant charge would be the force-displacement curve of the transducer measured under the open-circuit condition, when the two electrodes maintain a fixed amount of charge during deformation.

Following Gibbs's graphical method for the thermodynamics of fluids[29], we may choose any two of the four variables P, Φ, l and Q as coordinates. Each choice represents the transducer on a different plane. All these planes represent the same states of the transducer, because the transducer is capable of only two independent variations. Nonetheless, different planes emphasize different attributes of the states. For example, the (P, Φ) plane may be used to indicate loading conditions, while the (l, Q) plane may be used to indicate kinematic conditions. The (l, Φ) plane is often used to report the voltage-induced deformation, while the (P, Q) plane may be used to report force-induced charge. In mathematical terms, mapping the states of the transducer from one plane to another is a change of variables, or a transformation of coordinates. The mapping is carried out by using equations of state, to be described in §2.4.

2.2. Cyclic Operations of a Transducer

Many uses of transducers involve cyclic changes of states. A particular cycle of states is illustrated in Fig.5, on the (Q, Φ) plane. To operate a transducer in this cycle, we will need two batteries: one at a low voltage Φ_L, and the other at a high voltage Φ_H. The four sides of the rectangular cycle shown in Fig.5 represent the following processes:

(1) In changing from state A to state B, the transducer is connected to the battery of the low voltage, Φ_L. A change in the applied force reduces the spacing between the two electrodes, causing the charge on the electrodes to increase.

(2) In changing from state B to state C, the transducer is under an open-circuit condition and the electrodes maintain the constant charge Q_H. A change in the applied force increases the spacing between the two electrodes, raising the voltage to Φ_H.

(3) In changing from state C to state D, the transducer is connected to the battery of the high voltage, Φ_H. A change in the applied force increases the spacing between the two electrodes, causing the charge on the two electrodes to decrease.

(4) In changing from state D to state A, the transducer is under an open-circuit condition and the electrodes maintain the constant charge Q_L. A change in the applied force decreases the spacing between the two electrodes, lowering the voltage to Φ_L.

This cycle of operation of an electromechanical transducer is analogous to the Carnot cycle, provided we replace voltage with temperature, and replace charge with entropy. During the cycle, the transducer receives mechanical work from the environment, draws an amount of charge from the low-voltage battery, and deposits the same amount of charge to the high-voltage battery. Thus, the transducer is a generator, producing electric energy by receiving mechanical work. The mechanical work can be done, for example, by an animal or human during walking. The mechanical work can also be done on a large scale, for example, by ocean waves.

Indeed, a closed curve of any shape on the (Q, Φ) plane represents a cyclic operation of the transducer. To operate such a cycle would require a variable-voltage source. The amount of energy converted per cycle is given by the area enclosed by the cycle on the (Q, Φ) plane. When the states cycle counterclockwise on the (Q, Φ) plane, the transducer is a generator, converting mechanical energy to electrical energy. When the states cycle clockwise on the (Q, Φ) plane, the transducer is an actuator, converting electrical energy to mechanical energy.
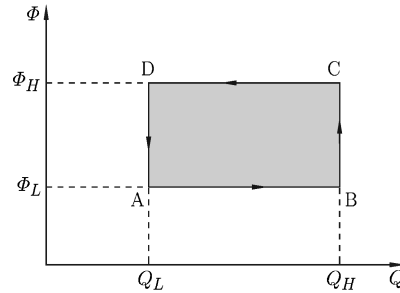

Fig. 5   In a plane with voltage and charge as coordinates, a point represents a state of a transducer. A use of the transducer typically involves a cyclic change of the state. The rectangle represents a cycle involving two levels of voltage and two values of charge.

Cyclic operation of a transducer can also be represented on the (l, P) plane. Figure 4 already contains force-displacement curves measured with two values of constant voltage. We could add force-displacement curves measured with two values of constant charge. The four curves would represent the same cycle of operation as that shown in Fig.5.

2.3. Modes of Failure and Region of Allowable States

A transducer may fail in multiple modes, such as mechanical rupture, electrical breakdown, electromechanical instability, and loss of tension[30–32]. The critical condition for each mode of failure can be represented on the (Q, Φ) plane by a curve. Curves of all modes of failure bound in the plane a region, which we call the region of allowable states of the transducer. Such graphic methods have been used to optimize actuators[33, 34] and calculate the maximal energy of conversion for generators[35–37]. Figure 6 shows an example[35]. By transformations of coordinates, we can represent the same modes of
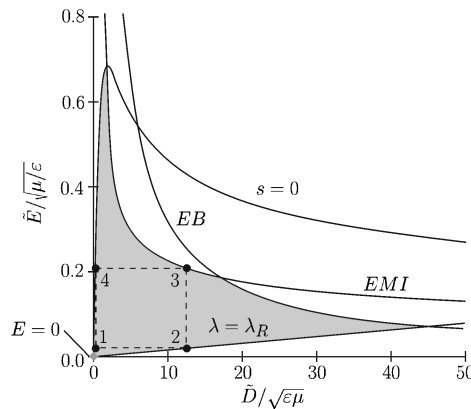
Fig. 6. A state of a dielectric membrane is represented by a point in the charge-voltage plane[35]. The coordinates are given in dimensionless forms, with the horizontal axis being the normalized charge, and the vertical axis being the normalized voltage. Plotted are curves representing various modes of failure: electrical breakdown (EB), electromechanical instability (EMI), loss of tension ($s = 0$), and rupture by stretch ($\lambda = \lambda_R$). These curves bound the region of allowable states of the transducer. A cycle involving two levels of voltage and two values of charge is represented by dotted lines.

failure on planes (l, P), (l, Q), etc.

2.4. Equations of State

We will analyze isothermal processes of a transducer, and remove temperature from explicit consideration. A main component of the transducer is an elastomer—a three-dimensional network of long and flexible polymer chains. The thermodynamic behavior of the transducer is highly entropic, characterized by the Helmholtz free energy, which we denote as F.

On dropping a small distance δl, the weight does work $P\delta l$. On pumping a small amount of charge δQ, the battery does work $\Phi\delta Q$. The force is work-conjugate to the displacement, and the voltage is work-conjugate to the charge. When the transducer equilibrates with the applied force and the applied voltage, the change in the free energy of the transducer equals the sum of the work done by the weight and the work done by the battery:

$$\delta F = P\delta l + \Phi\delta Q \tag{1}$$

This condition of equilibrium holds for arbitrary and independent small variations δl and δQ.

The two independent variables (l, Q) characterize the state of the transducer. The Helmholtz free energy of the transducer is a function of the two independent variables:

$$F = F(l, Q) \tag{2}$$

Associated with small variations δl and δQ, the free energy varies by

$$\delta F = \frac{\partial F(l, Q)}{\partial l}\delta l + \frac{\partial F(l, Q)}{\partial Q}\delta Q \tag{3}$$

A comparison of Eqs.(1) and (3) gives

$$\left[\frac{\partial F(l, Q)}{\partial l} - P\right]\delta l + \left[\frac{\partial F(l, Q)}{\partial Q} - \Phi\right]\delta Q = 0 \tag{4}$$

When the transducer equilibrates with the weight and the battery, the condition of equilibrium (4) holds for independent and arbitrary variations δl and δQ. Consequently, in equilibrium, the coefficients of the two variations in Eq.(4) both vanish, giving

$$P = \frac{\partial F(l, Q)}{\partial l} \tag{5}$$

$$\Phi = \frac{\partial F(l, Q)}{\partial Q} \tag{6}$$

Once the free-energy function $F(l, Q)$ is known, Eqs.(5) and (6) express P and Φ as functions of l and Q. That is, the two equations give the force and voltage needed to cause a certain displacement and a certain charge. The two equations (5) and (6) constitute the equations of state of the transducer. The equations represent the transformations that map the states of the transducer from one thermodynamic plane to another.

Equation (5) can be used to determine the free-energy function from the force-displacement curves of the transducer measured under the open-circuit conditions, when the electrodes maintain constant charges. For each value of Q, the free energy is the area under the force-displacement curve. Similarly, Eq.(6) can be used to determine the free-energy function from the voltage-charge curves of the transducer. As mentioned before, (l, P) and (Q, Φ) are convenient planes to represent the states of the transducer when we wish to highlight work and energy.

As an illustration, consider a parallel-plate capacitor—two plates of electrodes separated by a thin layer of a vacuum (Fig.7). The separation l between the two electrodes may vary, but the area A of either electrode remains fixed. Recall the elementary fact that the amount of charge on either
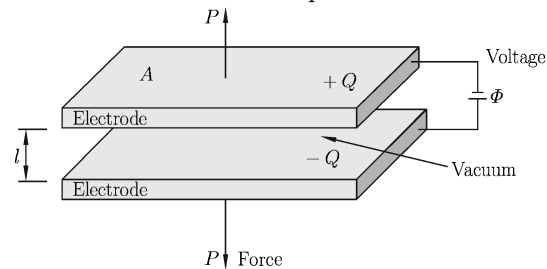

Fig. 7   A parallel-plate capacitor consists of two electrodes separated by a thin gap of a vacuum. When a voltage is applied, the two electrodes attract each other. The electrostatic attraction is balanced by applying a force.

electrode is linear in the voltage:

$$\Phi = \frac{lQ}{\varepsilon_0 A} \tag{7}$$

where ε_0 is the permittivity of the vacuum. Inserting Eq.(7) into Eq.(6), and integrating Eq.(6) with respect to Q while holding l fixed, we obtain that

$$F(l, Q) = \frac{lQ^2}{2\varepsilon_0 A} \tag{8}$$

Inserting Eq.(8) into Eq.(5), we obtain that

$$P = \frac{Q^2}{2A\varepsilon_0} \tag{9}$$

Equations (7) and (9) constitute the equations of state of the parallel-plate capacitor. They are readily interpreted. The applied voltage causes charge to flow from one electrode to the other, so that one electrode is positively charged, and the other negatively charged. Equation (7) relates the charge to the applied voltage. The oppositely charged electrodes attract each other. To maintain equilibrium, a force need be applied to each electrode. Equation (9) relates the applied force to the charge.

Define the electric field by $E = \Phi/l$ and the stress by $\sigma = P/A$. Rewrite Eq.(9) as

$$\sigma = \frac{1}{2}\varepsilon_0 E^2 \tag{10}$$

This equation gives the stress needed to be applied to the electrodes to counteract the electrostatic attraction. This stress is known as the Maxwell stress.

2.5. Stability of a State against Linear Perturbation

For a given transducer, the free energy $F(l, Q)$ may take a complicated functional form. The equations of state, Eqs.(5) and (6), are in general nonlinear. If the transducer operates in the neighborhood of a particular state (l, Q), the equations of state can be linearized in this neighborhood, written in an incremental form:

$$\delta P = \frac{\partial^2 F(l, Q)}{\partial l^2}\delta l + \frac{\partial^2 F(l, Q)}{\partial Q \partial l}\delta Q \tag{11}$$

$$\delta \Phi = \frac{\partial^2 F(l, Q)}{\partial l \partial Q}\delta l + \frac{\partial^2 F(l, Q)}{\partial Q^2}\delta Q \tag{12}$$

The increments of the loads, δP and $\delta \Phi$, are linear in the increments of the kinematic variables, δl and δQ. This procedure is known as linear perturbation. We call $\partial^2 F(l, Q)/\partial l^2$ the mechanical tangent stiffness of the transducer, and $\partial^2 F(l, Q)/\partial Q^2$ the electrical tangent stiffness of the transducer. The two electromechanical coupling effects are both characterized by the same cross derivative, $\partial^2 F(l, Q)/(\partial l \partial Q) = \partial^2 F(l, Q)/(\partial Q \partial l)$. The matrix

$$\boldsymbol{H}(l, Q) = \begin{bmatrix} \dfrac{\partial^2 F(l, Q)}{\partial l^2} & \dfrac{\partial^2 F(l, Q)}{\partial Q \partial l} \\ \dfrac{\partial^2 F(l, Q)}{\partial l \partial Q} & \dfrac{\partial^2 F(l, Q)}{\partial Q^2} \end{bmatrix} \tag{13}$$

is known as the Hessian of the free-energy function $F(l, Q)$.

As mentioned above, a state of the transducer can be represented by a point in the (l, Q) plane, as well as by a point in the (P, Φ) plane. For the same state of the transducer, the point in the (l, Q) plane is mapped to the point in the (P, Φ) plane by the equations of state, (5) and (6). The mapping may not always be invertible. That is, given a pair of the loads (P, Φ), the equations of state may not be invertible to determine a state (l, Q). For example, Eqs.(11) and (12) are not invertible when the Hessian is a singular matrix, $\det \boldsymbol{H} = 0$.

This singularity may be understood in terms of thermodynamics. The transducer and the loading mechanisms (i.e., the weight and the battery) together constitute a thermodynamic system. The free

energy of the system is the sum of the free energies of the individual parts—the transducer, the weight, and the battery. The free energy (i.e., the potential energy) of a constant weight is $-Pl$. The free energy of a battery of a constant voltage is $-\Phi Q$. Consequently, the free energy of the thermodynamic system combining the transducer and the loading mechanisms is

$$G(l, Q) = F(l, Q) - Pl - \Phi Q \tag{14}$$

The system has two independent variables, l and Q.

Thermodynamics requires that the system should reach a stable state of equilibrium when the free-energy function $G(l, Q)$ is a minimum against small changes in l and Q. When the weight moves by δl and the battery pumps charges δQ, the free energy of the system varies by

$$\delta G = \left[\frac{\partial F(l, Q)}{\partial l} - P\right] \delta l + \left[\frac{\partial F(l, Q)}{\partial Q} - \Phi\right] \delta Q + \frac{\partial^2 F(l, Q)}{2\partial l^2} (\delta l)^2$$

$$+ \frac{\partial^2 F(l, Q)}{\partial l \partial Q} (\delta l)(\delta Q) + \frac{\partial^2 F(l, Q)}{2\partial Q^2} (\delta Q)^2 \tag{15}$$

We have expanded the Taylor series of the function $F(l, Q)$ up to terms quadratic in δl and δQ. In a state of equilibrium, the coefficients of the first-order variations vanish, recovering the equations of state (5) and (6). To ensure that this state of equilibrium minimizes G, the sum of the second-order variations must be positive for arbitrary combination of δl and δQ. That is, a state of equilibrium is stable against small perturbation if the Hessian of the free energy of the transducer, $\boldsymbol{H}(l, Q)$, is positive-definite. The two-by-two matrix is positive-definite if and only if

$$\frac{\partial^2 F(l, Q)}{\partial l^2} > 0, \quad \frac{\partial^2 F(l, Q)}{\partial Q^2} > 0, \quad \left[\frac{\partial^2 F(l, Q)}{\partial l^2}\right]\left[\frac{\partial^2 F(l, Q)}{\partial Q^2}\right] > \left[\frac{\partial^2 F(l, Q)}{\partial l \partial Q}\right]^2 \tag{16}$$

When the Hessian of the free energy function is positive-definite, the function $F(l, Q)$ is convex at this state (l, Q).

As an illustration, consider the parallel-plate capacitor again. Given the free-energy function (8), the second derivatives are

$$\frac{\partial^2 F(l, Q)}{\partial l^2} = 0, \quad \frac{\partial^2 F(l, Q)}{\partial Q^2} = \frac{l}{\varepsilon_0 A}, \quad \left[\frac{\partial^2 F(l, Q)}{\partial l \partial Q}\right] = \frac{Q}{\varepsilon_0 A} \tag{17}$$

Consequently, the Hessian is not positive-definite in any state of equilibrium. That is, the parallel-plate capacitor subject to a constant force and a constant voltage cannot reach a stable state of equilibrium. The conclusion is readily understood. The weight is independent of the separation between the plates, but the electrostatic attractive force increases as the separation decreases. Subject to a fixed weight, the two plates will be pulled apart if the voltage is low, and will be pulled together if the voltage is high.

The capacitor can be stabilized by a modification of the loading mechanisms. For example, we can replace the weight with a spring that restrains the relative movement of the plates. Let K be the stiffness of the spring, and l_0 be the separation between the electrodes when the spring is unstretched, so that the force in the spring is $P = K(l_0 - l)$. The free energy of the system is the sum of the free energies of the capacitor, the spring and the battery:

$$G(l, Q) = \frac{lQ^2}{2\varepsilon_0 A} + \frac{1}{2}K(l - l_0)^2 - \Phi Q \tag{18}$$

In a state of equilibrium, the first derivatives of $G(l, Q)$ vanish, giving the same equations of state as Eqs.(10) and (12). The state of equilibrium is stable if and only if the Hessian of $G(l, Q)$ is positive-definite. The second derivatives of the function $G(l, Q)$ are

$$\frac{\partial^2 G(l, Q)}{\partial l^2} = K, \quad \frac{\partial^2 G(l, Q)}{\partial Q^2} = \frac{l}{\varepsilon_0 A}, \quad \frac{\partial^2 G(l, Q)}{\partial l \partial Q} = \frac{Q}{\varepsilon_0 A} \tag{19}$$

A state of equilibrium (l, Q) is stable if and only if

$$\frac{Kl}{\varepsilon_0 A} > \left(\frac{Q}{\varepsilon_0 A}\right)^2 \tag{20}$$

Thus, the transducer is stable when the spring is stiff and the applied voltage is small.

2.6. Nonconvex Free-Energy Surface

Following Gibbs[38], we may interpret above analytical statements geometrically. This geometric representation not only shows the geometric nature of the stability against linear perturbation, but also shows the possibility of coexistent states.

Consider a three-dimensional space with (l, Q) as the horizontal plane, and F as the vertical axis. In this space, the Helmholtz free energy $F(l, Q)$ is represented by a surface. Consider an inclined plane passing through the origin of the space, with P being the slope of the inclined plane with respect to the l axis, and Φ being the slope of the inclined plane with respect to the Q axis. According to Eq.(14), the vertical distance between the surface $F(l, Q)$ and the inclined plane is the function $G(l, Q)$. Thermodynamics dictates that this vertical distance $G(l, Q)$ should minimize when the transducer equilibrates with the loads (P, Φ).

Picture a plane simultaneously parallel to the inclined plane and tangent to the surface $F(l, Q)$. From the geometry, the tangent point minimizes the vertical distance $G(l, Q)$ if the surface $F(l, Q)$ is above the tangent plane—that is, if the surface $F(l, Q)$ is convex at the state (l, Q).

When the loads (P, Φ) change gradually, the inclined plane rotates, and the associated tangent plane rolls along the free-energy surface. If the surface $F(l, Q)$ is globally convex, every tangent plane touches the surface at only one point, and only one state of equilibrium is associated with a pair of given loads (P, Φ). By contrast, if part of the surface $F(l, Q)$ is concave, a tangent plane may touch the surface at two points, and the two states of equilibrium are associated with a pair of given loads (P, Φ).

It was discovered that the free-energy functions for dielectric elastomers are typically nonconvex[39]. Associated with a given set of loads, two states of equilibrium may coexist. This topic will be discussed in §5.4 and §6.8.

III. HOMOGENEOUS FIELD

We now develop a field theory of deformable dielectrics. The field theory assumes that a body is a sum of many small pieces, and the field in each small piece is homogeneous. This assumption enables us to define quantities per unit length, per unit area, and per unit volume. This section focuses on the homogeneous field of a small piece, and §VI considers inhomogeneous field in the body by summing up small pieces.

This section begins by setting up a thermodynamic framework for electromechanical coupling. We then consider several specific material models: a vacuum as an elastic dielectric of vanishing rigidity, incompressible materials, ideal dielectric elastomers, electrostrictive materials, and nonlinear dielectrics.

3.1. Condition of Thermodynamic Equilibrium

With reference to Fig.2, consider a membrane of an elastic dielectric, sandwiched between two compliant electrodes. In the reference state, the dielectric is subject to neither force nor voltage, and the dielectric is of dimensions L_1, L_2 and L_3. In the current state, the dielectric is subject to forces P_1, P_2 and P_3, and the two electrodes are connected to a battery of voltage Φ through a conducting wire. In the current state, the dimensions of the dielectric become l_1, l_2 and l_3, the two electrodes accumulate electric charges $\pm Q$, and the Helmholtz free energy of the membrane is F.

When the dimensions of the dielectric change by δl_1, δl_2 and δl_3, the forces do work $P_1 \delta l_1 + P_2 \delta l_2 + P_3 \delta l_3$. When a small quantity of charge δQ flows through the conducting wire, the voltage does work $\Phi \delta Q$. When the dielectric equilibrates with the forces and the voltage, the increase in the free energy equals the work done:

$$\delta F = P_1 \delta l_1 + P_2 \delta l_2 + P_3 \delta l_3 + \Phi \delta Q \tag{21}$$

The condition of equilibrium (21) holds for arbitrary small variations of the four independent variables, l_1, l_2, l_3 and Q.

3.2. Equations of State in Terms of Nominal Quantities

Define the nominal density of the Helmholtz free energy by $W = F/(L_1 L_2 L_3)$, stretches by $\lambda_1 = l_1/L_1$, $\lambda_2 = l_2/L_2$ and $\lambda_3 = l_3/L_3$, nominal stresses by $s_1 = P_1/(L_2 L_3)$, $s_2 = P_2/(L_1 L_3)$ and $s_3 = P_3/(L_1 L_2)$, nominal electric field by $\tilde{E} = \Phi/L_3$, and nominal electric displacement by $\tilde{D} = Q/(L_1 L_2)$.

The amount of charge on either electrode relates to the nominal electric displacement by $Q = \tilde{D}L_1L_2$. When the membrane is subject to forces and voltage, the variation of the charge is $\delta Q = L_1L_2\delta\tilde{D}$.

Divide both sides of Eq.(21) by $L_1L_2L_3$, the volume of the membrane in the reference state. We obtain that

$$\delta W = s_1\delta\lambda_1 + s_2\delta\lambda_2 + s_3\delta\lambda_3 + \tilde{E}\delta\tilde{D} \tag{22}$$

This condition of equilibrium holds for arbitrary and independent small variations $\delta\lambda_1$, $\delta\lambda_2$, $\delta\lambda_3$ and $\delta\tilde{D}$.

As a material model, the nominal density of the Helmholtz free energy is prescribed as a function of the four independent variables:

$$W = W\left(\lambda_1, \lambda_2, \lambda_3, \tilde{D}\right) \tag{23}$$

Inserting Eq.(23) into Eq.(22), we obtain that

$$\left(\frac{\partial W}{\partial\lambda_1} - s_1\right)\delta\lambda_1 + \left(\frac{\partial W}{\partial\lambda_2} - s_2\right)\delta\lambda_2 + \left(\frac{\partial W}{\partial\lambda_3} - s_3\right)\delta\lambda_3 + \left(\frac{\partial W}{\partial\tilde{D}} - \tilde{E}\right)\delta\tilde{D} = 0 \tag{24}$$

This condition of equilibrium holds for arbitrary and independent small variations $\delta\lambda_1$, $\delta\lambda_2$, $\delta\lambda_3$ and $\delta\tilde{D}$. Consequently, when the dielectric equilibrates with the applied forces and the applied voltage, the coefficient in front of each variation in Eq.(24) vanishes, giving

$$s_1 = \frac{\partial W\left(\lambda_1, \lambda_2, \lambda_3, \tilde{D}\right)}{\partial\lambda_1} \tag{25}$$

$$s_2 = \frac{\partial W\left(\lambda_1, \lambda_2, \lambda_3, \tilde{D}\right)}{\partial\lambda_2} \tag{26}$$

$$s_3 = \frac{\partial W\left(\lambda_1, \lambda_2, \lambda_3, \tilde{D}\right)}{\partial\lambda_3} \tag{27}$$

$$\tilde{E} = \frac{\partial W\left(\lambda_1, \lambda_2, \lambda_3, \tilde{D}\right)}{\partial\tilde{D}} \tag{28}$$

The equations of state (25)-(28) give the values of the forces and voltage needed to equilibrate with the dielectric in the state $\left(\lambda_1, \lambda_2, \lambda_3, \tilde{D}\right)$ once the free-energy function $W\left(\lambda_1, \lambda_2, \lambda_3, \tilde{D}\right)$ is prescribed as a material model.

In the above, we have defined stresses as applied forces divided by areas. In the absence of the applied forces, the stresses in the dielectric vanish. The stresses are zero even when the voltage causes the dielectric to deform. Thus, when the battery applies a voltage to the dielectric, the positive charge on one electrode and the negative charge on the other electrode cause the dielectric to thin down. We simply report what we have observed in this experiment: the voltage causes the dielectric to deform. We do not jump to the conclusion that the voltage causes a compressive stress. In this regard, we view the deformation caused by the voltage in the same way as we view the deformation caused by a change in temperature: both are stress-free deformation, so long as the material is unconstrained[12].

3.3. Equations of State in Terms of True Quantities

In §3.2, we have represented the equations of state in terms of the nominal quantities. We now represent the equations of state in terms of true quantities. The two sets of equations describe the same condition of thermodynamic equilibrium, but they are convenient under different circumstances.

Define the true stresses by $\sigma_1 = P_1/\left(l_2l_3\right)$, $\sigma_2 = P_2/\left(l_1l_3\right)$ and $\sigma_3 = P_3/\left(l_1l_2\right)$, true electric field by $E = \Phi/l_3$, and true electric displacement by $D = Q/(l_1l_2)$. The amount of charge on either electrode relates to the electric displacement by $Q = Dl_1l_2$. When the membrane is subject to forces and voltage, all three quantities D, l_1 and l_2 can vary, so that the variation of the charge is

$$\delta Q = Dl_2\delta l_1 + Dl_1\delta l_2 + l_1l_2\delta D \tag{29}$$

This equation should be contrasted with $\delta Q = L_1 L_2 \delta \tilde{D}$, where the nominal electric displacement is used.

Dividing both sides of Eq.(21) by $L_1 L_2 L_3$, the volume of the membrane in the reference state, and using Eq.(29), we obtain that

$$\delta W = (\sigma_1 + DE)\, \lambda_2 \lambda_3 \delta \lambda_1 + (\sigma_2 + DE)\, \lambda_1 \lambda_3 \delta \lambda_2 + \sigma_3 \lambda_1 \lambda_2 \delta \lambda_3 + \lambda_1 \lambda_2 \lambda_3 E \delta D \tag{30}$$

The condition of equilibrium (30) holds for arbitrary and independent variations $\delta \lambda_1$, $\delta \lambda_2$, $\delta \lambda_3$ and δD.

As a material model, the nominal density of Helmholtz free energy is taken to be a function of the four independent variables,

$$W = W\left(\lambda_1, \lambda_2, \lambda_3, D\right) \tag{31}$$

Comparing Eqs.(23) and (31), we remark that here the true electric displacement, rather than the nominal electric displacement, is used as an independent variable.

Inserting Eq.(31) into Eq.(30), we obtain that

$$\left[\frac{\partial W}{\partial \lambda_1} - (\sigma_1 + DE)\, \lambda_2 \lambda_3 \right] \delta \lambda_1 + \left[\frac{\partial W}{\partial \lambda_2} - (\sigma_2 + DE)\, \lambda_1 \lambda_3 \right] \delta \lambda_2$$

$$+ \left(\frac{\partial W}{\partial \lambda_3} - \sigma_3 \lambda_1 \lambda_2 \right) \delta \lambda_3 + \left(\frac{\partial W}{\partial D} - \lambda_1 \lambda_2 \lambda_3 E \right) \delta D = 0 \tag{32}$$

This condition of equilibrium holds for arbitrary and independent small variations $\delta \lambda_1$, $\delta \lambda_2$, $\delta \lambda_3$ and δD. Consequently, when the dielectric equilibrates with the applied forces and the applied voltage, the coefficient in front of each variation in Eq.(32) vanishes, giving

$$\sigma_1 = \frac{\partial W\left(\lambda_1, \lambda_2, \lambda_3, D\right)}{\lambda_2 \lambda_3 \partial \lambda_1} - ED \tag{33}$$

$$\sigma_2 = \frac{\partial W\left(\lambda_1, \lambda_2, \lambda_3, D\right)}{\lambda_1 \lambda_3 \partial \lambda_2} - ED \tag{34}$$

$$\sigma_3 = \frac{\partial W\left(\lambda_1, \lambda_2, \lambda_3, D\right)}{\lambda_1 \lambda_2 \partial \lambda_3} \tag{35}$$

$$E = \frac{\partial W\left(\lambda_1, \lambda_2, \lambda_3, D\right)}{\lambda_1 \lambda_2 \lambda_3 \partial D} \tag{36}$$

Equations (33)-(36) constitute the equations of state for an elastic dielectric once the function $W(\lambda_1, \lambda_2, \lambda_3, D)$ is given. These equations suggest that electromechanical coupling be classified into two kinds. First, the geometric coupling is characterized by Eq.(29), which results in the term DE in Eqs.(33) and (34). Second, the material coupling is characterized by the function $W\left(\lambda_1, \lambda_2, \lambda_3, D\right)$. Several specific material models are described next.

3.4. Vacuum

We think of a vacuum as an elastic dielectric with vanishing stiffness, undergoing a homogenous deformation λ_1, λ_2, and λ_3. Recall an elementary fact that in the vacuum the electric field relates to the electric displacement as $E = D/\varepsilon_0$. Integrating Eq.(30) with respect to D, while holding λ_1, λ_2 and λ_3 fixed, we obtain that

$$W\left(\lambda_1, \lambda_2, \lambda_3, D\right) = \frac{D^2}{2\varepsilon_0} \lambda_1 \lambda_2 \lambda_3 \tag{37}$$

This expression recovers a familiar result in electrostatics: $D^2 / (2\varepsilon_0)$ is the electrostatic energy per unit volume in the vacuum. The factor $\lambda_1 \lambda_2 \lambda_3$ appears in Eq.(37) because we have defined W as the nominal density of energy.

Inserting Eq.(37) into Eqs.(33)-(35), we obtain that

$$\sigma_1 = -\frac{1}{2}\varepsilon_0 E^2 \tag{38}$$

$$\sigma_2 = -\frac{1}{2}\varepsilon_0 E^2 \tag{39}$$

$$\sigma_3 = \frac{1}{2}\varepsilon_0 E^2 \tag{40}$$

Equations (38)-(40) recover the stresses obtained by Maxwell[40]. They are valid in the vacuum when the electric field is in direction 3.

The Maxwell stress is a tensor. In §2.4, we have interpreted the component of the Maxwell stress in the direction of the electric field. We now look at the two components of the Maxwell stress transverse to the direction of the electric field. Figure 8 illustrates a classic experiment of a capacitor, which is partly in the air and partly in a dielectric liquid. The applied voltage causes the liquid to rise to a height h. The height results from the balance of the Maxwell stress and the weight of the liquid. The Maxwell stress parallel to the electrodes in the air is $\sigma_a = -\varepsilon_a E^2/2$, where ε_a is the permittivity of the air. The Maxwell stress parallel to the electrodes in the liquid is $\sigma_l = -\varepsilon_l E^2/2$, where ε_l is the permittivity of the liquid. The electric field near the air/liquid interface is distorted, so that the above two formulas are correct only at some distance away from the interface. Because $\varepsilon_l > \varepsilon_a$, the difference in the Maxwell stresses in the two media will draw the liquid up against gravity. Examining the free-body diagram, and balancing the electrostatic forces with the weight of the liquid, we obtain that $\rho gh = (\varepsilon_l - \varepsilon_a) E^2/2$, where ρg is the weight per unit volume of the liquid.
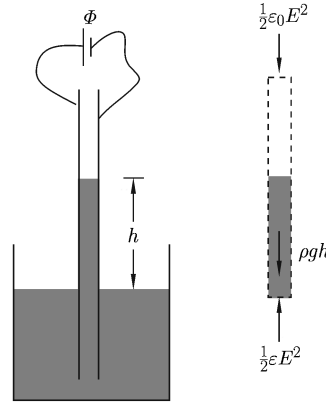

Fig. 8 A parallel-plate capacitor is partly in the air and partly in a dielectric liquid. When a voltage is applied, the liquid rises. As indicated by the free-body diagram on the right, the rise of the liquid is due to the balance between the Maxwell stress and the weight of the liquid.

3.5. Incompressibility

When an elastomer undergoes large deformation, the change in the shape of the elastomer is typically much larger than the change in the volume. Consequently, the elastomer is often taken to be incompressible—that is, the volume of the material remains unchanged during deformation, $l_1 l_2 l_3 = L_1 L_2 L_3$, so that

$$\lambda_1 \lambda_2 \lambda_3 = 1 \tag{41}$$

This assumption of incompressibility places a constraint among the three stretches. We regard λ_1 and λ_2 as independent variables, rewrite Eq.(41) as $\lambda_3 = 1/(\lambda_1\lambda_2)$, and express $\delta\lambda_3$ in terms of $\delta\lambda_1$ and $\delta\lambda_2$:

$$\delta\lambda_3 = -\frac{\delta\lambda_1}{\lambda_1^2\lambda_2} - \frac{\delta\lambda_2}{\lambda_1\lambda_2^2} \tag{42}$$

In terms of the variations of the independent variables, the condition of equilibrium (30) becomes

$$\delta W = \frac{\sigma_1 - \sigma_3 + DE}{\lambda_1}\delta\lambda_1 + \frac{\sigma_2 - \sigma_3 + DE}{\lambda_2}\delta\lambda_2 + E\delta D \tag{43}$$

For an incompressible dielectric, the condition of equilibrium (43) holds for arbitrary and independent variations $\delta\lambda_1$, $\delta\lambda_2$ and δD. For an incompressible elastic dielectric, the density of the free energy is a function of the three independent variables: $W = W(\lambda_1, \lambda_2, D)$. Equation (43) becomes that

$$\left(\frac{\partial W}{\partial\lambda_1} - \frac{\sigma_1 - \sigma_3 + DE}{\lambda_1}\right)\delta\lambda_1 + \left(\frac{\partial W}{\partial\lambda_2} - \frac{\sigma_2 - \sigma_3 + DE}{\lambda_2}\right)\delta\lambda_2 + \left(\frac{\partial W}{\partial D} - E\right)\delta D = 0 \tag{44}$$

Because $\delta\lambda_1$, $\delta\lambda_2$ and δD are independent variations, the condition of equilibrium (44) is equivalent to three equations:

$$\sigma_1 - \sigma_3 = \lambda_1 \frac{\partial W\left(\lambda_1, \lambda_2, D\right)}{\partial \lambda_1} - ED \tag{45}$$

$$\sigma_2 - \sigma_3 = \lambda_2 \frac{\partial W\left(\lambda_1, \lambda_2, D\right)}{\partial \lambda_2} - ED \tag{46}$$

$$E = \frac{\partial W\left(\lambda_1, \lambda_2, D\right)}{\partial D} \tag{47}$$

Once the function $W\left(\lambda_1, \lambda_2, D\right)$ is given for an incompressible dielectric elastomer, the four equations, (41) and (45)-(47), constitute the equations of state.

3.6. Ideal Dielectric Elastomers

An elastomer is a three-dimensional network of long and flexible polymers, held together by crosslinks (Fig.9). Each polymer chain consists of a large number of monomers. Consequently, the crosslinks have negligible effect on the polarization of the monomers—that is, the elastomer can polarize nearly as freely as a polymer melt. This molecular picture is consistent with the following experimental observation: the permittivity changes by only a few percent when a membrane of an elastomer is stretched to increase the area 25 times[41].
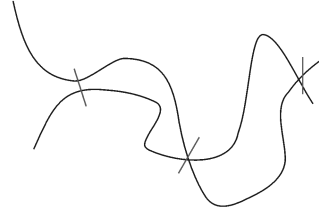

Fig. 9   An elastomer is a three dimensional network of long and flexible polymer chains. Each polymer chain consists of a large number of monomers.

As an idealization, we may assume that the dielectric behavior of an elastomer is exactly the same as that of a polymer melt—that is, the true electric field relates to the true electric displacement as

$$E = D/\varepsilon \tag{48}$$

where ε is the permittivity of the elastomer, taken to be a constant independent of deformation. Inserting Eq.(48) into Eq.(43), and integrating Eq.(43) with respect to D while holding λ_1 and λ_2 fixed, we obtain that

$$W\left(\lambda_1, \lambda_2, D\right) = W_s\left(\lambda_1, \lambda_2\right) + \frac{D^2}{2\varepsilon} \tag{49}$$

The constant of integration, $W_s\left(\lambda_1, \lambda_2\right)$, is the Helmholtz free energy associated with the stretching of the elastomer. The material is also taken to be incompressible, $\lambda_1\lambda_2\lambda_3 = 1$. This material model (49) is known as the model of ideal dielectric elastomers[39]. In this model (49), the stretches and the polarization contribute to the free energy independently. Consequently, the electromechanical coupling in an ideal dielectric elastomer is purely a geometric effect, in the sense as remarked at the end of §3.3.

Inserting Eq.(49) into Eqs.(45) and (46), and also using Eq.(48), we obtain that

$$\sigma_1 - \sigma_3 = \lambda_1 \frac{\partial W_s\left(\lambda_1, \lambda_2\right)}{\partial \lambda_1} - \varepsilon E^2 \tag{50}$$

$$\sigma_2 - \sigma_3 = \lambda_2 \frac{\partial W_s\left(\lambda_1, \lambda_2\right)}{\partial \lambda_2} - \varepsilon E^2 \tag{51}$$

Equations (41), (48), (50) and (51) constitute the equations of state for an incompressible, ideal dielectric elastomer, provided the permittivity ε and the function $W_s\left(\lambda_1, \lambda_2\right)$ are given. These equations of state have been used almost exclusively in all analyses of dielectric elastomers, and agree well with experimentally measured equations of state[42]. The equations are usually justified in terms of the Maxwell stress[3], and can be interpreted using the model of ideal dielectric elastomers[39]. That is, the Maxwell stress is valid when the dielectric behavior of the material is liquid-like, unaffected by deformation.

As shown in Eqs.(50) and (51), a through-thickness voltage induces a compressive stress of magnitude εE^2 in the two in-plane directions. This magnitude is twice the magnitude of the Maxwell stress. The apparent difference is readily understood (Fig.10). Because the elastomer is taken to be incompressible, superposition of a state of hydrostatic stress does not affect the state of deformation. Start from the state of triaxial stresses $\left(-\varepsilon E^2/2,\ -\varepsilon E^2/2,\ +\varepsilon E^2/2\right)$, as derived by Maxwell. A superposition of a state of hydrostatic stress $\left(+\varepsilon E^2/2,\ +\varepsilon E^2/2,\ +\varepsilon E^2/2\right)$ gives a state of uniaxial stress $\left(0,\ 0,\ +\varepsilon E^2\right)$. A superposition of a state of hydrostatic stress $\left(-\varepsilon E^2/2,\ -\varepsilon E^2/2,\ -\varepsilon E^2/2\right)$ gives a state of biaxial stress $\left(-\varepsilon E^2,\ -\varepsilon E^2,\ 0\right)$. For an incompressible material, the three states of stress illustrated in Fig.10 cause the same state of deformation.
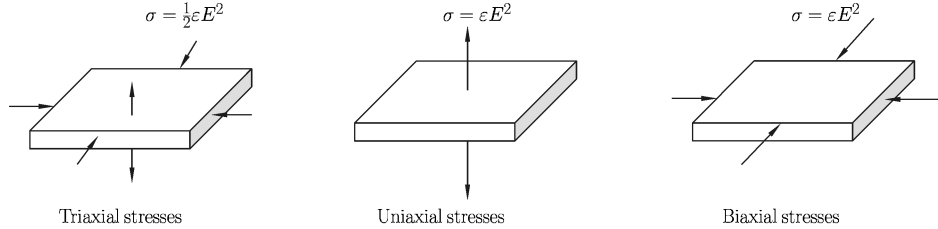

Fig. 10. A dielectric in three states of stresses.

The free energy due to the stretching of the elastomer, $W_s\left(\lambda_1, \lambda_2\right)$, may be selected from a large menu of well-tested functions in the theory of rubber elasticity. For example, the neo-Hookean model takes the form

$$W_s = \frac{\mu}{2}\left(\lambda_1^2 + \lambda_2^2 + \lambda_3^2 - 3\right) \tag{52}$$

where μ is the small-stress shear modulus. This free energy is due to the change of entropy when polymer chains are stretched[43].

In an elastomer, each individual polymer chain has a finite contour length. When the elastomer is subject to no loads, the polymer chains are coiled, allowing a large number of conformations. Subject to loads, the polymer chains become less coiled. As the loads increase, the end-to-end distance of each polymer chain approaches the finite contour length, and the elastomer approaches a limiting stretch. On approaching the limiting stretch, the elastomer stiffens steeply. This effect is absent in the neo-Hookean model, but is represented by the Arruda and Boyce model[44] and the Gent model[45]. The latter takes the form

$$W_s = -\frac{\mu J_{\lim}}{2}\log\left(1 - \frac{\lambda_1^2 + \lambda_2^2 + \lambda_3^2 - 3}{J_{\lim}}\right) \tag{53}$$

where μ is the small-stress shear modulus, and $J_{\lim}$ is a constant related to the limiting stretch. The stretches are restricted as $0 \leq \left(\lambda_1^2 + \lambda_2^2 + \lambda_3^2 - 3\right)/J_{\lim} < 1$. When $\left(\lambda_1^2 + \lambda_2^2 + \lambda_3^2 - 3\right)/J_{\lim} \to 0$, the Taylor expansion of Eq.(53) is Eq.(52). That is, the Gent model recovers the neo-Hookean model when deformation is small compared to the limiting stretch. When $\left(\lambda_1^2 + \lambda_2^2 + \lambda_3^2 - 3\right)/J_{\lim} \to 1$, the free energy diverges, and the elastomer approaches the limiting stretch.

3.7. Electrostriction

Subject to a voltage through the thickness, some dielectrics become thinner, but other dielectrics become thicker (Fig.11). For dielectrics that are nonpolar in the absence of electric field, the voltage-induced deformation has been analyzed by invoking stresses of two origins: electrostriction and the Maxwell stress. The electrostriction results from the effect of deformation on permittivity.

As a simplest model of electrostriction, we still assume that the true electric field is linear in the true electric displacement, but now the permittivity is a function of the stretches. Write the relation between the electric field and the electric displacement as

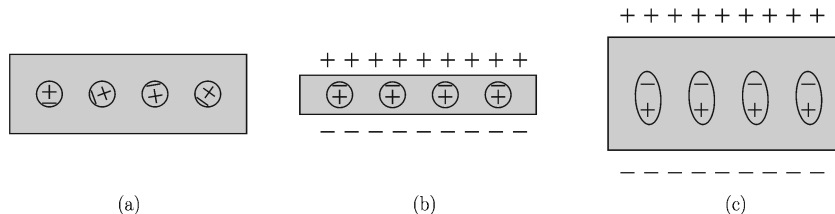$$E = \frac{D}{\varepsilon\left(\lambda_1, \lambda_2\right)} \tag{54}$$

Fig. 11. Consider a dielectric that is nonpolar in the absence of applied voltage (a). Subject to a voltage, some dielectrics become thinner (b), but other dielectrics become thicker (c).

The same procedure as that in §3.6 gives[46]

$$\sigma_1 - \sigma_3 = \lambda_1 \frac{\partial W_s\left(\lambda_1, \lambda_2\right)}{\partial \lambda_1} - \left[\varepsilon\left(\lambda_1, \lambda_2\right) + \frac{\lambda_1}{2}\frac{\partial \varepsilon\left(\lambda_1, \lambda_2\right)}{\partial \lambda_1}\right] E^2 \tag{55}$$

$$\sigma_2 - \sigma_3 = \lambda_2 \frac{\partial W_s\left(\lambda_1, \lambda_2\right)}{\partial \lambda_2} - \left[\varepsilon\left(\lambda_1, \lambda_2\right) + \frac{\lambda_2}{2}\frac{\partial \varepsilon\left(\lambda_1, \lambda_2\right)}{\partial \lambda_2}\right] E^2 \tag{56}$$

Equations (41) and (54)-(56) constitute the equations of state for an incompressible, electrostrictive dielectric elastomer.

When the permittivity varies significantly with the stretches, the terms $\partial \varepsilon\left(\lambda_1, \lambda_2\right)/\partial \lambda_1$ and $\partial \varepsilon(\lambda_1, \lambda_2)/\partial \lambda_2$ in Eqs.(55) and (56) make sizable contributions. In particular, when the permittivity decreases substantially as the stretches increase, the stresses associated with the electric field may change the sign. It is well known that, for many dielectrics other than elastomers, a voltage applied through the thickness causes the dielectrics to increase the thickness, as shown in Fig.11(c).

The variation of the permittivity with stretches has been observed experimentally for a dielectric elastomer[47]. Further measurements are needed to ascertain the practical significance of electrostriction in dielectric elastomers.

3.8. Nonlinear Dielectrics

In the models described so far, the electric field is taken to be linear in the electric displacement. We now consider a nonlinear dielectric. In an elastomer, each polymer chain may consist of monomers of electric dipoles. In the absence of the applied voltage, the dipoles undergo thermal fluctuation, and are randomly oriented. The situation is similar to water molecules. When the elastomer is subject to a voltage, the dipoles rotate toward the direction of the electric field. When the voltage becomes sufficiently high, the dipoles become perfectly aligned with the electric field, and the polarization of the material saturate. This nonlinear dielectric behavior may be included by generalizing the model of ideal dielectric elastomers[48].

As an idealization, we may assume that the dielectric behavior of an elastomer is exactly the same as that of a polymer melt, so that the electric field relates to the electric displacement by a function independent of deformation:

$$E = f\left(D\right) \tag{57}$$

The function $f\left(D\right)$ can be nonlinear. Following the same procedure as that in §3.6, we obtain that

$$\sigma_1 - \sigma_3 = \lambda_1 \frac{\partial W_s\left(\lambda_1, \lambda_2\right)}{\partial \lambda_1} - ED \tag{58}$$

$$\sigma_2 - \sigma_3 = \lambda_2 \frac{\partial W_s\left(\lambda_1, \lambda_2\right)}{\partial \lambda_2} - ED \tag{59}$$

Equations (41) and (57)-(59) constitute the equations of state for an incompressible, ideal dielectric elastomer, accounting for nonlinear dielectric behavior.

To study the effect of polarization saturation, here we assume that the elastomer is characterized a nonlinear D-E relation, $D = D_s \tanh\left(\varepsilon E/D_s\right)$, where ε and D_s are constants[48]. When electric field is low, $\varepsilon E/D_s << 1$, the D-E relation recovers the linear dielectric behavior, $E = D/\varepsilon$, so that ε is

the small-field permittivity. When the electric field is high, $\varepsilon E/D_s >> 1$, the D-E relation becomes $D = D_s$, so that D_s is the saturated electric displacement.

The effect of polarization saturation is appreciated by inspecting the equations of state, (58) and (59). When the dielectric behavior is linear, $D = \varepsilon E$, the term DE recovers the Maxwell stress εE^2. As polarization saturates, however, the term DE becomes $D_s E$, which increases with the electric field linearly. Consequently, polarization saturation makes the stress associated with voltage rise less steeply. This behavior may markedly affect electromechanical coupling[48].

IV. NONEQUILIBRIUM THERMODYNAMICS OF DIELECTRIC ELASTOMERS

An elastomer responds to forces and voltage by time-dependent, dissipative processes[49–51]. Viscoelastic relaxation may result from slippage between long polymers and rotation of joints between monomers. Dielectric relaxation may result from distortion of electron clouds and rotation of polar groups. Conductive relaxation may result from migration of electrons and ions through the elastomer. This section describes an approach to construct models of dissipative dielectric elastomers, guided by nonequilibrium thermodynamics[52].

Thermodynamics requires that the increase in the free energy should not exceed the total work done, namely,

$$\delta F \leq P_1 \delta l_1 + P_2 \delta l_2 + P_3 \delta l_3 + \Phi \delta Q \tag{60}$$

For the inequality to be meaningful, the small changes are time-directed: δf means the change of the quantity f from one time to a slightly later time.

Divide both sides of Eq.(60) by the volume of the membrane, $L_1 L_2 L_3$, and the thermodynamic inequality becomes

$$\delta W \leq s_1 \delta \lambda_1 + s_2 \delta \lambda_2 + s_3 \delta \lambda_3 + \tilde{E} \delta \tilde{D} \tag{61}$$

As a model of the dielectric elastomer, the free-energy density is prescribed as a function:

$$W = W \left(\lambda_1, \lambda_2, \lambda_3, \tilde{D}, \xi_1, \xi_2, ... \right) \tag{62}$$

We characterize the state of a dielectric by λ_1, λ_2, λ_3 and $\tilde{D}$, along with additional parameters $(\xi_1, \xi_2, ...)$. Inspecting Eq.(61), we note that λ_1, λ_2, λ_3 and $\tilde{D}$ are the kinematic parameters through which the external loads do work. By contrast, the additional parameters $(\xi_1, \xi_2, ...)$ are not associated with the external loads in this way. These additional parameters describe the degrees of freedom associated with dissipative processes, and are known as internal variables.

Inserting Eq.(62) into Eq.(61), we rewrite the thermodynamic inequality as

$$\left(\frac{\partial W}{\partial \lambda_1} - s_1 \right) \delta \lambda_1 + \left(\frac{\partial W}{\partial \lambda_2} - s_2 \right) \delta \lambda_2 + \left(\frac{\partial W}{\partial \lambda_3} - s_3 \right) \delta \lambda_3 + \left(\frac{\partial W}{\partial \tilde{D}} - \tilde{E} \right) \delta \tilde{D} + \sum_i \frac{\partial W}{\partial \xi_i} \delta \xi_i \leq 0 \tag{63}$$

As time goes forward, this thermodynamic inequality holds for any change in the independent variables $\left(\lambda_1, \lambda_2, \lambda_3, \tilde{D}, \xi_1, \xi_2, ... \right)$. We next specify a model consistent with this inequality.

We assume that the system is in mechanical and electrostatic equilibrium, so that in Eq.(63) the factors in front of $\delta \lambda_1$, $\delta \lambda_2$, $\delta \lambda_3$ and $\delta \tilde{D}$ vanish:

$$s_1 = \frac{\partial W \left(\lambda_1, \lambda_2, \lambda_3, \tilde{D}, \xi_1, \xi_2, ... \right)}{\partial \lambda_1} \tag{64}$$

$$s_2 = \frac{\partial W \left(\lambda_1, \lambda_2, \lambda_3, \tilde{D}, \xi_1, \xi_2, ... \right)}{\partial \lambda_2} \tag{65}$$

$$s_3 = \frac{\partial W \left(\lambda_1, \lambda_2, \lambda_3, \tilde{D}, \xi_1, \xi_2, ... \right)}{\partial \lambda_3} \tag{66}$$

$$\tilde{E} = \frac{\partial W \left(\lambda_1, \lambda_2, \lambda_3, \tilde{D}, \xi_1, \xi_2, ... \right)}{\partial \tilde{D}} \tag{67}$$

Equations (64)-(67) constitute the thermodynamic equations of state of the dielectric elastomer.

Once the elastomer is assumed to be in mechanical and electrostatic equilibrium, the inequality (63) becomes

$$\sum_i \frac{\partial W\left(\lambda_1, \lambda_2, \lambda_3, \tilde{D}, \xi_1, \xi_2, ...\right)}{\partial \xi_i} \delta \xi_i \leq 0 \tag{68}$$

This thermodynamic inequality may be satisfied by prescribing a suitable relation between $(\delta\xi_1, \delta\xi_2, ...)$ and $(\partial W/\partial\xi_1, \partial W/\partial\xi_2, ...)$. For example, one may adopt a kinetic model of the type

$$\frac{\mathrm{d}\xi_i}{\mathrm{d}t} = -\sum_j M_{ij} \frac{\partial W\left(\lambda_1, \lambda_2, \lambda_3, \tilde{D}, \xi_1, \xi_2, ...\right)}{\partial \xi_j} \tag{69}$$

Here M_{ij} is a positive-definite matrix, which may depend on the independent variables $(\lambda_1, \lambda_2, \lambda_3, \tilde{D}, \xi_1, \xi_2, ...)$.

To represent a dissipative dielectric elastomer using the above approach, we need to specify a set of internal variables $(\xi_1, \xi_2, ...)$, and then specify the functions $W\left(\lambda_1, \lambda_2, \lambda_3, \tilde{D}, \xi_1, \xi_2, ...\right)$ and $M_{ij}\left(\lambda_1, \lambda_2, \lambda_3, \tilde{D}, \xi_1, \xi_2, ...\right)$. There is considerable flexibility in choosing kinetic models to fulfill the thermodynamic inequality (68). To develop a kinetic model for a given material, one also draws upon mechanistic pictures and experimental data.

Viscoelastic relaxation is commonly pictured with an array of springs and dashpots, known as the rheological models; see recent examples[52, 53]. Similarly, dielectric relaxation is commonly pictured with models consisting of resistors and capacitors. By contrast, electrical conduction involves the transport of charged species over a long distance. Coupled large deformation and transport of charged species are significant in polyelectrolytes[21], and will not be discussed here.

V. ELECTROMECHANICAL INSTABILITY

While all dielectrics deform under voltage, the amount of deformation differs markedly among different materials. Under voltage, piezoelectric ceramics attain strains of typically less than 1%. Glassy and semi-crystalline polymers can attain strains of less than 10%[54]. Strains about 30% were observed in some elastomers[55]. In the last decade, strains over 100% have been achieved in several ways, by pre-stretching an elastomer[3], by using an elastomer of interpenetrating networks[56, 57], by swelling an elastomer with a solvent[58], and by spraying charge on an electrode-free elastomer[59].

These experimental advances have prompted a theoretical question: What is the fundamental limit of deformation that can be induced by voltage? After all, one can easily increase the length of a rubber band several times by using a mechanical force. Why is it difficult to do so by using a voltage?

5.1. Electrical Breakdown and Electromechanical Instability

The difficulty to achieve large deformation by voltage has to do with two modes of failure: electrical breakdown and electromechanical instability. For a stiff dielectric such as a ceramic or a glassy polymer, voltage-induced deformation is limited by electrical breakdown, when the voltage mobilizes charged species in the dielectric to produce a path of electrical conduction. For a compliant dielectric such as an elastomer, the voltage-induced deformation is often limited by electromechanical instability.

Stark and Garton[60] described a model that accounted for the following experimental observation: the breakdown fields of a polymer reduces when the polymer becomes soft at elevated temperatures. As the applied voltage increases, the polymer thins down, so that the same voltage induces an even higher electric field. This positive feedback results in a mode of instability, known as electromechanical instability or pull-in instability, which causes the polymer to reduce the thickness drastically, often leading to electrical breakdown. Electromechanical instability has been recognized as a mode of failure for insulators in power transmission cables.

5.2. Desirable Stress-Stretch Behavior for Large Voltage-Induced Deformation

Electromechanical instability is sensitive to the stress-stretch behavior of the elastomer[39]. Figure 12(a) sketches a dielectric membrane pulled by biaxial stresses σ. The length of the membrane in any direction in the plane is stretched by a ratio λ. As will become clear, to attain a large voltage-induced stretch, the dielectric should have a stress-stretch curve $\sigma(\lambda)$ of the following desirable features[61]: (a) The dielectric is compliant at small stretches, and (b) the dielectric stiffens steeply at modest stretches. That is, the limiting stretch, λ_{lim}, should not be excessive.
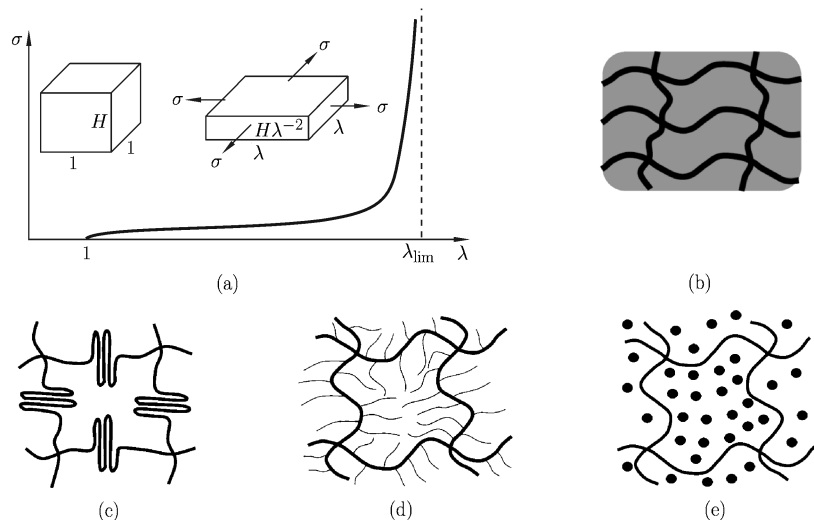

Fig. 12. Several molecular structures can lead to a stress-stretch curve of a desirable form [61]. (a) Stress-stretch curve of a membrane under biaxial stresses. (b) Fibers embedded in a compliant matrix. (c) A network of polymers with folded domains. (d) A network of polymers with side chains. (e) A network of polymers swollen with a solvent.

Also sketched are several designs of materials that exhibit the stress-stretch curve of the desirable form. Many biological tissues, such as skins and vascular walls, deform readily, but avert excessive deformation. Figure 12(b) sketches a design of such a tissue, consisting of stiffer fibers in a compliant matrix. At small stretches, the fibers are loose, and the tissue is compliant. At large stretches, the fibers are taut, and the tissue stiffens steeply. As another example, Fig.12(c) sketches a network of polymers with folded domains. The domains unfold when the network is pulled, giving rise to substantial deformation. After all the domains unfold, the network stiffens steeply.

Consider an elastomer, i.e., a network of polymer chains. When the individual chains are short, the initial modulus of the elastomer is large and the limiting stretch λ_{lim} is small. When the individual chains are long, the initial modulus of the elastomer is small and the limiting stretch λ_{lim} is large. Consequently, it is difficult to achieve the stress-stretch curve of the desirable form by adjusting the density of crosslinks alone. The stress-stretch curve, however, can be shaped into the desirable form in several ways. For example, the widely used dielectric elastomer, VHB, is a network of polymers with side chains (Fig. 12(d)). The side chains fill the space around the networked chains. The motion of the networked chains is lubricated, lowering the glass transition temperature. Also the density of the networked chains is reduced, lowering the stiffness of the elastomer when the stretch is small. While the side chains do not change the contour length of the networked chains, the side chains pull the networked chains towards their full contour length even when the elastomer is not loaded. Once loaded, the elastomer may stiffen sharply, averting electromechanical instability. Similar behavior is expected for a network swollen with a solvent (Fig.12(e)). The stress-stretch curve can also be shaped into the desirable form by prestretch[3], or by using interpenetrating networks[56,57].

5.3. Voltage-Stretch Curve Goes Up, Down, and Up Again

We now use the stress-stretch curve $\sigma(\lambda)$ to deduce the voltage-stretch curve $\Phi(\lambda)$. As illustrated in Fig.13(a), when a membrane of an elastomer, thickness H in the undeformed state, is subject to a
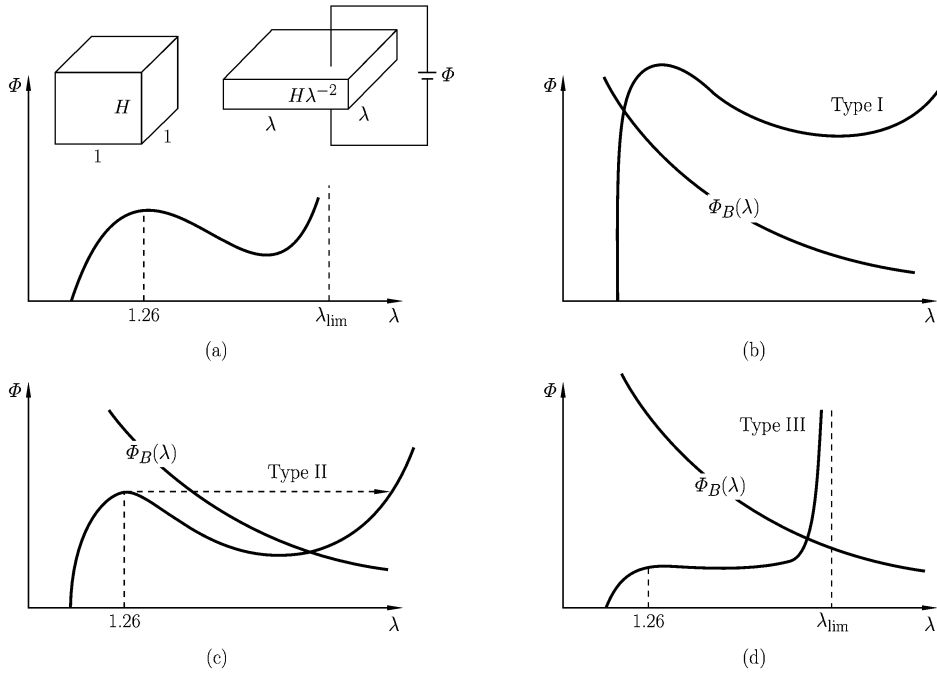
Fig. 13. Three types of behavior of a dielectric transducer[61]. (a) A membrane of a dielectric elastomer subject to a voltage reduces thickness and expands area. The voltage-stretch curve is typically not monotonic, (b)-(d) Three types behavior are distinguished, depending on where the two curves $\Phi(\lambda)$ and $\Phi_B(\lambda)$ intersect.

voltage Φ, the membrane is stretched by λ in both directions in the plane, the thickness of the membrane reduces to $H\lambda^{-2}$, and the electric field in the membrane is $E = \lambda^2\Phi/H$. The membrane is taken to be incompressible. The voltage-induced stretch can be described by using the Maxwell stress (50), namely, $\sigma(\lambda) - \varepsilon E^2 = 0$.

A combination of the above considerations relates the voltage to the stretch:

$$\Phi = H\lambda^{-2}\sqrt{\sigma(\lambda)/\varepsilon} \tag{70}$$

This voltage-stretch relation is sketched in Fig.13(a). Even though the stress-stretch curve $\sigma(\lambda)$ is monotonic, the voltage-stretch curve $\Phi(\lambda)$ is usually not[39]. At a small stretch ($\lambda \sim 1$), the rising $\sigma(\lambda)$ dominates, and the voltage increases with the stretch. At an intermediate stretch, the factor λ^{-2} due to thinning of the membrane becomes important, and the voltage falls as the stretch increases. As the elastomer approaches the limiting stretch λ_{lim}, the steep rise of $\sigma(\lambda)$ prevails, and the voltage rises again. The shape of the voltage-stretch curve $\Phi(\lambda)$ indicates a snap-through electromechanical instability[39].

The local maximum voltage represents a critical condition, which can be estimated as follows. Under the equal-biaxial stresses, Hooke's law takes the form $\sigma(\lambda) = 6\mu(\lambda - 1)$, where μ is the shear modulus. Inserting this expression into Eq.(70), and maximizing the function $\Phi(\lambda)$, we find local maximum voltage $\Phi_c \approx 0.80H\sqrt{\mu/\varepsilon}$ and the critical $\lambda_c = 4/3 = 1.33$. The critical values vary somewhat with the stress-stretch relation. For example, for the neo-Hookean model, $\sigma(\lambda) = \mu(\lambda^2 - \lambda^{-4})$, the maximum voltage is $\Phi_c \approx 0.69H\sqrt{\mu/\varepsilon}$ and the critical stretch is $\lambda_c = 2^{1/3} \approx 1.26$. This electromechanical instability has been analyzed systematically by using the Hessian[62–66]. Stability analysis has also been carried out by considering inhomogeneous perturbation[67,68].

The stretch also affects the voltage for electrical breakdown, Φ_B. The function $\Phi_B(\lambda)$ may be determined experimentally as follows. Before a voltage is applied, an elastomer is prestretched to λ by a mechanical force, and is then fixed by rigid electrodes. Subsequently, when the voltage is applied, the elastomer will not deform further. The measured voltage at failure is taken to be the voltage for electrical breakdown, Φ_B. Experiments indicate that the breakdown voltage is a monotonically decreasing function of the prestretch[30,41]. This trend may be understood as follows: the larger the prestretch, the thinner the membrane, and the higher the electric field for the same applied voltage.

According to where the curves $\Phi(\lambda)$ and $\Phi_B(\lambda)$ intersect, we distinguish three types of transducers[61]. A type I transducer suffers electrical breakdown prior to electromechanical instability, and is capable of small voltage-induced deformation, Fig.13(b). A type II transducer reaches the peak of the $\Phi(\lambda)$ curve, and thins down excessively, leading to electrical breakdown, Fig.13(c). The transducer is recorded to fail at the peak of $\Phi(\lambda)$, which can be much below the breakdown voltage Φ_B. The voltage-induced deformation is limited by the stretch at which the voltage reaches the peak. A type III transducer eliminates or survives electromechanical instability, reaches a stable state before the electrical breakdown, and attains a large voltage-induced deformation, Fig.13(d). This classification accounts for existing experimental observations, and suggests alternative routes to achieve giant voltage-induced deformation[61].

5.4. Coexistent States

A new experimental manifestation of the electromechanical instability has been reported recently[30]. Under certain conditions, an applied voltage can deform a layer of a dielectric elastomer into a mixture of two regions, one being flat and the other wrinkled (Fig.14). This observation has been interpreted as the coexistence of two states, resulting from the shape of voltage-stretch curve (Fig.13(a))[39]. In one state, the membrane is thick and has a small area. In the other state, the membrane is thin and has a large area. The two states may coexist at a specific applied voltage, so that some regions of the membrane are in the thick state, while other regions are in the thin state. The regions of the thick state constrain the regions of the thin state. To accommodate the large area in the thin state, the regions of the thin state form wrinkles.

Fig. 14. An experimental observation of electromechanical instability (courtesy of Plante J. S. and Dubowsky S.). A layer of a dielectric elastomer, coated with conductive grease on top and bottom faces, is pre-stretched using a frame. An electric voltage is applied between the two electrodes. The layer deforms into a mixture of two regions, one being flat and the other wrinkled.

VI. INHOMOGENEOUS FIELDS

Following §III, we now sum up small pieces to describe a body of inhomogeneous deformation. No new physical ingredient is needed, but the reader is assumed of more sophisticated background in mathematics. Studies of inhomogeneous fields of coupled large deformation and electric potential date back to classic works of Toupin[69], Eringen[70] and Tiersten[71]. These works have been reexamined recently for applications to dielectric elastomers[9–13]. This section begins by expressing the condition of thermodynamic equilibrium in terms of a variational statement. The same condition of thermodynamic equilibrium is also expressed in terms of partial differential equations. We formulate the model of ideal dielectric elastomers, and describe a finite element method for a dielectric membrane of any arbitrary shape. We then perturb a state of static equilibrium, a technique commonly used to analyze oscillation

and bifurcation. We conclude with a discussion of coexistent phases.

6.1. Variational Statement of Thermodynamic Equilibrium

A body of an elastic dielectric is represented by a sum of many small pieces, called material particles. Each material particle is named after the coordinate $\boldsymbol{X}$ of its place when the body is in a reference state. In the current state, at time t, the particle $\boldsymbol{X}$ moves to a place with coordinate $\boldsymbol{x}$. The function

$$\boldsymbol{x} = \boldsymbol{x}\left(\boldsymbol{X}, t\right) \tag{71}$$

describes the history of the deformation of the body. Define the deformation gradient $\boldsymbol{F}$ as

$$F_{iK} = \frac{\partial x_i\left(\boldsymbol{X}, t\right)}{\partial X_K} \tag{72}$$

The deformation gradient generalizes the notion of the stretches.

In the current state at time t, the electric potential at particle $\boldsymbol{X}$ is denoted as

$$\varPhi = \varPhi\left(\boldsymbol{X}, t\right) \tag{73}$$

The gradient of the electric potential defines the nominal electric field $\tilde{\boldsymbol{E}}$, namely,

$$\tilde{E}_K = -\frac{\partial \varPhi\left(\boldsymbol{X}, t\right)}{\partial X_K} \tag{74}$$

The negative sign in Eq.(74) follows the convention that the electric field points from a material particle of a high voltage to a material particle of a low voltage.

Motivated by Eq.(22), we write the variation of the nominal density of the Helmholtz free energy, δW, in the form

$$\delta W = s_{iK}\delta F_{iK} + \tilde{E}_K \delta\tilde{D}_K \tag{75}$$

where δF_{iK} is a small change in the deformation gradient, and $\delta\tilde{D}_K$ is a small change in the nominal electric displacement. Equation (75) defines the nominal stress $\boldsymbol{s}$ as a tensor work-conjugate to the deformation gradient $\boldsymbol{F}$, and the nominal electric displacement $\tilde{\boldsymbol{D}}$ as a vector work-conjugate to the nominal electric field $\tilde{\boldsymbol{E}}$.

Inspecting Eqs.(72) and (74), we wish to use the deformation gradient and the nominal electric field as the independent variables. Introducing a new quantity $\hat{W}$ by

$$\hat{W} = W - \tilde{E}_K \tilde{D}_K \tag{76}$$

The quantity $\hat{W}$ may be called the electrical Gibbs free energy. A combination of Eqs.(75) and (76) gives

$$\delta\hat{W} = s_{iK}\delta F_{iK} - \tilde{D}_K \delta\tilde{E}_K \tag{77}$$

We may call the quantity $\tilde{D}_K \delta\tilde{E}_K$ the complementary electrical work.

A material model is prescribed by a function $\hat{W} = \hat{W}\left(\boldsymbol{F}, \tilde{\boldsymbol{E}}\right)$. When the body undergoes a rigid-body motion, the free energy is invariant. Consequently, the function depends on the deformation gradient $\boldsymbol{F}$ through the Green deformation tensor, $C_{KL} = F_{iK}F_{iL}$. Associated with small changes δF_{iK} and $\delta\tilde{E}_K$, the electrical Gibbs free energy changes by

$$\delta\hat{W} = \frac{\partial\hat{W}\left(\boldsymbol{F}, \tilde{\boldsymbol{E}}\right)}{\partial F_{iK}}\delta F_{iK} + \frac{\partial\hat{W}\left(\boldsymbol{F}, \tilde{\boldsymbol{E}}\right)}{\partial\tilde{E}_K}\delta\tilde{E}_K \tag{78}$$

On each material element of volume $\mathrm{d}V\left(\boldsymbol{X}\right)$, we prescribe mass $\rho\left(\boldsymbol{X}\right)\mathrm{d}V$, force $\boldsymbol{B}\left(\boldsymbol{X}, t\right)\mathrm{d}V$ and charge $q\left(\boldsymbol{X}, t\right)\mathrm{d}V$. The effect of inertia may be represented by adding to the force the inertial force, so that the combination of the applied force and the inertial force on the element of volume is $\left(\boldsymbol{B} - \rho\partial^2\boldsymbol{x}/\partial t^2\right)\mathrm{d}V$. The body may consist of dissimilar dielectrics and conductors, separated by interfaces. Consider an interface separating two parts of the body labeled as $-$ and $+$. When the body is in the reference state, denote an element of the interface by $\mathrm{d}A\left(\boldsymbol{X}\right)$, and denote the unit vector normal to the element of the

interface by $\boldsymbol{N}$, pointing toward part $+$. On the element of the interface, we prescribe force $\boldsymbol{T}(\boldsymbol{X}, t)\,\mathrm{d}A$ and charge $\omega(\boldsymbol{X}, t)\,\mathrm{d}A$.

Let $\delta x_i = \xi_i(\boldsymbol{X})$ be a field of virtual displacement of the body. Associated with the field of virtual displacement, the forces do virtual work $\int (B_i - \rho \partial^2 x_i/\partial t^2)\,\delta x_i \mathrm{d}V + \int T_i \delta x_i \mathrm{d}A$. Similarly, let $\delta \Phi = \eta(\boldsymbol{X})$ be a field of virtual electric potential of the body. Associated with the field of electric potential, the charges do virtual complementary work $\int q\delta\Phi \mathrm{d}V + \int \omega\delta\Phi \mathrm{d}A$. The virtual deformation gradient is $\delta F_{iK} = \partial \xi_i(\boldsymbol{X})/\partial X_K$, and the virtual nominal electric field is $\delta \tilde{E}_K = -\partial \eta(\boldsymbol{X})/\partial X_K$. The virtual change in the electrical Gibbs free energy is $\int \delta \hat{W} \mathrm{d}V$, where $\delta \hat{W}$ is given by Eq.(78).

When the body is in thermodynamic equilibrium, the change in the electrical Gibbs free energy equals the mechanical work minus the complementary electrical work:

$$\int \delta \hat{W} \mathrm{d}V = \int \left(B_i - \rho \frac{\partial^2 x_i}{\partial t^2} \right) \delta x_i \mathrm{d}V + \int T_i \delta x_i \mathrm{d}A - \int q\delta\Phi \mathrm{d}V - \int \omega\delta\Phi \mathrm{d}A \qquad (79)$$

This condition of thermodynamic equilibrium has the similar physical content as Eqs.(1) and (21), and holds for arbitrary and independent variations $\delta\boldsymbol{x}$ and $\delta\Phi$. The above presentation follows closely a previous paper[12]. Variational statements of various forms may be developed from alternative starting points[72–75].

Once the loads and the electrical Gibbs free-energy function $\hat{W}\left(\boldsymbol{F}, \tilde{\boldsymbol{E}}\right)$ are prescribed, the variational statement (79), along with the definitions (72) and (74), is the basis for the finite element method, determining the field of deformation $\boldsymbol{x}(\boldsymbol{X}, t)$ and the field of electric potential $\Phi(\boldsymbol{X}, t)$ simultaneously. Several implementations of the finite element method have been reported[75–78], but few practical examples are available. Significant effort is needed to develop the finite element method, and to apply the method to analyze phenomena and devices.

6.2. Differential Equations

A comparison of Eqs.(77) and (78) gives that

$$s_{iK} = \frac{\partial \hat{W}\left(\boldsymbol{F}, \tilde{\boldsymbol{E}}\right)}{\partial F_{iK}} \qquad (80)$$

$$\tilde{D}_K = -\frac{\partial \hat{W}\left(\boldsymbol{F}, \tilde{\boldsymbol{E}}\right)}{\partial \tilde{E}_K} \qquad (81)$$

Once the electrical free-energy function $\hat{W}\left(\boldsymbol{F}, \tilde{\boldsymbol{E}}\right)$ is prescribed, Eqs.(80) and (81) constitute the equations of state.

Inserting Eqs.(72), (74) and (77) into the condition of thermodynamic equilibrium (79), and recalling that the condition holds for arbitrary and independent variations in $\delta\boldsymbol{x}$ and $\delta\Phi$, we obtain that

$$\frac{\partial s_{iK}(\boldsymbol{X}, t)}{\partial X_K} + B_i(\boldsymbol{X}, t) = \rho(\boldsymbol{X}) \frac{\partial^2 x_i(\boldsymbol{X}, t)}{\partial t^2} \qquad (82)$$

in the volume

$$\left(s_{iK}^- - s_{iK}^+ \right) N_K = T_i \qquad (83)$$

on the interfaces

$$\frac{\partial \tilde{D}_K(\boldsymbol{X}, t)}{\partial X_K} = q(\boldsymbol{X}, t) \qquad (84)$$

in the volume, and

$$\left(\tilde{D}_K^+ - \tilde{D}_K^- \right) N_K = \omega(\boldsymbol{X}, t) \qquad (85)$$

on the interfaces. Equations (82) and (83) reproduce the equations for momentum balance, and Eqs.(84) and (85) reproduce Gauss's law of electrostatics.

Equations (71)-(74) and (80)-(85) are governing equations to determine the field of deformation $\boldsymbol{x}(\boldsymbol{X}, t)$ and the field of electric potential $\Phi(\boldsymbol{X}, t)$ simultaneously, once the loads and the free-energy

function $\hat{W}\left(\boldsymbol{F}, \tilde{\boldsymbol{E}}\right)$ are prescribed. These partial differential equations have been used to solve boundary-value problems[79–86]. Observe that the equations of mechanics, (71), (72), (82) and (83), decouple from those of electrostatics, (73), (74), (84) and (85). The only coupling between mechanics and electrostatics arises from the material model, (80) and (81).

6.3. True Quantities

The true stress σ_{ij} relates to the nominal stress by

$$\sigma_{ij} = \frac{F_{jK}}{\det \boldsymbol{F}} s_{iK} \tag{86}$$

The true electric displacement D_i relates to the nominal electric displacement as

$$D_i = \frac{F_{iK}}{\det \boldsymbol{F}} \tilde{D}_K \tag{87}$$

The true electric field E_i relates to the nominal electric field as

$$E_i = H_{iK} \tilde{E}_K \tag{88}$$

where H_{iK} is the inverse of the deformation gradient, namely, $H_{iK} F_{iL} = \delta_{KL}$ and $H_{iK} F_{jK} = \delta_{ij}$. The true quantities may be taken as functions of $\boldsymbol{x}$ and t, and satisfy the familiar partial differential equations in mechanics and electrostatics.

6.4. Ideal Dielectric Elastomers

For an isotropic elastic dielectric, the free-energy density is a function of six invariants of the deformation gradient tensor and the electric field vector[69]. Function of this complexity is unavailable for any real material. We next describe ideal dielectric elastomers—a material model nearly exclusively used in the literature. As discussed before in connection with Fig.9, for an ideal dielectric elastomer, the dielectric behavior is the same as that of a liquid—that is, the dielectric behavior is unaffected by deformation[39].

As a simplest model of a dielectric liquid, assume that the true electric displacement D_m is linear in the true electric field E_m:

$$D_m = \varepsilon E_m \tag{89}$$

The permittivity ε is taken to be independent of deformation.

Using Eqs.(87) and (88), we express Eq.(89) in terms of the nominal fields:

$$\tilde{D}_N = \varepsilon \tilde{E}_L H_{mN} H_{mL} \det \boldsymbol{F} \tag{90}$$

Inserting Eq.(90) into Eq.(81) and integrating with respect to $\tilde{\boldsymbol{E}}$ while holding $\boldsymbol{F}$ fixed, we obtain the nominal density of the electrical Gibbs free energy:

$$\hat{W}\left(\boldsymbol{F}, \tilde{\boldsymbol{E}}\right) = W_s\left(\boldsymbol{F}\right) - \frac{\varepsilon}{2} \tilde{E}_N \tilde{E}_L H_{mN} H_{mL} \det \boldsymbol{F} \tag{91}$$

The constant of integration, $W_s\left(\boldsymbol{F}\right)$, is the free energy associated with the elasticity of the elastomer, which may be selected from a large menu in the theory of elasticity. While the elastomer is nearly incompressible, in the finite element method, it is convenient to allow the material to be compressible with a large bulk modulus.

Insert Eq.(91) into Eq.(80), and recall mathematical identities $\partial H_{mN}/\partial F_{iK} = -H_{mK} H_{iN}$ and $\partial \det \boldsymbol{F}/\partial F_{iK} = H_{iK} \det \boldsymbol{F}$. We obtain that

$$s_{iK} = \frac{\partial W_s\left(\boldsymbol{F}\right)}{\partial F_{iK}} + \varepsilon \tilde{E}_N \tilde{E}_L \left(H_{iN} H_{mK} H_{mL} - \frac{1}{2} H_{mN} H_{mL} H_{iK} \right) \det \boldsymbol{F} \tag{92}$$

This equation of state relates the nominal stress to the deformation gradient and the nominal electric field.

A combination of Eqs.(86) and (92) gives

$$\sigma_{ij} = \frac{F_{jK}}{\det \boldsymbol{F}} \frac{\partial W_s\left(\boldsymbol{F}\right)}{\partial F_{iK}} + \varepsilon \left(E_i E_j - \frac{1}{2} E_m E_m \delta_{ij} \right) \tag{93}$$

This equation relates the true stress to the deformation gradient and the true electric field. The contribution due to the deformation gradient results from the stretching of the elastomer, and is the same as that in the theory of elasticity. The contribution due to the electric field is identical to that derived by Maxwell[40]. When $\sigma_{ij} = 0$, Eq.(93) balances elasticity and electrostatics, and determines the voltage-induced deformation. As commented before, the Maxwell stress correctly accounts for the voltage-induced deformation only when the dielectric behavior is liquid-like, an idealization that works well with elastomers, but not for other solid dielectrics.

Equations (89) and (93) constitute the equations of state, for an ideal dielectric elastomer, in terms of the true quantities. The equations of state exhibit one-way coupling: the deformation does not affect the electrical behavior, but the electric field contributes to the stress-stretch relation. As noted before, the partial differential equations of mechanics decouple from those of electrostatics. One may solve electrostatic boundary-value problems in terms of the true fields, and then add the Maxwell stress in solving the elastic field. Of course, the deformation will change the shape of the boundary of the body. This change must be included in solving the electrostatic problems. The one-way coupling may not bring any advantage after all.

The model of ideal dielectric elastomers can be generalized to account for nonlinear dielectric behavior, by replacing Eq.(89) with a nonlinear relation between the electric field and electric displacement[12,48]. Furthermore, the model of ideal dielectric elastomers can be modified to include dissipative processes, such as viscoelasticity, dielectric relaxation, and electrical conduction[21,52].

6.5. Weak Statements

We recall a well known statement in continuum mechanics. On each part of the boundary of a body, either the displacement or the traction is prescribed. Let $\xi_i\left(\boldsymbol{X}\right)$ be a function defined in the body and on the boundary. Set $\xi_i\left(\boldsymbol{X}\right) = 0$ on the part of the boundary where the displacement is prescribed, but leave the function $\xi_i\left(\boldsymbol{X}\right)$ otherwise arbitrary. Equations (82) and (83) are equivalent to requiring that

$$\int s_{iK} \frac{\partial \xi_i}{\partial X_K} \mathrm{d}V = \int \left(B_i - \rho \frac{\partial^2 x_i}{\partial t^2} \right) \xi_i \mathrm{d}V + \int T_i \xi_i \mathrm{d}A \tag{94}$$

hold for any arbitrary function $\xi_i\left(\boldsymbol{X}\right)$ as described above. The volumetric integrals extend over the volume of the body, and the surface integral extends over the boundary where the traction is prescribed. The statement (94) is readily confirmed by applying the divergence theorem.

Also recall a weak statement concerning electrostatics. On each part of the boundary, either the electric potential or the charge is prescribed. Let $\eta\left(\boldsymbol{X}\right)$ be a function defined in the body and on the boundary. Set $\eta\left(\boldsymbol{X}\right) = 0$ on the part of the boundary where the electric potential is prescribed, and leave $\eta\left(\boldsymbol{X}\right)$ otherwise arbitrary. Equations (84) and (85) are equivalent to requiring that

$$\int \left(-\frac{\partial \eta}{\partial X_K} \right) \tilde{D}_K \mathrm{d}V = \int \eta q \mathrm{d}V + \int \eta \omega \mathrm{d}A \tag{95}$$

hold for any arbitrary functions $\eta\left(\boldsymbol{X}\right)$ as specified above. The volumetric integrals extend over the volume, and the surface integral extends over the boundary where charge is prescribed.

6.6. Finite Element Method for an Elastic Dielectric Membrane

To induce a significant deformation, the voltage needs to be on the order $\Phi \sim H\sqrt{\mu/\varepsilon}$, where H is the thickness over which the voltage is applied, μ the shear modulus, and ε the permittivity. A representative order of magnitude is $\sqrt{\mu/\varepsilon} \sim 10^8$ V/m, which is a very high electric field. Consequently, to reduce the voltage needed for actuation, nearly all dielectric elastomer transducers are in the form of thin membranes, with the thickness much smaller than the in-plane dimensions of the membranes.

Membranes of circular, spherical and axisymmetric symmetries have been analyzed[9,30,47,80–85]. Here we briefly review a finite element method for dielectric membranes of arbitrary shapes, coupled with passive materials of arbitrary shapes[77].

Consider a dielectric membrane sandwiched between two electrodes. The nominal electric field in the membrane takes the form

$$\tilde{E} = \frac{\Phi}{H(X)} N(X) \tag{96}$$

where Φ is the voltage applied between the electrodes, $H(X)$ is the thickness of the membrane, and $N(X)$ is the unit vector normal to the membrane. Both the thickness and the unit normal vector are measured when the membrane is in the reference state, where the thickness may vary from place to place, and the shape of the membrane can be curved. When the voltage Φ is prescribed, Eq.(96) gives the nominal electric field $\tilde{E}$.

For any material specified by a function $\hat{W}\left(F, \tilde{E}\right)$, the nominal stress relates to the deformation gradient and the nominal electric field by $s_{iK} = \partial\hat{W}\left(F, \tilde{E}\right)/\partial F_{iK}$. This equation, along with Eq.(96), relates the nominal stress to the deformation gradient, with Φ as a known parameter. The weak statement (94) defines a boundary-value problem in elasticity. This boundary-value problem is readily implemented in the commercial finite element package ABAQUS by using a user-defined subroutine[77]. ABAQUS readily accounts for passive materials of arbitrary shapes.

6.7. Linear Perturbation around a State of Static Equilibrium

We now return to the general theory in §6.1 and §6.2. Subject to a set of time-independent loads, a body may attain a state of static equilibrium. We may perturb this state with fields small in strain and electric field[12,67,68]. Such a linear perturbation analysis is commonly used to analyze oscillation and bifurcation[87].

Perturb the state of static equilibrium with a small-amplitude oscillation of frequency Ω. Write the time-dependent fields of deformation and electric potential as

$$x(X,t) = x^\circ(X) + x'(X)\exp(i\Omega t) \tag{97}$$

$$\Phi(X,t) = \Phi^\circ(X) + \Phi'(X)\exp(i\Omega t) \tag{98}$$

where $x^\circ(X)$ and $\Phi^\circ(X)$ are the state of static equilibrium, and $x'(X)$ and $\Phi'(X)$ are the amplitude fields of the oscillation. Associated with the static state of equilibrium, the deformation gradient is $F_{iK}^\circ = \partial x_i^\circ(X)/\partial X_K$, and the nominal electric field is $\tilde{E}_K^\circ = -\partial\Phi^\circ(X)/\partial X_K$. Associated with the oscillation, the amplitudes of the deformation gradient and the nominal electric field are

$$F'_{iK} = \frac{\partial x'_i(X)}{\partial X_K} \tag{99}$$

$$\tilde{E}'_K = -\frac{\partial\Phi'(X)}{\partial X_K} \tag{100}$$

The amplitudes of the oscillation are small, so that the amplitudes of the nominal stress and the nominal electric displacement can be obtained by linear perturbation:

$$s'_{iK} = K_{iKjL}F'_{jL} - e_{iKL}\tilde{E}'_L \tag{101}$$

$$\tilde{D}'_L = e_{iKL}F'_{iK} + \varepsilon_{LK}\tilde{E}'_K \tag{102}$$

The tangent moduli are calculated from

$$K_{iKjL} = \frac{\partial^2\hat{W}}{\partial F_{jL}\partial F_{iK}}, \quad \varepsilon_{KL} = -\frac{\partial^2\hat{W}}{\partial\tilde{E}_K\partial\tilde{E}_L}, \quad e_{iKL} = -\frac{\partial^2\hat{W}}{\partial F_{iK}\partial\tilde{E}_L} \tag{103}$$

The derivatives are calculated at the static field $\left(F^\circ, \tilde{E}^\circ\right)$. So long as the oscillation around a static field is concerned, all dielectrics, including vacuum, act like a linear piezoelectric. Because the static state of equilibrium can be inhomogeneous, the tangent moduli may vary from one material particle to another.

The amplitude fields satisfy the partial differential equations and boundary conditions:

$$\frac{\partial s'_{iK}}{\partial X_K} = -\rho \Omega^2 x'_i \tag{104}$$

$$\frac{\partial \tilde{D}'_K}{\partial X_K} = 0 \tag{105}$$

$$\left(s'^-_{iK} - s'^+_{iK} \right) N_K = 0 \tag{106}$$

$$\left(\tilde{D}'^+_K - \tilde{D}'^-_K \right) N_K = 0 \tag{107}$$

The governing equations for the oscillation constitute an eigenvalues problem, with the frequency as the eigenvalue.

Applying the weak statements (94) and (95) to the amplitude fields, we obtain that

$$\int s'_{iK} F'_{iK} \mathrm{d}V = \Omega^2 \int \rho x'_i x'_i \mathrm{d}V \tag{108}$$

$$\int \tilde{E}'_K \tilde{D}'_K \mathrm{d}V = 0 \tag{109}$$

Adding these two equations, and using Eqs.(101) and (102), we obtain that

$$\int \left(K_{iKjL} F'_{iK} F'_{jL} + \varepsilon_{KL} \tilde{E}'_K \tilde{E}'_L \right) \mathrm{d}V = \Omega^2 \int \rho x'_i x'_i \mathrm{d}V \tag{110}$$

When the tangent moduli K_{iKjL} and ε_{KL} are positive-definite, the frequency Ω is real-valued.

Several oscillatory dielectric elastomer actuators have been analyzed[82, 83]. The above analysis also applies when the frequency is set to be zero. Equations (97) and (98) then pose a bifurcation problem: in the neighborhood of a given state of static equilibrium we look for other states of static equilibrium. This bifurcation problem has been discussed briefly in connection with the electromechanical instability in §V. More detailed discussions are available elsewhere[67, 68].

6.8. Coexistent Phases

As noted in §V, coexistent phases have been observed experimentally for dielectric elastomers. Such coexistence has been analyzed by matching regions of two homogeneous states[39, 88], and the transitional region has been analyzed by using finite element method[76]. Here we consider coexistent phases in general terms. The displacement and electric potential are continuous across the phase boundary, but the deformation gradient, stress, electric field and electric displacement can be discontinuous across the phase boundary. We next derive the conditions of equilibrium following an established method[89–91].

The elastomer, the applied forces and the applied charge together constitute a thermodynamic system. The free energy of the system includes contributions from all the parts:

$$G = \int \hat{W} \mathrm{d}V - \int B_i x_i \mathrm{d}V - \int T_i x_i \mathrm{d}A + \int \Phi q \mathrm{d}V + \int \Phi \omega \mathrm{d}A \tag{111}$$

The free energy varies when the displacement and electric potential vary, and when the phase boundary moves. In equilibrium, the variation of the free energy associated with any such change must vanish.

§6.1 has considered the variation of the free energy associated with the variations of deformation and electric potential, while holding the phase boundary immobile. Setting this variation of free energy to zero, Eq.(79), we obtain field equations in §6.2. In particular, consider an element of the phase boundary $\mathrm{d}A(\boldsymbol{X})$, with $\boldsymbol{N}(\boldsymbol{X})$ being the unit vector normal to the element. Equation (83) requires that the traction be continuous across the phase boundary:

$$\langle s_{iK} \rangle N_K = 0 \tag{112}$$

The notation $\langle f \rangle$ stands for the jump of f across the phase boundary. Similarly, Eq.(85) requires that the component of the electric displacement normal to the phase boundary be continuous across the phase boundary:

$$\left\langle \tilde{D}_K \right\rangle N_K = 0 \tag{113}$$

We next consider the change in the free energy associated with the motion of the phase boundary. The motion of the phase boundary is tracked in the reference state. When the phase boundary moves by a displacement $\delta\alpha\left(\boldsymbol{X}\right)$, the element of the phase boundary at material particle $\boldsymbol{X}$ moves to a new material particle $\boldsymbol{X} + \boldsymbol{N}\left(\boldsymbol{X}\right)\delta\alpha\left(\boldsymbol{X}\right)$. Associated with the movement of the phase boundary, the free energy varies by

$$\delta G = \int \left\langle \hat{W} + \left(\tilde{D}_K \tilde{E}_L - s_{iK} F_{iL}\right) N_K N_L \right\rangle \delta\alpha \mathrm{d}A \tag{114}$$

The integral extends over the phase boundary. Equation (114) corrects a mistake in Eq.(4.22) in my previous paper[91].

When the two phases are in equilibrium, δG vanishes for arbitrary small movement of the phase boundary $\delta\alpha\left(\boldsymbol{X}\right)$, so that the jump in front of $\delta\alpha\left(\boldsymbol{X}\right)$ in Eq.(114) vanishes:

$$\left\langle \hat{W} + \left(\tilde{D}_K \tilde{E}_L - s_{iK} F_{iL}\right) N_K N_L \right\rangle = 0 \tag{115}$$

One can readily confirm that Eq.(115) recovers the condition for coexistence for a special case[39].

VII. CONCLUDING REMARKS

A large number of examples in biology demonstrate that deformation of soft materials connect many stimuli to many functions essential to life. Using soft active materials to create soft machines has emerged as an exciting field of engineering. To participate in advancing the field of soft active materials and soft machines effectively, mechanicians must retool our laboratories and our software, as well as adapt our theories. While theories are being developed for diverse soft active materials, this review focuses on one class of soft active materials: dielectric elastomers. This focus allows us to review the theory of dielectric elastomers in some depth, within the framework of nonlinear continuum mechanics and nonequilibrium thermodynamics, while motivating the theory by empirical observations, molecular pictures and applications. It is hoped that the theory will be used to develop software, study intriguing phenomena of electromechanical coupling, and aid the design of electromechanical transducers. It is also hoped that this review of dielectric elastomers will contribute to the rapid development of theories for diverse soft active materials and soft machines.

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