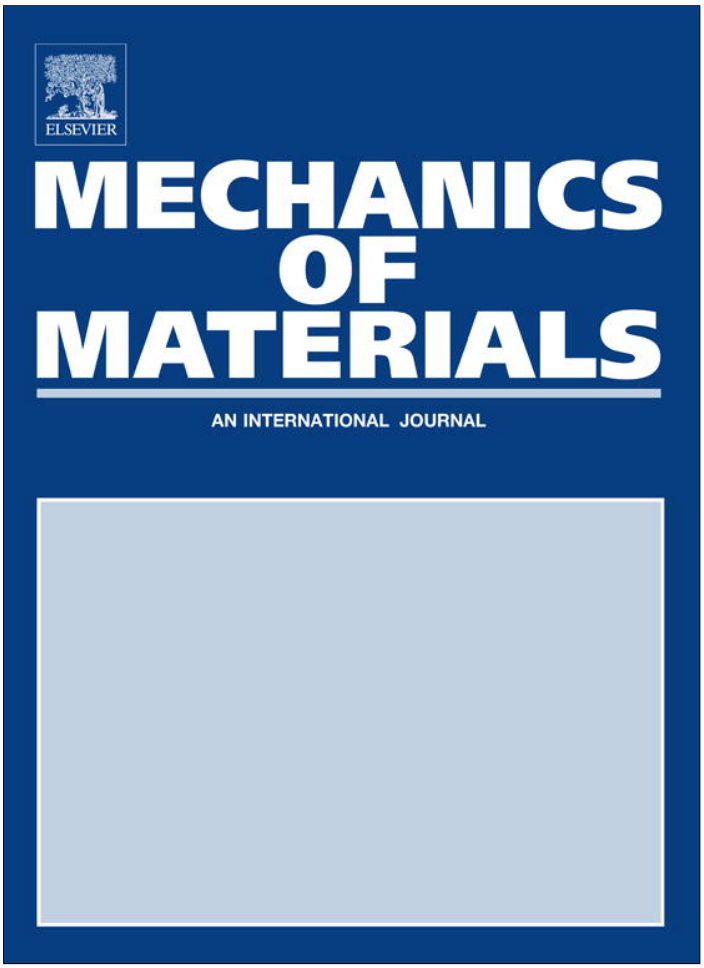

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Electromechanical instability and snap-through instability of dielectric elastomers undergoing polarization saturation

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ABSTRACT

By applying a voltage, electric charge will be induced on the surface of dielectric elastomers. Generally, the charge increases with the level of voltage. When the voltage reaches to a certain value, the charge would not increase any more due to the polarization saturation of dielectric materials. In this paper, a thermodynamic constitutive model, combined both the nonlinear dielectric and hyperelastic behavior as dielectric elastomers undergoing polarization saturation, has been developed. Analytical solutions have been obtained for situations incorporating strain-stiffening effect, electromechanical instability and snap-through instability. The numerical results reveal the marked influence of the extension and polarization saturation limits of elastomer material on its electromechanical instability and the snap-through instability. The developed constitutive model would be helpful in future research of dielectric elastomer based high-performance transducers.

1. Introduction

As one kind of soft electroactive polymers which contain molecular chains cross-linking to each other into 3D network, dielectric elastomers have extraordinary attractive advantages such as large deformation, high elastic energy density, high efficiency, and high responsive speed (Pelrine et al., 2000). When subject to a voltage, dielectric elastomers shrink in thickness and expand in area (Pelrine et al., 2000; Brochu and Pei, 2010; O'Halloran et al., 2008). Based on this electrically activated mechanism of operation, dielectric elastomers have tremendous potential in applications in robotics, aerospace, medical, and bionics engineering. Some practical devices including kinds of actuators, sensors, energy harvesters, tactile displays, and manipulators have already be designed and fabricated (Pelrine et al., 2000; Brochu and Pei, 2010; O'Halloran et al., 2008; Suo, 2010).

For typical dielectric elastomer based transducers, their performances are significantly affected by the coupling effects of both mechanical forces and electric fields applied on them (Brochu and Pei, 2010; Plante and Dubowsky, 2006). Hence, in order to obtain a better performance of dielectric elastomers in transducers, it is necessary to have a thorough understanding of the mechanisms of large deformation, as well as the electromechanical behavior of these materials.

Recently, a lot of research work focused on either large deformation or electromechanical stability has been carried out (Suo, 2010; Zhou et al., 2008; Plante and Dubowsky, 2006; Liu et al., 2010a–c, 2009a–c, 2011; Zhao and Suo, 2008b; Xu et al., 2010; Zhao et al., 2011; Zhu et al., 2010b; Bertoldi and Gei, 2011; He et al., 2010; Adrian Koh et al., 2011a; Huang and Suo, 2012; Li et al., 2011a,b, Kong et al., 2011). Zhao and Suo proposed a theoretical method to investigate the electromechanical instability of

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ideal dielectric elastomers. Their preliminary research results indicated that the simulation predictions of critical electric field coincide with those from experimental observations and they proved theoretically that the pre-stretch cannot only improve the critical nominal electric field but also enhances the electromechanical stability (Zhao and Suo, 2007a). In the statement of this theory, other researchers started to investigate the electromechanical stability of various types of dielectric elastomer combining the strain energy function for hyperelastic materials such as Ogden model, Neo-Hookean model, Arruda–Boyce model, and Mooney–Rivlin model. From these researches, the relations among the nominal stress, the pre-stretch, the nominal electric displacement and the nominal electric field were described accurately (Liu et al., 2008, 2009a–c; Díaz-Calleja et al., 2008; He et al., 2009; Zhao and Suo, 2008a; Suo and Zhu, 2009; Norris, 2008). Meantime, some research results indicated the stable or unstable domain of different types of dielectric elastomer (Liu et al., 2009c; Díaz-Calleja and Díaz-Calleja et al., 2008). All these investigations contribute to the understanding the electromechanical behaviors of dielectric elastomers thoroughly.

For the actuators made of dielectric elastomers, such as balloon, spring-roll and tube, similar theoretical work has been done by several researchers. The dynamic performances of such actuators undergoing finite deformation have been analyzed. (Zhu et al., 2011, 2010a,c; Moscardo et al., 2008). Koh et al. studied an energy harvester based on Neo-Hookean type of dielectric elastomer and evaluated the maximum energy generated by the harvester in one working cycle (Adrian Koh et al., 2009).

For a dielectric elastomer, the dielectric energy per unit volume can be expressed as $D^2/2\varepsilon$, while the permittivity ε can be either a constant (Zhao and Suo, 2007a), linearly dependent of deformation as (He et al., 2009), or nonlinearly dependent of deformation in form of (Leng et al., 2009). Sometimes, to improve the dielectric properties of dielectric elastomer, particles with higher dielectric permittivity will be filled into the elastomer. In this case, the permittivity is not only dependent of deformation, but also dependent of the weight percentage of fillers (Liu et al., 2010b). A recent experimental research discloses the fact that the dielectric permittivity of dielectric elastomer can also be influenced by temperature or frequency (Jean-Mistral et al., 2010). Koh et al. discussed on the failure due to electric breakdown in the snap-through. They concluded that the dielectric elastomers actuation may respond to voltage in two ways: one that undergoes electro-mechanical instability, characterized by a peak in the voltage–stretch response, and the other with a monotonic voltage–stretch response (Adrian Koh et al., 2011b).

In the above-mentioned theoretical study of dielectric elastomers, linear electrostatic free energy has been employed to characterize the polarization, actuation, and instability. This linearity is applicable previously because the conventional dielectric elastomer is susceptible to electromechanical instability, a constant voltage squeezing the elastomer, resulting in a higher electric field and hence ceaselessly thinning the film until failure, and the polarization at electromechanical instability still resides within the linear regime.

In this paper, a free energy function which consists of nonlinear elastic strain energy and nonlinear electric field energy is proposed to construct the constitutive equation and predicate the electromechanical instability and the snap-through instability of dielectric elastomer undergoing polarization saturation. We show that the electromechanical stability and snap-through instability is markedly affected by both the extension limit of polymer chains and the polarization saturation of dipoles.

2. Free energy of dielectric elastomer

A schematic representation of dielectric elastomer is shown in Fig. 1. Generally, due to the fact that dielectric elastomer need to work under a certain electric field, a pair of compliant electrodes made of carbon grease (silver nanowires or carbon nanotubes) are attached on both surfaces of a thin dielectric elastomer film. When the dielectric elastomer is subjected to an electric field, charge flows through a conducting wire from one electrode to the other. The charges of opposite sign on the two electrodes cause the membrane thickness to reduce and the area to be enlarged. Both the mechanical stiffness and electrical resistance can be neglected.

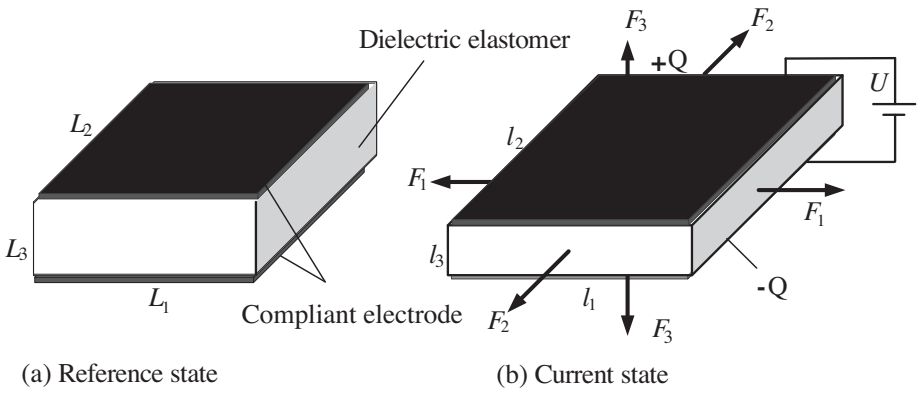

Fig. 1. A membrane of a dielectric elastomer is sandwiched between two compliant electrodes. (a) In the reference state, the dielectric is subject to neither forces nor voltage. (b) In the current state, subject to forces and voltage, the membrane deforms, and charge flows from one electrode to the other through the external conducting wire.

Then we apply the basic theory of thermodynamic system, continuum mechanics, and variation principle to build the equilibrium equation of dielectric elastomers.

As shown is Fig. 1, in the reference state, the elastomer has an initial dimensions of L_1, L_2, and L_3 and is subject to zero force and voltage. While in the current state, with the applied forces F_1, F_2, and F_3, and voltage U, the elastomer's dimensions deforms to l_1, l_2, and l_3 and the accumulated charges generated on the electrodes are $+Q$, $-Q$, respectively.

As the dimensions of the elastomer change by δl_1, δl_2, and δl_3, therefore, the work done by the forces is $F_1\delta l_1 + F_2\delta l_2 + F_3\delta l_3$. Meanwhile, the work done by the voltage is $U\delta Q$ assuming that charges in amount of δQ flows through the thin film. When the dielectric elastomer is in an equilibrium state of mechanical forces and electrical voltage, the increase in the free energy should be equal to the total work,

$$\delta H = F_1\delta l_1 + F_2\delta l_2 + F_3\delta l_3 + U\delta Q \tag{1}$$

Here H is the Helmholtz free energy of such a thermodynamic system. In a Cartesian coordinate system, the elastomer's stretches in the three direction are defined as: $\lambda_1 = l_1/L_1$, $\lambda_2 = l_2/L_2$, and $\lambda_3 = l_3/L_3$. The nominal stresses s_1, s_2, and s_3 are defined by dividing the pre-stretch forces by the area before deformation, i.e., $s_1 = F_1/(l_2l_3)$, $s_2 = F_2/(l_1l_3)$, and $s_3 = F_3/(l_1l_2)$. Similarly, the nominal electric field can be defined as $E^{\sim} = U/L_3$ and the nominal electrical displacement as $D^{\sim} = Q/(L_1L_2)$. The nominal density of the Helmholtz free energy is defined as $W = H/(L_1L_2L_3)$. Furthermore, define the true stresses by $\sigma_1 = F_1/(\lambda_2\lambda_3L_2L_3)$, $\sigma_2 = F_2/(\lambda_1\lambda_3L_1L_3)$, $\sigma_3 = F_3/(\lambda_1\lambda_2L_1L_2)$, the true electric field by $E = U/(\lambda_3L_3)$, and the true electric displacement by $D = Q/(\lambda_1\lambda_2L_1L_2)$.

The condition of equilibrium (1) holds in any current state. Based on the above definitions, the small change of the Helmholtz free energy density is expressed as:

$$\delta W = s_1\delta\lambda_1 + s_2\delta\lambda_2 + s_3\delta\lambda_3 + E^{\sim}\delta D^{\sim} \tag{2}$$

This equilibrium condition will hold for arbitrary small variations of the four independent variables, $\lambda_1, \lambda_2, \lambda_3$, and $D^{\sim}$. Neglecting the variation of temperature, to characterize the dielectric elastomer, the nominal density of the Helmholtz free energy is prescribed as a function of the four independent variables,

$$W = W(\lambda_1, \lambda_2, \lambda_3, D^{\sim}) \tag{3}$$

Under the coupling effect of electric and mechanical fields, the small variation of the four independent variables are $d\lambda_1, d\lambda_2, d\lambda_3$,, and $dD^{\sim}$, respectively. The variation of free energy of dielectric elastomer's electromechanical coupling system can be expressed as following:

$$\delta W = \frac{\partial W(\lambda_1,\lambda_2,\lambda_3,D^{\sim})}{\partial\lambda_1}\delta\lambda_1 + \frac{\partial W(\lambda_1,\lambda_2,\lambda_3,D^{\sim})}{\partial\lambda_2}\delta\lambda_2 + \frac{\partial W(\lambda_1,\lambda_2,\lambda_3,D^{\sim})}{\partial\lambda_3}\delta\lambda_3 + \frac{\partial(\lambda_1,\lambda_2,\lambda_3,D^{\sim})}{\partial D^{\sim}}\delta D^{\sim} \tag{4}$$

Inserting Eq. (4) into Eq. (2), then we can obtain a thermodynamic equilibrium equation in form of,

$$\left[\frac{\partial W(\lambda_1,\lambda_2,\lambda_3,D^{\sim})}{\partial\lambda_1} - s_1\right]\delta\lambda_1 + \left[\frac{\partial W(\lambda_1,\lambda_2,\lambda_3,D^{\sim})}{\partial\lambda_2} - s_2\right]\delta\lambda_2 + \left[\frac{\partial W(\lambda_1,\lambda_2,\lambda_3,D^{\sim})}{\partial\lambda_3} - s_3\right]\delta\lambda_3 + \left[\frac{\partial(\lambda_1,\lambda_2,\lambda_3,D^{\sim})}{\partial D^{\sim}} - E^{\sim}\right]\delta D^{\sim} = 0 \tag{5}$$

This condition of equilibrium holds for any small variations of the four independent variables. Consequently, when the dielectric elastomer is in equilibrium with the applied forces and the applied voltage, the coefficient in front of the variation of each independent variable vanishes, giving,

$$s_1 = \frac{\partial W(\lambda_1,\lambda_2,\lambda_3,D^{\sim})}{\partial\lambda_1} \tag{6}$$

$$s_2 = \frac{\partial W(\lambda_1,\lambda_2,\lambda_3,D^{\sim})}{\partial\lambda_2} \tag{7}$$

$$s_3 = \frac{\partial W(\lambda_1,\lambda_2,\lambda_3,D^{\sim})}{\partial\lambda_3} \tag{8}$$

$$E^{\sim} = \frac{\partial W(\lambda_1,\lambda_2,\lambda_3,D^{\sim})}{\partial D^{\sim}} \tag{9}$$

According to the above-mentioned definition, the true stress and the true electric filed can be defined as

$$\sigma_1 = \frac{\partial W(\lambda_1,\lambda_2,\lambda_3,D^{\sim})}{\lambda_2\lambda_3\partial\lambda_1} \tag{10}$$

$$\sigma_2 = \frac{\partial W(\lambda_1,\lambda_2,\lambda_3,D^{\sim})}{\lambda_1\lambda_3\partial\lambda_2} \tag{11}$$

$$\sigma_3 = \frac{\partial W(\lambda_1,\lambda_2,\lambda_3,D^{\sim})}{\lambda_1\lambda_2\partial\lambda_3} \tag{12}$$

$$E = \frac{\partial W(\lambda_1,\lambda_2,\lambda_3,D^{\sim})}{\lambda_3\partial D^{\sim}} \tag{13}$$

Once the free energy function $W(\lambda_1, \lambda_2, \lambda_3, D^{\sim})$ is determined, Eqs. (6)–(13) will constitute the equations of state for the dielectric elastomer.

Actually, as a dielectric elastomer thermodynamic system, the nominal density of Helmholtz free energy density is taken to be a function of the four independent variables, $W = W(\lambda_1, \lambda_2, \lambda_3, D)$, according to the above-mentioned method, using the true electric displacement to express Eqs. (10)–(13), we obtain that,

$$\sigma_1 = \frac{\partial W(\lambda_1,\lambda_2,\lambda_3,D)}{\lambda_2\lambda_3\partial\lambda_1} - ED \tag{14}$$

$$\sigma_2 = \frac{\partial W(\lambda_1,\lambda_2,\lambda_3,D)}{\lambda_1\lambda_3\partial\lambda_2} - ED \tag{15}$$

$$\sigma_3 = \frac{\partial W(\lambda_1,\lambda_2,\lambda_3,D)}{\lambda_1\lambda_2\partial\lambda_3} \tag{16}$$

$$E = \frac{\partial W(\lambda_1,\lambda_2,\lambda_3,D)}{\lambda_1\lambda_2\lambda_3\partial D} \tag{17}$$

The Eqs. (14)–(17) coincide with Suo's investigations (Suo, 2010). Eqs. (14)–(17) constitute the equations of state in terms of true quantities for an elastic dielectric once the function $(\lambda_1, \lambda_2, \lambda_3, D)$ is given.

The permittivity of vacuum is $\varepsilon_0 = D/E$, consider the Eq. (17), the free energy density is $W(\lambda_1, \lambda_2, \lambda_3, D) = D^2\lambda_1\lambda_2\lambda_3/(2\varepsilon_0)$, the true stress are $\sigma_1 = \sigma_2 = -\varepsilon_0E^2/2$ and $\sigma_3 = \varepsilon_0E^2/2$ (Suo, 2010). The permittivity of ideal dielectric

elastomer is $\varepsilon = D/E$, consider the Eq. (17), the free energy density is $W(\lambda_1,\lambda_2,\lambda_3,D) = U(\lambda_1,\lambda_2,\lambda_3) + D^2\lambda_1\lambda_2\lambda_3/(2\varepsilon)$, the true stress are $\sigma_1 = \lambda_1(\partial U/\partial\lambda_1) - \varepsilon E^2/2, \sigma_2 = \lambda_2(\partial U/\partial\lambda_2) - \varepsilon E^2/2$, and $\sigma_3 = \lambda_3(\partial U/\partial\lambda_3)$, which are suitable to Suo's results (Suo, 2010). The permittivity of the quasi linear dielectric elastomer is a function of the stretches, $\varepsilon(\lambda_1,\lambda_2,\lambda_3) = D/E$, the free energy density is $W(\lambda_1,\lambda_2,\lambda_3,D) = U(\lambda_1,\lambda_2,\lambda_3) + D^2\lambda_1\lambda_2\lambda_3/[2\varepsilon(\lambda_1,\lambda_2,\lambda_3)]$, the true stress are $\sigma_1 = \lambda_1(\partial U/\partial\lambda_1) - \varepsilon(\lambda_1,\lambda_2,\lambda_3)E^2/2 - [\partial\varepsilon(\lambda_1,\lambda_2,\lambda_3)/(2\partial\lambda_1)]\lambda_1E^2, \sigma_2 = \lambda_2(\partial U/\partial\lambda_2) - \varepsilon(\lambda_1,\lambda_2,\lambda_3)E^2/2 - [\partial\varepsilon(\lambda_1,\lambda_2,\lambda_3)/(2\partial\lambda_2)]\lambda_2E^2$, and $\sigma_3 = \lambda_3(\partial U/\partial\lambda_3) + \varepsilon(\lambda_1,\lambda_2,\lambda_3)E^2/2 - [\partial\varepsilon(\lambda_1,\lambda_2,\lambda_3)/(2\partial\lambda_3)]\lambda_3E^2$, which are suitable to Suo's results (Suo, 2010).

The free energy of dielectric elastomer includes two parts of contributions: the stretch and the polarization. Therefore, the free energy density of a dielectric elastomer can be can be written as follows (Suo, 2010; Suo et al., 2008; Zhao and Suo, 2007a; Zhao et al., 2007b):

$$W(\lambda_1,\lambda_2,\lambda_3,D^\sim) = U(\lambda_1,\lambda_2,\lambda_3) + V(\lambda_1,\lambda_2,\lambda_3,D^\sim) \tag{18}$$

where $U(\lambda_1,\lambda_2,\lambda_3)$ is the elastic energy density of dielectric elastomer, and $V(\lambda_1,\lambda_2,\lambda_3,D^\sim)$ is the electric energy density.

Substitute Eq. (18) to Eqs. (6)–(9), the equilibrium functions of dielectric elastomers' thermodynamical system expressed by nominal stress and nominal electric field are as follows:

$$s_1 = \frac{\partial U(\lambda_1,\lambda_2,\lambda_3,D^\sim)}{\partial\lambda_1} + \frac{\partial V(\lambda_1,\lambda_2,\lambda_3,D^\sim)}{\partial\lambda_1} \tag{19}$$

$$s_2 = \frac{\partial U(\lambda_1,\lambda_2,\lambda_3,D^\sim)}{\partial\lambda_2} + \frac{\partial V(\lambda_1,\lambda_2,\lambda_3,D^\sim)}{\partial\lambda_2} \tag{20}$$

$$s_3 = \frac{\partial U(\lambda_1,\lambda_2,\lambda_3,D^\sim)}{\partial\lambda_3} + \frac{\partial V(\lambda_1,\lambda_2,\lambda_3,D^\sim)}{\partial\lambda_3} \tag{21}$$

$$E^\sim = \frac{\partial V(\lambda_1,\lambda_2,\lambda_3,D^\sim)}{\partial D^\sim} \tag{22}$$

Substitute Eq. (18) to Eqs. (10)–(13), the equilibrium functions of dielectric elastomers' thermodynamical system expressed by true stress and true electric field are as follows:

$$\sigma_1 = \frac{\partial U(\lambda_1,\lambda_2,\lambda_3,D^\sim)}{\lambda_2\lambda_3\partial\lambda_1} + \frac{\partial V(\lambda_1,\lambda_2,\lambda_3,D^\sim)}{\lambda_2\lambda_3\partial\lambda_1} \tag{23}$$

$$\sigma_2 = \frac{\partial U(\lambda_1,\lambda_2,\lambda_3,D^\sim)}{\lambda_1\lambda_3\partial\lambda_2} + \frac{\partial V(\lambda_1,\lambda_2,\lambda_3,D^\sim)}{\lambda_1\lambda_3\partial\lambda_2} \tag{24}$$

$$\sigma_3 = \frac{\partial U(\lambda_1,\lambda_2,\lambda_3,D^\sim)}{\lambda_1\lambda_2\partial\lambda_3} + \frac{\partial V(\lambda_1,\lambda_2,\lambda_3,D^\sim)}{\lambda_1\lambda_2\partial\lambda_3} \tag{25}$$

$$E = \frac{\partial V(\lambda_1,\lambda_2,\lambda_3,D^\sim)}{\lambda_3\partial D^\sim} \tag{26}$$

Eqs. (19)–(26) constitute yet another representation of the condition of equilibrium – they are called the equations of state. Once the elastic energy density function $U(\lambda_1,\lambda_2,\lambda_3,D^\sim)$ and electric energy density function $V(\lambda_1,\lambda_2,\lambda_3,D^\sim)$ are prescribed, the equations of state (19)–(26) give the values of the forces and voltage needed to equilibrate with the dielectric elastomer in the state $(\lambda_1,\lambda_2,\lambda_3,D^\sim)$.

3. Dielectric elastomers undergoing polarization saturation

Dielectric materials will be polarized in the presence of an external electric field E (Suo, 2010; He et al., 2009). For liquid dielectric, the polarization is the orientation polarization, attributed from the rotation of molecular dipoles, seen in Fig. 2(a). The electric displacement, interpreted as polarized charge density, is small and linear at weak electric field (Fig. 2(b)), and will approach a saturation value D_s when all the dipoles are perfectly aligned in the direction of a strong electric field (Fig. 2(c)). The overall nonlinear polarization curve is sketched in Fig. 2(d).

Dielectric elastomer soft materials usually undergo electromechanical instability. In the experiments conducted by Plante et al., the instability phenomenon of dielectric elastomers undergoing mechanical force and electrical force coupling field is observed (Plante and Dubowsky, 2006). Under certain voltage, the dielectric elastomers will deform, with part of the thin film smooth and the other parts wrinkled. For the voltage of each point is the same, the two states exist at the same time. In the wrinkled region, the film suffers from larger deformation and become thinner. The experiment above suggests that when suffering from electromechanical coupling effect, the electrical breakdown of the dielectric elastomer soft materials, which is induced by the electromechanical instability, will occur.

Under the electromechanical coupling effect, the dielectric elastomers become electromechanically instable. If the true electric field at the time is not beyond the critical electric field, the thin film will avoid breakdown and undergo snap-through deformation safely, reaching the equilibrium position eventually. The electric field increases drastically for the sharply decrease of the thickness. Further increase of the voltage can lead to the electrical breakdown. This kind of electromechanical instability dominated by snap-through deformation is called the snap-through instability of the dielectric elastomers.

The dielectric elastomers within strain stiffening effect may avoid electro-mechanical instability and the membrane may undergo snap-through instability. The dielectric elastomers may jump to larger deformation. This conclusion points out a kind of method to avoid and suppress the instability of dielectric elastomers, which can guide the design and manufacture of dielectric elastomer transducers within large deformation. What's more, it is also helpful to the research of the failure of dielectric elastomers.

The electrostriction of the dielectric elastomer soft materials is influenced by the two kinds of nonlinear electromechanical coupling behaviors which suffer from instability first and then final breakdown. Next, we further described the influence of these two kinds of instabilities to the dielectric elastomer's electrostriction.

Figs. 2 and 3 illustrate the influence on the electrostriction deformation of two material limits: the extension limit and the polarization saturation. As Fig. 3(a), when the dielectric elastomer is subject to mechanical forces, on approaching the extension limit, $\lambda_{\lim}$, the elastomer stiffens

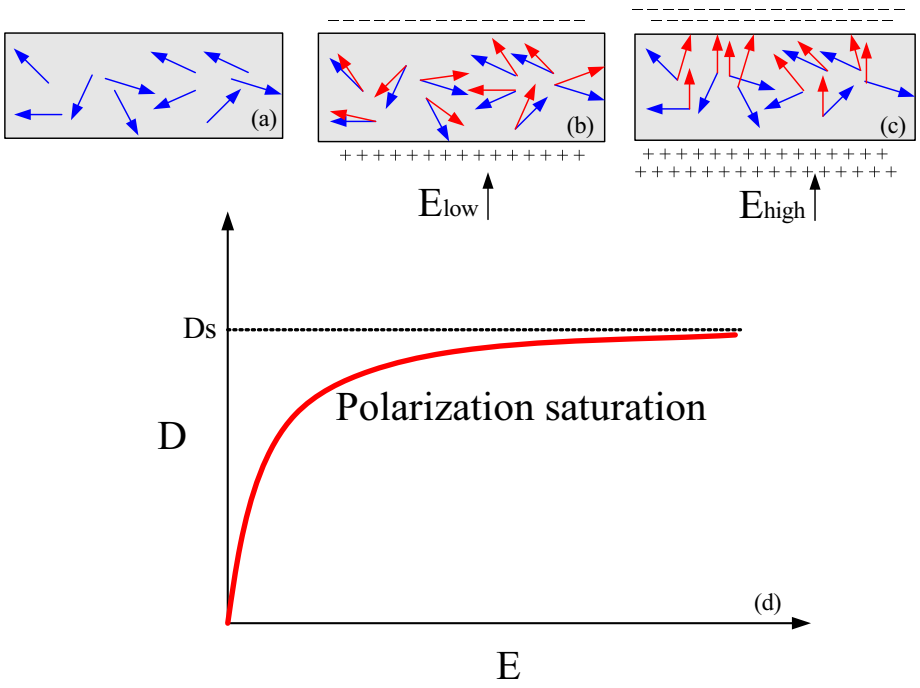

Fig. 2. Dielectric liquid contains (a) lots of molecular dipoles. (b) The polarization is a result of dipole rotation under an electric field, often interpreted as linear, and (c) polarization saturation is obtained when all dipoles are perfectly aligned, when the voltage is high enough as displayed in (d) the overall polarization curve.

steeply. The phenomenon is called the strain-stiffening of the dielectric elastomers.

In Fig. 3(b), when the deformation is caused by voltage, the voltage–stretch curve is typically not monotonic. The voltage attains a maximum at stretch λ_c. The critical stretch λ_c will be 1.26 if we use the Neo-Hookean elastic energy model (Zhao and Suo, 2007a), and λ_c will be 1.37 if we use the Mooney–Rivlin elastic energy model (Liu et al., 2008), these results are quite fit with the experiment results (Pelrine et al., 1998).

In Fig. 3(c), when the deformation is caused by voltage rather than the mechanical forces, the voltage–stretch curve is typically not monotonic. The voltage attains a local maximum at stretch λ_c. As the voltage ramps up further, the dielectric elastomer material snaps, and is stabilized at a stretch close to $\lambda_{\lim}$. In Fig. 3(c), we predict that by tuning loading path subjected to materials can lead to the snap-through instability of dielectric elastomers and then avoid the electrical breakdown. It is also been proved by the experiments of Christoph et al. (Keplinger et al., 2012).

As Fig. 3(d), when the applied pre-stretch λ_p are strong large, the local maximum disappears, leading to a monotonic voltage–deformation curve. Before the electric field is applied, the applied pre-stretch pull the dielectric elastomer toward the extension limit, so that the steep stiffening removes the local maximum of the voltage–stretch response. The mechanism may explain why mechanical forces enhance voltage–induced large deformation.

The critical stretch λ_c, corresponds to the onset of the electromechanical instability, the extension limit $\lambda_{\lim}$, corresponds to the stable state of the snap-through stability. The following content is to introduce the electromechanical instability and the snap-through instability of the dielectric elastomers. Subject to an electric voltage, a layer of a dielectric elastomer reduces its thickness, so that the voltage induces a high electric field. When the electric voltage ramps up, the membrane thins down, so that the same voltage will induce an even higher electric field. This unique physical process results in the electromechanical instability (Zhao and Suo, 2007a).

However, a dielectric elastomer may survive the electromechanical instability without electrical breakdown, and be stabilized in a state of a much smaller thickness, resulting in the snap-through instability (Zhao et al., 2007b; Li et al., 2011a,b). To attain a larger deformation of actuation, the dielectric elastomers should undergo the snap-through instability.

Illustrated in Fig. 2(a)–(c), when the voltage is applied on a dielectric elastomer with randomly oriented dipoles, the dipoles rotate and rearranges. When the voltage is high enough, the polarization of the material may saturate. It is necessary to think about nonlinear dielectric performance of the dielectric elastomer, because it would revise voltage–extension curve, furthermore it is helpful in designing high performance dielectric elastomer drivers.

A dielectric elastomer is a three-dimensional network of long and flexible polymer chains, held together by cross-links. Each polymer chain consists of such a large number of monomers that the cross-links affect polarization of the monomers negligibly. That is, the dielectric elastomer can polarize nearly as freely as a polymer melt. As an idealization, that the relation between the electric field is a function of the electric displacement independent of deformation (Li et al., 2011a,b),

$$E = f(D) \tag{27}$$

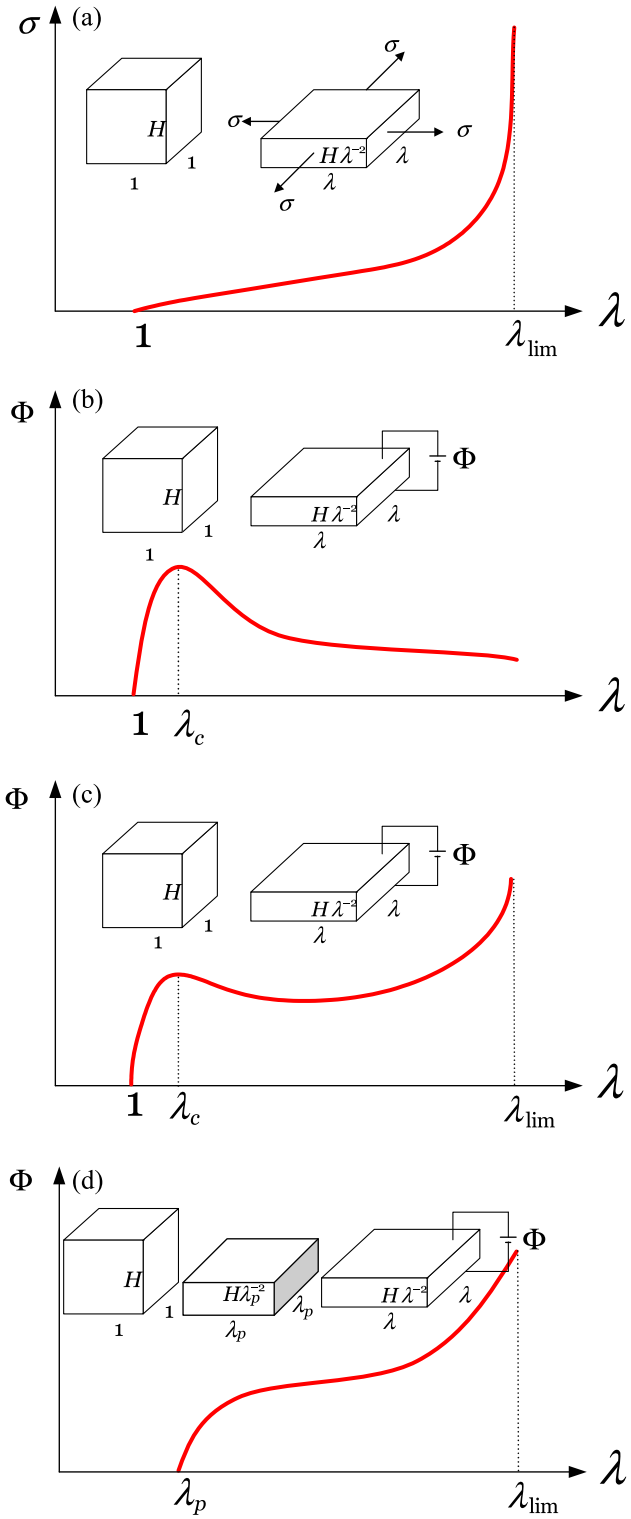

Fig. 3. (a) Stress–stretch curve of a dielectric elastomer membrane under biaxial stresses. The curve stiffens steeply upon approaching the extension limit. (b) Voltage–stretch curve of a dielectric elastomer membrane undergoing the pull-in electromechanical instability. (c) Voltage–stretch curve of a dielectric elastomer membrane undergoing the snap-through instability. Voltage–stretch curve of a dielectric elastomer membrane is typically not monotonic. (d) Voltage–stretch curve of a dielectric elastomer membrane undergoing the equal-biaxial pre-stretch. When the applied pre-stretch λ_p are strong large, the local maximum disappears, leading to a monotonic voltage–stretch curve.

Considering the work conjugated parameters: the nominal electric filed and the nominal electric displacement, the electric energy density of a dielectric elastomer undergoing large deformation can be written as follows (Suo, 2010; Suo et al., 2008):

$$V(D^{\sim}) = \int_0^{D^{\sim}} E^{\sim} dD^{\sim} \tag{28}$$

Considering equations $E^{\sim} = E\lambda_3, D = D^{\sim}\lambda_1^{-1}\lambda_2^{-1}$ and (28), we obtain that

$$V(\lambda_1, \lambda_2, \lambda_3, D^{\sim}) = \lambda_3 \int_0^{D^{\sim}} f\left(D^{\sim}\lambda_1^{-1}\lambda_2^{-1}\right) dD^{\sim} \tag{29}$$

Inserting Eq. (29) into Eqs. (19)–(22), we get that

$$s_1 = \frac{\partial U(\lambda_1, \lambda_2, \lambda_3)}{\partial \lambda_1} + \lambda_3 \frac{\partial}{\partial \lambda_1}\left[\int_0^{D^{\sim}} f(D^{\sim}\lambda_1^{-1}\lambda_2^{-1}) dD^{\sim}\right] \tag{30}$$

$$s_2 = \frac{\partial U(\lambda_1, \lambda_2, \lambda_3)}{\partial \lambda_2} + \lambda_3 \frac{\partial}{\partial \lambda_2}\left[\int_0^{D^{\sim}} f(D^{\sim}\lambda_1^{-1}\lambda_2^{-1}) dD^{\sim}\right] \tag{31}$$

$$s_3 = \frac{\partial U(\lambda_1, \lambda_2, \lambda_3)}{\partial \lambda_3} + \lambda_3 \frac{\partial}{\partial \lambda_3}\left[\int_0^{D^{\sim}} f(D^{\sim}\lambda_1^{-1}\lambda_2^{-1}) dD^{\sim}\right] \tag{32}$$

$$E^{\sim} = \lambda_3 \frac{\partial}{\partial D^{\sim}}\left[\int_0^{D^{\sim}} f(D^{\sim}\lambda_1^{-1}\lambda_2^{-1}) dD^{\sim}\right] \tag{33}$$

Inserting Eq. (29) into Eqs. (23)–(26), we obtain the following expressions of the true stress and the true electric filed

$$\sigma_1 = \frac{\partial U(\lambda_1, \lambda_2, \lambda_3)}{\lambda_2\lambda_3 \partial \lambda_1} + \frac{1}{\lambda_2}\frac{\partial}{\partial \lambda_1}\left[\int_0^{D^{\sim}} f\left(D^{\sim}\lambda_1^{-1}\lambda_2^{-1}\right) dD^{\sim}\right] \tag{34}$$

$$\sigma_2 = \frac{\partial U(\lambda_1, \lambda_2, \lambda_3)}{\lambda_1\lambda_3 \partial \lambda_2} + \frac{1}{\lambda_1}\frac{\partial}{\partial \lambda_2}\left[\int_0^{D^{\sim}} f\left(D^{\sim}\lambda_1^{-1}\lambda_2^{-1}\right) dD^{\sim}\right] \tag{35}$$

$$\sigma_3 = \frac{\partial U(\lambda_1, \lambda_2, \lambda_3)}{\lambda_1\lambda_2 \partial \lambda_3} + \frac{\lambda_3}{\lambda_1\lambda_2}\frac{\partial}{\partial \lambda_3}\left[\int_0^{D^{\sim}} f\left(D^{\sim}\lambda_1^{-1}\lambda_2^{-1}\right) dD^{\sim}\right] \tag{36}$$

$$E = \frac{\partial}{\partial D^{\sim}}\left[\int_0^{D^{\sim}} f\left(D^{\sim}\lambda_1^{-1}\lambda_2^{-1}\right) dD^{\sim}\right] \tag{37}$$

Eqs. (30)–(37) are the equilibrium equations of dielectric elastomer undergoing polarization saturation. Once the elastic energy density and the electric energy density are prescribed as a material model, the equations of state (30)–(37) give the values of the forces and voltage needed to equilibrate with the dielectric elastomer undergoing polarization saturation in the state $(\lambda_1, \lambda_2, \lambda_3, D^{\sim})$.

4. Incompressible dielectric elastomer undergoing polarization saturation

The dielectric elastomer is taken to be incompressible, $\lambda_1\lambda_2\lambda_3 = 1, \delta\lambda_3 = -\lambda_1^{-2}\lambda_2^{-1}\delta\lambda_1 - \lambda_1^{-1}\lambda_2^{-2}\delta\lambda_2$, Inserting it into Eq. (2), $\delta W = (s_1 - \lambda_1^{-2}\lambda_2^{-1}s_3)\delta\lambda_1 + (s_2 - \lambda_1^{-1}\lambda_2^{-2}s_3)\delta\lambda_2 + E^{\sim}\delta D^{\sim}$. The nominal stress in the two planar principal directions, the nominal electric field in the thickness direction of incompressible dielectric elastomer's thermodynamic system are obtained respectively, as is shown in the followings:

$$s_1-\frac{s_3}{\lambda_1^2\lambda_2}=\frac{\partial U(\lambda_1,\lambda_2,\lambda_1^{-1}\lambda_2^{-1})}{\partial\lambda_1}+\frac{\partial}{\partial\lambda_1}\left\{\int_0^{D^\sim}\left[\lambda_1^{-1}\lambda_2^{-1}f(D^\sim\lambda_1^{-1}\lambda_2^{-1})\right]dD^\sim\right\} \tag{38}$$

$$s_2-\frac{s_3}{\lambda_1\lambda_2^2}=\frac{\partial U(\lambda_1,\lambda_2,\lambda_1^{-1}\lambda_2^{-1})}{\partial\lambda_2}+\frac{\partial}{\partial\lambda_2}\left\{\int_0^{D^\sim}\left[\lambda_1^{-1}\lambda_2^{-1}f(D^\sim\lambda_1^{-1}\lambda_2^{-1})\right]dD^\sim\right\} \tag{39}$$

$$E^\sim=\lambda_1^{-1}\lambda_2^{-1}\frac{\partial}{\partial D^\sim}\left[\int_0^{D^\sim}f(D^\sim\lambda_1^{-1}\lambda_2^{-1})dD^\sim\right] \tag{40}$$

And the true stress and the true electric filed can be expressed as followings

$$\sigma_1-\sigma_3=\lambda_1\frac{\partial U(\lambda_1,\lambda_2,\lambda_1^{-1}\lambda_2^{-1})}{\partial\lambda_1}+\lambda_1\times\frac{\partial}{\partial\lambda_1}\left\{\int_0^{D^\sim}\left[\lambda_1^{-1}\lambda_2^{-1}f(D^\sim\lambda_1^{-1}\lambda_2^{-1})\right]dD^\sim\right\} \tag{41}$$

$$\sigma_2-\sigma_3=\lambda_2\frac{\partial U(\lambda_1,\lambda_2,\lambda_1^{-1}\lambda_2^{-1})}{\partial\lambda_2}+\lambda_2\times\frac{\partial}{\partial\lambda_2}\left\{\int_0^{D^\sim}\left[\lambda_1^{-1}\lambda_2^{-1}f(D^\sim\lambda_1^{-1}\lambda_2^{-1})\right]dD^\sim\right\} \tag{42}$$

$$E=\frac{\partial}{\partial D^\sim}\left[\int_0^{D^\sim}f(D^\sim\lambda_1^{-1}\lambda_2^{-1})dD^\sim\right] \tag{43}$$

Eqs. (38)–(43) constitute the equations of state of incompressible dielectric elastomer undergoing polarization saturation. If the elastic energy and the electric energy are given, we can obtain the stress and the electric field of incompressible dielectric elastomer in the equilibrium state.

5. A special electric energy

To obtain the special electric field energy, we consider the nonlinear dielectric behavior of a dielectric elastomer. According to Eq. (27), for example, we get the expression of the true electric field and the true electric displacement of dielectric elastomers undergoing polarization saturation as follows (Gong and Suo, 1996; Yang and Suo, 1994; Li et al., 2011a,b):

$$E=\frac{D_s}{2\varepsilon}\log\left(\frac{1+D/D_s}{1-D/D_s}\right) \tag{44}$$

Here D_s is the saturated electric displacement, E_s is a characteristic electric field. When E is small, D is linearly proportional to E, recovers the linear dielectric behavior. When E is high enough, dielectric elastomer is in the state of polarization saturation, $D\to D_s$. According to Eqs. (44), (27)–(29), we obtain the special electric energy of dielectric elastomer,

$$V(\lambda_1,\lambda_2,\lambda_3,D^\sim)=\frac{D_sD^\sim\lambda_3}{2\varepsilon}\log\left(\frac{1+D^\sim\lambda_1^{-1}\lambda_2^{-1}/D_s}{1-D^\sim\lambda_1^{-1}\lambda_2^{-1}/D_s}\right)+\frac{D_s^2\lambda_1\lambda_2\lambda_3}{2\varepsilon}\log\left(1-\frac{D^{\sim2}\lambda_1^{-2}\lambda_2^{-2}}{D_s^2}\right) \tag{45}$$

Considering the Eq. (45), submit Eq. (18) into Eqs. (30)–(33), the nominal stress of dielectric elastomer's thermodynamic system in the three planar principal directions, the nominal electric field in the thickness direction are obtained respectively, as is shown in the followings:

$$s_1=\frac{\partial U(\lambda_1,\lambda_2,\lambda_3,D^\sim)}{\partial\lambda_1}+\frac{D_s^2}{2\varepsilon}\lambda_2\lambda_3\log\left(1-\frac{D^{\sim2}\lambda_1^{-2}\lambda_2^{-2}}{D_s^2}\right) \tag{46}$$

$$s_2=\frac{\partial U(\lambda_1,\lambda_2,\lambda_3,D^\sim)}{\partial\lambda_2}+\frac{D_s^2}{2\varepsilon}\lambda_1\lambda_3\log\left(1-\frac{D^{\sim2}\lambda_1^{-2}\lambda_2^{-2}}{D_s^2}\right) \tag{47}$$

$$s_3=\frac{\partial U(\lambda_1,\lambda_2,\lambda_3,D^\sim)}{\partial\lambda_3}+\frac{D_s^{\sim2}}{2\varepsilon}\lambda_1\lambda_2\log\left(1-\frac{D^{\sim2}\lambda_1^{-2}\lambda_2^{-2}}{D_s^2}\right)+\frac{D_sD^\sim}{2\varepsilon}\log\left(\frac{1+D^\sim\lambda_1^{-1}\lambda_2^{-1}/D_s}{1-D^\sim\lambda_1^{-1}\lambda_2^{-1}/D_s}\right) \tag{48}$$

$$E^\sim=\frac{D_s\lambda_3}{2\varepsilon}\log\left(\frac{1+D^\sim\lambda_1^{-1}\lambda_2^{-1}/D_s}{1-D^\sim\lambda_1^{-1}\lambda_2^{-1}/D_s}\right) \tag{49}$$

Inserting Eqs. (46)–(49) into Eqs. (34)–(37), we can write the true stress and the true electric field, respectively, as

$$\sigma_1=\frac{\partial U(\lambda_1,\lambda_2,\lambda_3,D^\sim)}{\lambda_2\lambda_3\partial\lambda_1}+\frac{D_s^2}{2\varepsilon}\log\left(1-\frac{D^{\sim2}\lambda_1^{-2}\lambda_2^{-2}}{D_s^2}\right) \tag{50}$$

$$\sigma_2=\frac{\partial U(\lambda_1,\lambda_2,\lambda_3,D^\sim)}{\lambda_1\lambda_3\partial\lambda_2}+\frac{D_s^2}{2\varepsilon}\log\left(1-\frac{D^{\sim2}\lambda_1^{-2}\lambda_2^{-2}}{D_s^2}\right) \tag{51}$$

$$\sigma_3=\frac{\partial U(\lambda_1,\lambda_2,\lambda_3,D^\sim)}{\lambda_1\lambda_2\partial\lambda_3}+\frac{D_s^{\sim2}}{2\varepsilon}\log\left(1-\frac{D^{\sim2}\lambda_1^{-2}\lambda_2^{-2}}{D_s^2}\right)+\frac{D_sD^\sim}{2\varepsilon}\lambda_1^{-1}\lambda_2^{-1}\log\left(\frac{1+D^\sim\lambda_1^{-1}\lambda_2^{-1}/D_s}{1-D^\sim\lambda_1^{-1}\lambda_2^{-1}/D_s}\right) \tag{52}$$

$$E=\frac{D_s}{2\varepsilon}\log\left(\frac{1+D/D_s}{1-D/D_s}\right) \tag{53}$$

For incompressible dielectric elastomer, submit Eq. (33) into Eqs. (30)–(35), the nominal stress and the nominal electric field become

$$s_1-\frac{s_3}{\lambda_1^2\lambda_2}=\frac{\partial U(\lambda_1,\lambda_2,\lambda_1^{-1}\lambda_2^{-1})}{\partial\lambda_1}-\frac{D_sD^\sim\lambda_1^{-2}\lambda_2^{-1}}{2\varepsilon}\log\left(\frac{1+D^\sim\lambda_1^{-1}\lambda_2^{-1}/D_s}{1-D^\sim\lambda_1^{-1}\lambda_2^{-1}/D_s}\right) \tag{54}$$

$$s_2-\frac{s_3}{\lambda_1\lambda_2^2}=\frac{\partial U(\lambda_1,\lambda_2,\lambda_1^{-1}\lambda_2^{-1})}{\partial\lambda_2}-\frac{D_sD^\sim\lambda_1^{-1}\lambda_2^{-2}}{2\varepsilon}\log\left(\frac{1+D^\sim\lambda_1^{-1}\lambda_2^{-1}/D_s}{1-D^\sim\lambda_1^{-1}\lambda_2^{-1}/D_s}\right) \tag{55}$$

$$E^\sim=\frac{D_s\lambda_1^{-1}\lambda_2^{-1}}{2\varepsilon}\log\left(\frac{1+D^\sim\lambda_1^{-1}\lambda_2^{-1}/D_s}{1-D^\sim\lambda_1^{-1}\lambda_2^{-1}/D_s}\right) \tag{56}$$

According to the above-mentioned definition, the true stress and true electric field can be expressed as followings:

$$\sigma_1-\sigma_3=\lambda_1\frac{\partial U(\lambda_1,\lambda_2,\lambda_1^{-1}\lambda_2^{-1})}{\partial\lambda_1}-\frac{D_sD^{\sim}\lambda_1^{-1}\lambda_2^{-1}}{2\varepsilon}\log\left(\frac{1+D^{\sim}\lambda_1^{-1}\lambda_2^{-1}/D_s}{1-D^{\sim}\lambda_1^{-1}\lambda_2^{-1}/D_s}\right) \tag{57}$$

$$\sigma_2-\sigma_3=\lambda_2\frac{\partial U(\lambda_1,\lambda_2,\lambda_1^{-1}\lambda_2^{-1})}{\partial\lambda_2}-\frac{D_sD^{\sim}\lambda_1^{-1}\lambda_2^{-1}}{2\varepsilon}\log\left(\frac{1+D^{\sim}\lambda_1^{-1}\lambda_2^{-1}/D_s}{1-D^{\sim}\lambda_1^{-1}\lambda_2^{-1}/D_s}\right) \tag{58}$$

$$E=\frac{D_s}{2\varepsilon}\log\left(\frac{1+D/D_s}{1-D/D_s}\right) \tag{59}$$

In Eqs. (57) and (58), the first item is the stress due to hyperelasticity of the dielectric elastomers, the second item is Maxwell stress, which is related to the polarization saturation. Considering of the nonlinear dielectric behavior, the Maxwell stress can be expressed as

$$\sigma_M=\frac{1}{2}\varepsilon E_s^2\left[\log\left(\frac{1+\tanh(E/E_s)}{1-\tanh(E/E_s)}\right)\right] \tag{60}$$

As we know, if we assume that the elastomer is a nonlinear dielectric behavior, the Maxwell stress of ideal dielectric elastomer is (Suo, 2010; Zhao and Suo, 2007a)

$$\sigma_M=\varepsilon E^2 \tag{61}$$

Based on Eqs. (60) and (61), a dimensionless parameter $\sigma_M/\varepsilon E_s^2$ is used to characterize the Maxwell stress. And a dimensionless parameter E/E_s is used to characterize the electric field. Fig. 4 shows the Maxwell stress of the dielectric elastomers undergoing the linear and the nonlinear dielectric behaviors, respectively. When dielectric elastomer is linear dielectric behavior, $D=\varepsilon E$, the Maxwell stress is $\sigma_M=\varepsilon E^2$. However thinking about the nonlinear dielectric behavior, the Maxwell stress DE becomes to D_sE, which goes up linearly along with electric field increasing. Comparing to the linear dielectric behavior, the Maxwell stress goes up slowly due to the polarization saturation behavior. Due to that the Maxwell stress is limited, it is hard for dielectric elastomer under polarization saturation to fail. Therefore the nonlinear dielectric elastomer more tends to be in a stable state. Therefore, the nonlinear dielectric elastomer more tends to be in a stable state, which suggests that dielectric elastomer undergoing polarization saturation may qualify super-large deformation.

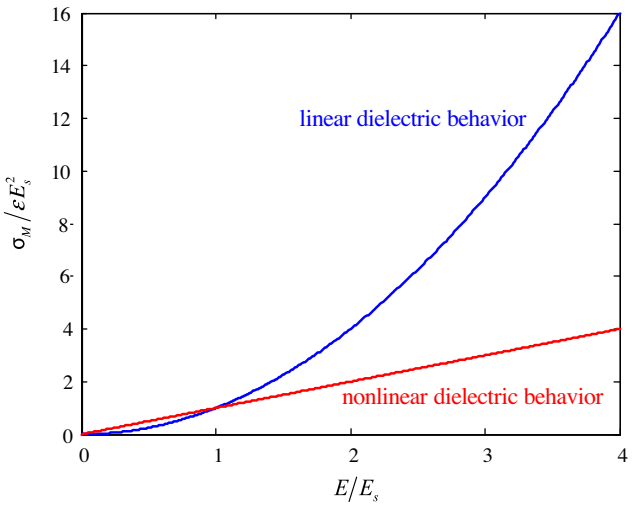

Fig. 4. Maxwell stress of a dielectric elastomer undergoing linear dielectric behavior and nonlinear dielectric behavior.

6. Electromechanical stability and snap-through stability

We next focus on the electromechanical stability and the snap stability of dielectric elastomer undergoing the polarization saturation behavior. Considering a equal biaxial forces $F_1=F_2=F$, $F_3=0$ applied on the dielectric elastomer, as well as voltage U. Write the three stretches as $\lambda_1=\lambda_2=\lambda$ and $\lambda_3=\lambda^{-2}$ (due to the incompressibility). The equal-biaxial planar stretch process ($\lambda_1=\lambda_2=\lambda$) stands for the physical process of using mechanical force to make the dielectric elastomer pre-stretched and then apply electric field to it. Based on the above-mentioned study of dielectric elastomer actuation (Liu et al., 2008, 2010b), we introduce the special elastic strain energy, Mooney–Rivlin model, $U(\lambda)=\frac{C_1}{2}(2\lambda^2+\lambda^{-4}-3)+\frac{C_2}{2}(2\lambda^{-2}+\lambda^4-3)$, C_1 and C_2 are the material constants. To analyze the stability of different materials or structure of dielectric elastomers, we propose a dimensionless parameter k (Liu et al., 2008). Let $C_2=kC_1$ (k is a constant), k is determinated by the material and the structure. The free energy can be simplified as

$$W(\lambda,D^{\sim})=\frac{C_1}{2}(2\lambda^2+\lambda^{-4}-3)+\frac{kC_1}{2}(2\lambda^{-2}+\lambda^4-3)+\frac{D_sD^{\sim}\lambda^{-2}}{2\varepsilon}\log\left(\frac{1+D^{\sim}\lambda^{-2}/D_s}{1-D^{\sim}\lambda^{-2}/D_s}\right)+\frac{D_s^2}{2\varepsilon}\log\left(1-\frac{D^{\sim 2}\lambda^{-4}}{D_s^2}\right) \tag{62}$$

We obtain the dimensionless nominal stress and the nominal electric field of dielectric elastomerm respectively

$$\frac{s}{C_1}=2(\lambda-\lambda^{-5})+2k(\lambda^3-\lambda^{-3})-\frac{D_s}{\sqrt{C_1\varepsilon}}\frac{D^{\sim}\lambda^{-3}}{\sqrt{C_1\varepsilon}}\log\left(\frac{1+\frac{D^{\sim}\lambda^{-2}}{\sqrt{C_1\varepsilon}}\Big/\frac{D_s}{\sqrt{C_1\varepsilon}}}{1-\frac{D^{\sim}\lambda^{-2}}{\sqrt{C_1\varepsilon}}\Big/\frac{D_s}{\sqrt{C_1\varepsilon}}}\right) \tag{63}$$

$$\frac{E^{\sim}}{E_s}=\frac{\lambda^{-2}}{2}\log\left(\frac{1+D^{\sim}\lambda^{-2}/D_s}{1-D^{\sim}\lambda^{-2}/D_s}\right) \tag{64}$$

By Eq. (64), we obtain that $D^{\sim}=\left[\frac{\exp(2E^{\sim}\lambda^2/E_s)-1}{\exp(2E^{\sim}\lambda^2/E_s)+1}\right]D_s\lambda^2$, $\log\left(\frac{1+D^{\sim}\lambda^{-2}/D_s}{1-D^{\sim}\lambda^{-2}/D_s}\right)=\frac{2E^{\sim}\lambda^2}{E_s}$, inserting it into Eq. (59), the relationship between nominal electric field and the stretch can be given, respectively, as

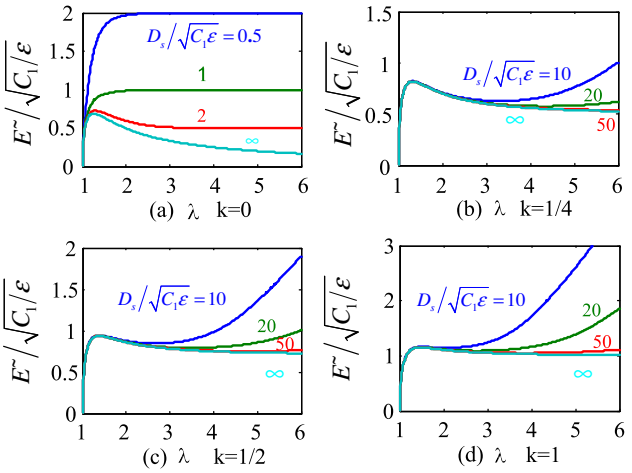

Fig. 5. Nominal electric field and stretch of dielectric elastomer undergoing polarization saturation under the loading condition $\lambda_1 = \lambda_2 = \lambda$.

$$\frac{s}{C_1} = 2(\lambda - \lambda^{-5}) + 2k(\lambda^3 - \lambda^{-3}) - 2 \times \frac{D_s}{\sqrt{C_1\varepsilon}} \left[\frac{\exp\left(2\frac{E^{\sim}}{\sqrt{C_1/\varepsilon}}\lambda^2 \Big/ \frac{D_s}{\sqrt{C_1\varepsilon}}\right) - 1}{\exp\left(2\frac{E^{\sim}}{\sqrt{C_1/\varepsilon}}\lambda^2 \Big/ \frac{D_s}{\sqrt{C_1\varepsilon}}\right) + 1} \right] \frac{E^{\sim}}{\sqrt{C_1/\varepsilon}}\lambda \quad (65)$$

Fig. 5 illustrates the relation between the nominal electric field and the stretch for different dielectric elastomer materials under biaxial stretch, in which $\lambda_1 = \lambda_2 = \lambda$. Here if $\frac{s}{C_1} = 0$, it means no mechanical force load on (a zero value of $\frac{s}{C_1}$ indicates the absence of mechanical force loads applied). The different material constant ratios k which differentiate Fig. 5(a)–(d), denote different dielectric elastomer materials (such as BJB TC-A/B-C, Dow Corning HS3silicone, Nusil CF19-2186 silicone, 3 M VHB 4905/4910, Dow Corning Sylgard 186, VHB 4910-HDDA, VHB 4905-TMPTMA, and CF19-2186 silicone, (Brochu and Pei, 2010; O'Halloran et al., 2008) or their various structures (such as rolling, folding, stacking, flatting, spring roll, tube, stacked linear contractile, and circular, Brochu and Pei, 2010; O'Halloran et al., 2008). If $k = 0$, $\lambda \to \infty$ and $\frac{D_s}{\sqrt{C_1\varepsilon}} = 0.5$, 1 and 2, $\frac{E^{\sim}_{\lim}}{\sqrt{C_1/\varepsilon}} \to 2$, 1 and 0.5, respectively, which is consistent with the conclusions given by our previous work (Li et al., 2011a,b). When k and $\frac{D_s}{\sqrt{C_1\varepsilon}}$ are both small enough, the dielectric elastomer undergoes snap instability. On the contrary, if $\frac{D_s}{\sqrt{C_1\varepsilon}} \to \infty$, the dielectric elastomer undergoes the electromechanical instability, which can be predigested by the previous investigation done by Liu et al. (2008). When k bears different values as 0, 1/4, 1/2, and 1, the corresponding critical nominal electric fields are $0.686\sqrt{C_1/\varepsilon}, 0.817\sqrt{C_1/\varepsilon}, 0.936\sqrt{C_1/\varepsilon}$, and $1.148\sqrt{C_1/\varepsilon}$, with the corresponding critical stretch being 1.26, 1.32, 1.38, and 1.47, respectively, which goes well with both of the conclusions given by Suo and Liu (Zhao and Suo, 2007a; Liu et al., 2008).

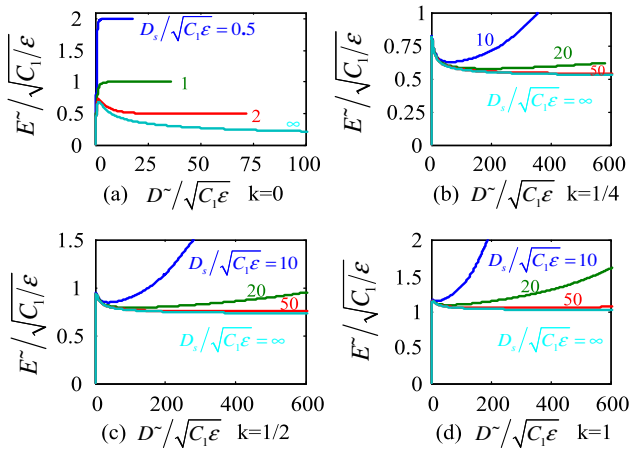

Fig. 6. Nominal electric field and nominal electric displacement of dielectric elastomer undergoing polarization saturation under the loading condition $\lambda_1 = \lambda_2 = \lambda$.

Fig. 6 shows the relation between the nominal electric field and the nominal electric displacement for different dielectric elastomer materials undergoing the polarization saturation and the equal biaxial stretch. When k is large and $\frac{D_s}{\sqrt{C_1\varepsilon}}$ is small, the nominal electric field increases monotonously along with the nominal electric displacement increasing. The local maximum of the voltage–stretch curve disappears; and dielectric elastomer reaches its stable state.

As a second special case, we take the stretching perpendicular to the elastomer film as $\lambda_3 = \lambda$, the thickness direction stretch process stands for the physical process of applying electric field to the dielectric elastomers. Owing to the incompressibility assumption for the dielectric elastomer, we have $\lambda_1 = \lambda_2 = \lambda^{-1/2}$, the free energy function can be expressed as follows:

$$W(\lambda, D^{\sim}) = \frac{C_1}{2}(2\lambda^{-1} + \lambda^2 - 3) + \frac{kC_1}{2}(2\lambda + \lambda^{-2} - 3) + \frac{D_s D^{\sim}\lambda}{2\varepsilon}\log\left(\frac{1 + D^{\sim}\lambda/D_s}{1 - D^{\sim}\lambda/D_s}\right) + \frac{D_s^2}{2\varepsilon}\log\left(1 - \frac{D^{\sim 2}\lambda^2}{D_s^2}\right) \quad (66)$$

The non-dimensional nominal electric field and nominal stress is

$$\frac{s}{C_1} = (\lambda - \lambda^{-2}) + k(1 - \lambda^{-3}) + \frac{D_s}{\sqrt{C_1\varepsilon}}\frac{D^{\sim}}{2\sqrt{C_1\varepsilon}}\log\left(\frac{1 + \frac{D^{\sim}}{\sqrt{C_1\varepsilon}}\lambda \Big/ \frac{D_s}{\sqrt{C_1\varepsilon}}}{1 - \frac{D^{\sim}}{\sqrt{C_1\varepsilon}}\lambda \Big/ \frac{D_s}{\sqrt{C_1\varepsilon}}}\right) \quad (67)$$

$$\frac{E^{\sim}}{E_s} = \frac{\lambda}{2}\log\left(\frac{1 + D^{\sim}\lambda/D_s}{1 - D^{\sim}\lambda/D_s}\right) \quad (68)$$

According to Eq. (60), we obtain that $D^{\sim} = \left[\frac{\exp(2E^{\sim}\lambda^{-1}/E_s) - 1}{\exp(2E^{\sim}\lambda^{-1}/E_s) + 1}\right] D_s\lambda^{-1}$, $\frac{2E^{\sim}\lambda^{-1}}{E_s} = \log\left(\frac{1 + D^{\sim}\lambda/D_s}{1 - D^{\sim}\lambda/D_s}\right)$, and deduced the governing equations of the nominal electric field and the nominal electric displacement in the equilibrium state

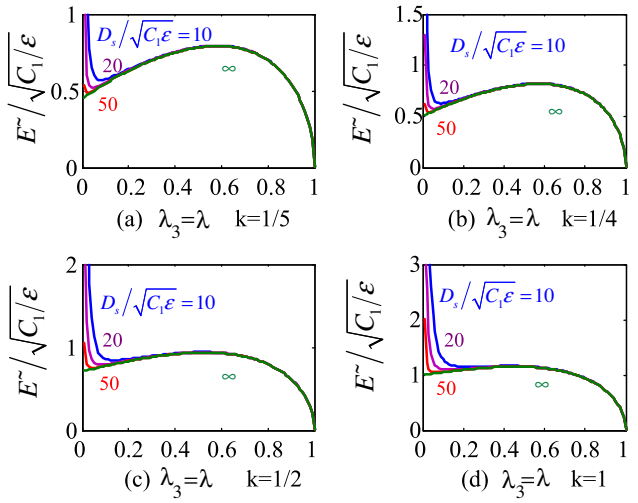

Fig. 7. Nominal electric field and stretch of dielectric elastomer undergoing polarization saturation under the loading condition $\lambda_3 = \lambda$.

$$\frac{s}{C_1}=(\lambda-\lambda^{-2})+k(1-\lambda^{-3}) +\frac{D_s}{\sqrt{C_1\varepsilon}}\left[\frac{\exp\left(2\frac{\tilde{E}}{\sqrt{C_1/\varepsilon}}\lambda^{-1}\Big/\frac{D_s}{\sqrt{C_1\varepsilon}}\right)-1}{\exp\left(2\frac{\tilde{E}}{\sqrt{C_1/\varepsilon}}\lambda^{-1}\Big/\frac{D_s}{\sqrt{C_1\varepsilon}}\right)+1}\right]\frac{\tilde{E}}{\sqrt{C_1/\varepsilon}}\lambda^{-2} \tag{69}$$

Fig. 7 depicts the relation between the nominal electric field and the stretch of dielectric elastomer undergoing the polarization saturation behavior when the Maxwell stress induced by the electric field is applied in the thickness direction. For the thickness strain λ_3 changing from 1 to 0, when k and $\frac{D_s}{\sqrt{C_1\varepsilon}}$ are both small enough, the nominal electric field can attain a local maximum value which indicates an electromechanical instability. If dielectric elastomer survived the electromechanical instability without electrical breakdown, the membrane would keep stable with a much smaller thickness, which could result in the snap-through instability. Therefore, such a theory can be used to find the dielectric elastomer transducers with high electrostriction performance. When $k=\frac{1}{5}$ and $\frac{D_s}{\sqrt{C_1\varepsilon}}=10$, the critical nominal electric field induced by the pull-in electromechanical instability stability is $0.794\sqrt{C_1/\varepsilon}$, with the corresponding critical stretch being 0.58.

On the other hand, when k is large and $\frac{D_s}{\sqrt{C_1\varepsilon}}$ is relatively small, the nominal electric field goes up monotonously before reaching the stable state. If $\frac{D_s}{\sqrt{C_1\varepsilon}}=\infty$, k = 1/5, 1/4, 1/2, and 1, the dielectric elastomer is under the electromechanical instable state and the critical nominal electric fields are $0.792\sqrt{C_1/\varepsilon}, 0.817\sqrt{C_1/\varepsilon}, 0.936\sqrt{C_1/\varepsilon}$ and $1.148\sqrt{C_1/\varepsilon}$ with the critical stretches being 0.58, 0.57, 0.53, and 0.46, respectively.

7. Effect of strain-stiffening on electromechanical instability and snap-through instability

In the study above, the material is assumed to be an ideal dielectric elastomer, where the dielectric behavior of the elastomer is taken to be liquid-like, unaffected by deformation. An ideal dielectric elastomer is a three-dimensional network of long and flexible polymer chains. Each polymer chain consists of a large number of monomers. When the polymer is applied to the mechanical force, it elongates. Upon release, the polymers spontaneously return to the original arrangement.

However, dielectric elastomers used in practice may exhibit strain-stiffening due to the finite contour length of polymer chains. This phenomenon cannot be described by a Mooney–Rivlin or Neo-Hookean model (Zhu et al., 2010c). Due to the polymer is compliant at small stretches, but stiffens steeply when pulled near the full length of the polymer (Zhao and Suo, 2010). Considering the incompressible dielectric elastomer, therefore, the strain energy model developed by Gent is employed to characterize the stiffening behavior, as

$$U(\lambda_1,\lambda_2)=-\frac{\mu}{2}J_{\lim}\log\left(1-\frac{I_1-3}{J_{\lim}}\right) \tag{70}$$

where I_1 is left Cauchy–Green deformation tensor, $I_1=\lambda_1^2+\lambda_2^2+\lambda_1^{-2}\lambda_2^{-2}$, $J_{\lim}$ is a constant related to the limiting stretch, $J_{\lim}=\lambda_{1\lim}^2+\lambda_{2\lim}^2+\lambda_{1\lim}^{-2}\lambda_{2\lim}^{-2}-3$, and μ is the small-strain shear modulus. When $(I_1-3)/J_{\lim}\to 0$, the Taylor expansion of (70) gives $U=\frac{\mu}{2}(I_1-3)$. That is, the Gent model recovers the Neo-Hookean model when deformation is small compared to the limiting stretch. When $(I_1-3)/J_{\lim}\to 1$, the elastomer approaches the limiting stretch. Further, we expand the natural logarithm of Gent model as

$$U(\lambda_1,\lambda_2)=\frac{\mu}{2}\left[(I_1-3)+\frac{1}{2J_{\lim}}(I_1-3)^2+\cdots+\frac{1}{(n+1)J_{\lim}^n}(I_1-3)^{n+1}\right],$$

a general form is $U(\lambda_1,\lambda_2)=\sum_{i=1}^{n}C_i(J_{\lim})(I_1-3)^i$, which can be viewed as a expression of Rivlin elastic strain energy model $U(\lambda_1,\lambda_2)=\sum_{i,j=0}^{\infty}C_{ij}(I_1-3)^i(I_2-3)^j$ when j = 0.

Inserting Eq. (70) into Eqs. (54), (55) and (57), (58), we get the nominal stress and the true stress of dielectric elastomer undergoing strain-stiffening as shown below

$$s_1-\frac{s_3}{\lambda_1^2\lambda_2}=\frac{\mu J_{\lim}}{J_{\lim}-(I_1-3)}\left(\lambda_1-\lambda_1^{-3}\lambda_2^{-2}\right) -\frac{D_sD^{\sim}\lambda_1^{-2}\lambda_2^{-1}}{2\varepsilon}\log\left(\frac{1+D^{\sim}\lambda_1^{-1}\lambda_2^{-1}/D_s}{1-D^{\sim}\lambda_1^{-1}\lambda_2^{-1}/D_s}\right) \tag{71}$$

$$s_2-\frac{s_3}{\lambda_1\lambda_2^2}=\frac{\mu J_{\lim}}{J_{\lim}-(I_1-3)}\left(\lambda_2-\lambda_1^{-2}\lambda_2^{-3}\right) -\frac{D_sD^{\sim}\lambda_1^{-1}\lambda_2^{-2}}{2\varepsilon}\log\left(\frac{1+D^{\sim}\lambda_1^{-1}\lambda_2^{-1}/D_s}{1-D^{\sim}\lambda_1^{-1}\lambda_2^{-1}/D_s}\right) \tag{72}$$

$$\sigma_1-\sigma_3=\frac{\mu J_{\lim}}{J_{\lim}-(I_1-3)}\left(\lambda_1^2-\lambda_1^{-2}\lambda_2^{-2}\right)-\frac{D_sD^{\sim}\lambda_1^{-1}\lambda_2^{-1}}{2\varepsilon}\log\left(\frac{1+D^{\sim}\lambda_1^{-1}\lambda_2^{-1}/D_s}{1-D^{\sim}\lambda_1^{-1}\lambda_2^{-1}/D_s}\right) \tag{73}$$

$$\sigma_2-\sigma_3=\frac{\mu J_{\lim}}{J_{\lim}-(I_1-3)}\left(\lambda_2^2-\lambda_1^{-2}\lambda_2^{-2}\right)-\frac{D_sD^{\sim}\lambda_1^{-1}\lambda_2^{-1}}{2\varepsilon}\log\left(\frac{1+D^{\sim}\lambda_1^{-1}\lambda_2^{-1}/D_s}{1-D^{\sim}\lambda_1^{-1}\lambda_2^{-1}/D_s}\right) \tag{74}$$

We now investigate an effect of the strain-stiffening on the electromechanical stability and the snap stability of dielectric elastomer undergoing the polarization saturation. Consider the unequal-biaxial condition of dielectric elastomer, $\lambda_1=\lambda_2=\lambda$, we get that

$$\frac{s}{\mu}=\frac{2J_{\lim}}{J_{\lim}-(2\lambda^2+\lambda^{-4}-3)}(\lambda-\lambda^{-5})-\frac{D_s}{\sqrt{C_1\varepsilon}}\frac{D^{\sim}\lambda^{-3}}{\sqrt{C_1\varepsilon}}\times\log\left(\frac{1+\frac{D^{\sim}\lambda^{-2}}{\sqrt{C_1\varepsilon}}\Big/\frac{D_s}{\sqrt{C_1\varepsilon}}}{1-\frac{D^{\sim}\lambda^{-2}}{\sqrt{C_1\varepsilon}}\Big/\frac{D_s}{\sqrt{C_1\varepsilon}}}\right) \tag{75}$$

Let $\frac{s}{\mu}=0$, Eq. (75) is the relation between the nominal electric field and the electrical displacement. Substituting Eq. (68) into Eq. (75), we obtain that

$$\frac{s}{\mu}=\frac{2J_{\lim}}{J_{\lim}-(2\lambda^2+\lambda^{-4}-3)}(\lambda-\lambda^{-5})-2\times\frac{D_s}{\sqrt{C_1\varepsilon}}\left[\frac{\exp\left(2\frac{E^{\sim}}{\sqrt{C_1/\varepsilon}}\lambda^2\Big/\frac{D_s}{\sqrt{C_1\varepsilon}}\right)-1}{\exp\left(2\frac{E^{\sim}}{\sqrt{C_1/\varepsilon}}\lambda^2\Big/\frac{D_s}{\sqrt{C_1\varepsilon}}\right)+1}\right]\frac{E^{\sim}}{\sqrt{C_1/\varepsilon}}\lambda \tag{76}$$

Eq. (76) reveals the relation between the dimensionless nominal electric field and the stretch of the dielectric elastomer undergoing the polarization saturation while considering the effect of the strain-stiffening. The voltage–stretch curve with different extension limit is showed in Fig. 8. When $J_{\lim}$ and $\frac{D_s}{\sqrt{C_1\varepsilon}}$ are both small enough, the nominal electric field increases monotonously with the stretch, avoiding any local maximum value and the dielectric elastomer can therefore approach its stable state. When $\frac{D_s}{\sqrt{C_1\varepsilon}}\to\infty$ and $J_{\lim}$ is a finite value, the nominal electric field attains the local maximum value first. Then, the dielectric elastomer goes through the snap instability with the voltage increasing. When $J_{\lim}\to\infty$ and $\frac{D_s}{\sqrt{C_1\varepsilon}}\to\infty$, the dielectric elastomer undergoes the pull-in electromechanical instability, and the critical breakdown electric field is $E^{\sim}=0.686\sqrt{C_1/\varepsilon}$, the corresponding critical stretch $\lambda_c=1.26$ (Zhao and Suo, 2007a). Therefore, to get a large deformation, the applied voltage should achieve two limits, lower than the breakdown voltage of the material, and approaching the voltage undergoing the snap instability.

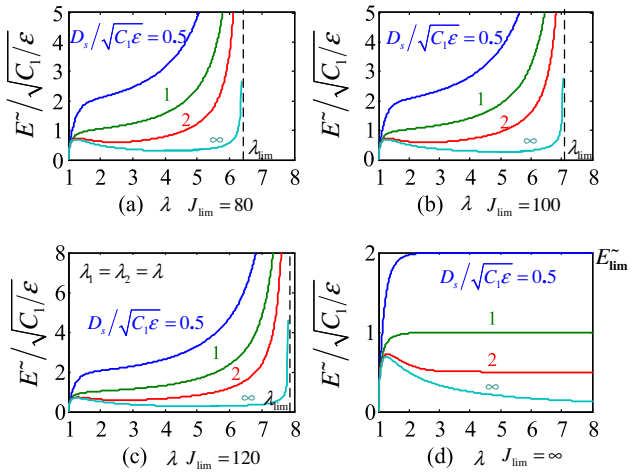

Fig. 8. Consider the effect of the strain-stiffening, nominal electric field, and stretch of dielectric elastomer undergoing polarization saturation under the loading condition $\lambda_1=\lambda_2=\lambda$.

Next, for another case, assuming $\lambda_3=\lambda$, i.e. $\lambda_1=\lambda_2=\lambda^{-1/2}$, applying the similar method of investigation, we express the nominal electric field and the stretch as

$$\frac{s}{\mu}=\frac{J_{\lim}}{J_{\lim}-(2\lambda^{-1}+\lambda^2-3)}(\lambda-\lambda^{-2})+\frac{D_s}{\sqrt{C_1\varepsilon}}\frac{D^{\sim}}{2\sqrt{C_1\varepsilon}}\times\log\left(\frac{1+\frac{D^{\sim}}{\sqrt{C_1\varepsilon}}\lambda\Big/\frac{D_s}{\sqrt{C_1\varepsilon}}}{1-\frac{D^{\sim}}{\sqrt{C_1\varepsilon}}\lambda\Big/\frac{D_s}{\sqrt{C_1\varepsilon}}}\right) \tag{77}$$

Substituting Eq. (68) into Eq. (77), we obtain that

$$\frac{s}{\mu}=\frac{J_{\lim}}{J_{\lim}-(2\lambda^{-1}+\lambda^2-3)}(\lambda-\lambda^{-2})+\frac{D_s}{\sqrt{C_1\varepsilon}}\left[\frac{\exp\left(2\frac{E^{\sim}}{\sqrt{C_1/\varepsilon}}\lambda^{-1}\Big/\frac{D_s}{\sqrt{C_1\varepsilon}}\right)-1}{\exp\left(2\frac{E^{\sim}}{\sqrt{C_1/\varepsilon}}\lambda^{-1}\Big/\frac{D_s}{\sqrt{C_1\varepsilon}}\right)+1}\right]\frac{E^{\sim}}{\sqrt{C_1/\varepsilon}}\lambda^{-2} \tag{78}$$

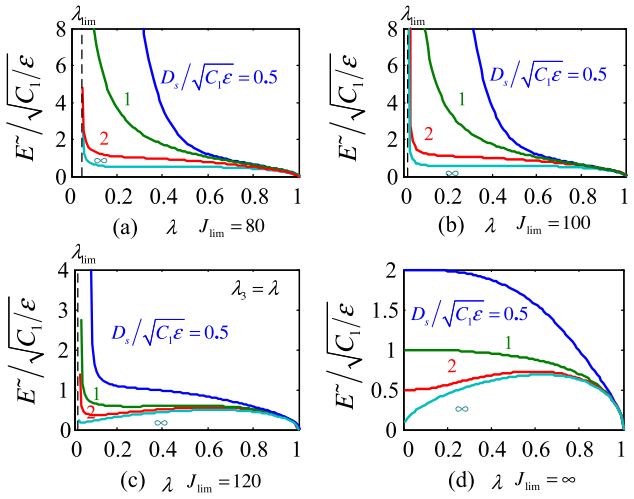

Fig. 9. Consider the effect of the strain-stiffening, nominal electric field, and stretch of dielectric elastomer undergoing polarization saturation under the loading condition $\lambda_3=\lambda$.

Eq. (78) obtains the relation between the voltage and the deformation of dielectric elastomer undergoing polarization saturation while consider the effect of the strain-stiffening under the loading condition as $\lambda_3 = \lambda$.

As shown in Fig. 9, if $J_{\lim}$ and $\frac{D_s}{\sqrt{C_1\varepsilon}}$ are both the finite values, due to the persistent compression of the Maxwell stress induced by electric field, the nominal electric field goes up monotonously. Resulting from its strain-stiffening, the dielectric elastomer can attain its stable state while performing large deformation. When $J_{\lim}$ is a finite value and $\frac{D_s}{\sqrt{C_1\varepsilon}} \to \infty$, the dielectric elastomer will undergo snap instability after the electromechanical instability. On the other hand, when $J_{\lim} \to \infty$ and $\frac{D_s}{\sqrt{C_1\varepsilon}} \to \infty$, the dielectric elastomer merely undergoes the electromechanical instability.

8. Conclusions

When a dielectric elastomer with randomly oriented dipoles is subject to a voltage, the dipoles rotate to align with the electric field. The polarization of the material may be saturated when the voltage is high enough. We develop a thermodynamic model of dielectric elastomers undergoing polarization saturation to account for both nonlinear elastic and dielectric behavior. An analytical solution of the constitutive equation of dielectric elastomer is obtained, and electromechanical instability and snap-through instability are sequentially investigated. Extension and polarization saturation limits of elastomer material can dramatically influence the electromechanical instability and snap-through instability. To get a large deformation, the applied voltage should achieve two conditions, lower than the breakdown voltage of the material, and approaching the voltage undergoing the snap-through instability. For Mooney–Rivlin type dielectric elastomer, as an example, when the material constant ratio k and the polarization saturation parameter $\frac{D_s}{\sqrt{C_1\varepsilon}}$ are both small, the dielectric elastomer undergoing the snap instability. When extension limit $J_{\lim}$ is a finite value and $\frac{D_s}{\sqrt{C_1\varepsilon}} \to \infty$, the dielectric elastomer undergoing the snap instability, capable of giant deformation of actuation. These results are thought by the authors competent in facilitating the design and the manufacture of dielectric elastomer transducers.

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