Extension limit, polarization saturation, and snap-through instability of dielectric elastomers

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Abstract

A dielectric elastomer is capable of large voltage-induced deformation, particularly when the voltage is applied on the verge of snap-through instability. This paper describes a model to show that the snap-through instability is markedly affected by both the extension limit of polymer chains and the polarization saturation of dipoles. The model may guide the search for high-performance dielectric elastomer transducers.

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Subject to a voltage through its thickness, a membrane of a dielectric elastomer reduces thickness and enlarges area (Fig. 1). This electromechanical coupling is being studied intensely for diverse applications, including soft robots, adaptive optics, Braille displays, and electric generators.[1-10] Voltage-induced strains over 100% have been achieved in several ways, by pre-stretching an elastomer,[11] by using an elastomer of interpenetrating networks,[12] by swelling an elastomer with a solvent,[13] and by spraying charge on an electrode-free elastomer.[14]

When the voltage ramps up, the membrane thins down, so that the same voltage will induce an even higher electric field. This positive feedback results in the pull-in instability.[15] The pull-in instability is commonly considered a mode of failure: the voltage causes the elastomer to reduce the thickness drastically, possibly leading to electrical breakdown.[16] In recent years, the pull-in instability has been analyzed within the context of dielectric elastomer transducers.[17-24]

It is recognized recently, however, an elastomer may survive the pull-in instability without electrical breakdown, and be stabilized in a state of a much smaller thickness, resulting in the snap-through instability.[25] This behavior is understood as follows. When the elastomer is subject to mechanical forces (Fig. 2a), on approaching the extension limit, $\lambda_{\lim}$, the elastomer stiffens steeply. When the deformation is caused by voltage rather than the mechanical forces, the voltage-stretch curve is typically not monotonic (Fig. 2b). The voltage attains a local maximum at stretch λ_c, corresponding to the onset of the pull-in instability. As the voltage ramps up further, the membrane snaps, and is stabilized at a stretch close to $\lambda_{\lim}$. Indeed, giant voltage-induced strains well above 100% are possible, so long as the elastomer snaps to a state safe from electrical breakdown.[26,27]

This paper shows that the snap-through instability can also be markedly affected by polarization saturation (Fig. 2c) When a dielectric with randomly oriented dipoles is subject to a voltage, the dipoles rotate to align with the electric field. The polarization of the material may

saturate when the voltage is high enough.[28,29] This nonlinear dielectric behavior will be incorporated in equations of state, and will be shown to modify the voltage-stretch curve.

In the reference state (Fig. 1a), the membrane is subject to neither forces nor voltage, and is of dimensions L_1, L_2 and L_3. In the current state (Fig. 1b), subject to forces P_1, P_2 and P_3, and voltage Φ, the membrane is of dimensions 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 membrane 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 $\Phi\delta Q$. In equilibrium, the combined work equals the increase in the free energy of the membrane:

$$\delta F = P_1\delta l_1 + P_2\delta l_2 + P_3\delta l_3 + \Phi\delta Q . \tag{1}$$

Define the specific Helmholtz free energy by $W = F/(L_1L_2L_3)$, stretches by $\lambda_1 = l_1/L_1$, $\lambda_2 = l_2/L_2$ and $\lambda_3 = l_3/L_3$, stresses by $\sigma_1 = P_1/(l_2l_3)$, $\sigma_2 = P_2/(l_1l_3)$ and $\sigma_3 = P_3/(l_1l_2)$, electric field by $E = \Phi/l_3$, and electric displacement by $D = Q/(l_1l_2)$. The amount of charge on either electrode relates to the electric displacement by $Q = Dl_1l_2$, so that the variation of the charge is

$$\delta Q = Dl_2\delta l_1 + Dl_1\delta l_2 + l_1l_2\delta D . \tag{2}$$

The elastomer is taken to be incompressible—that is, the volume of the material remains unchanged during deformation, $l_1l_2l_3 = L_1L_2L_3$, so that

$$\lambda_1\lambda_2\lambda_3 = 1 . \tag{3}$$

This assumption of incompressibility places a constraint among the three stretches. We regard λ_1 and λ_2 as independent variables, so that $\lambda_3 = \lambda_1^{-1}\lambda_2^{-1}$, and $\delta\lambda_3 = -\lambda_1^{-2}\lambda_2^{-1}\delta\lambda_1 - \lambda_1^{-1}\lambda_2^{-2}\delta\lambda_2$. Divide both sides of (1) by the volume of the membrane, $L_1L_2L_3$, and using (2) and (3), we obtain that

$$\delta W = (\sigma_1 - \sigma_3 + DE)\lambda_1^{-1}\delta\lambda_1 + (\sigma_1 - \sigma_3 + DE)\lambda_2^{-1}\delta\lambda_2 + E\delta D . \tag{4}$$

For an incompressible dielectric, the condition of equilibrium (4) holds for arbitrary and independent variations $\delta\lambda_1$, $\delta\lambda_2$ and δD.

As a material model, the specific free energy is taken to be a function of the three independent variables, $W(\lambda_1,\lambda_2,D)$, so that (4) is equivalent to the following equations:

$$\sigma_1-\sigma_3=\lambda_1\frac{\partial W(\lambda_1,\lambda_2,D)}{\partial\lambda_1}-ED, \tag{5}$$

$$\sigma_2-\sigma_3=\lambda_2\frac{\partial W(\lambda_1,\lambda_2,D)}{\partial\lambda_2}-ED, \tag{6}$$

$$E=\frac{\partial W(\lambda_1,\lambda_2,D)}{\partial D}. \tag{7}$$

Electromechanical coupling may be classified into two kinds: the geometric coupling characterized by (2), and the material coupling characterized by the function $W(\lambda_1,\lambda_2,D)$.

We next focus on a model known as ideal dielectric elastomers.[25] An elastomer is a three-dimensional network of long and flexible polymer chains, held together by crosslinks. Each polymer chain consists of such a large number of monomers that the crosslinks affect polarization of the monomers negligibly. That is, the elastomer can polarize nearly as freely as a polymer melt. 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 relation between the electric field is a function of the electric displacement independent of deformation:

$$E=f(D). \tag{8}$$

Holding λ_1 and λ_2 fixed, and integrating (4) with respect to *D*, we obtain that

$$W(\lambda_1,\lambda_2,D)=W_s(\lambda_1,\lambda_2)+\int_0^D f(D)dD. \tag{9}$$

The constant of integration, $W_s(\lambda_1,\lambda_2)$, is the Helmholtz free energy associated with the stretching of the elastomer. Equations (5) and (6) become

$$\sigma_1-\sigma_3=\lambda_1\frac{\partial W_s(\lambda_1,\lambda_2)}{\partial\lambda_1}-ED, \tag{10}$$

$$\sigma_2 - \sigma_3 = \lambda_2 \frac{\partial W_s(\lambda_1, \lambda_2)}{\partial \lambda_2} - ED . \tag{11}$$

The four equations, (3), (8), (10) and (11) constitute the equations of state for an incompressible, ideal dielectric elastomer, provided the functions $f(D)$ and $W_s(\lambda_1, \lambda_2)$ are given.

When the model of ideal dielectric elastomers was proposed,[25] the elastomer was taken to be a linear dielectric, $E = D/\varepsilon$, where ε is the permittivity. To study the effect of polarization saturation, here we assume that the elastomer is a nonlinear dielectric, characterized by the function[29]

$$D = D_s \tanh(\varepsilon E / D_s), \tag{12}$$

where D_s is the saturated electric displacement. When electric field is low, $\varepsilon E / D_s << 1$, (12) recovers the linear dielectric behavior, $E = D/\varepsilon$. When the electric field is high, $\varepsilon E / D_s >> 1$, (12) becomes $D = D_s$.

The free energy due to the stretching of the elastomer, $W_s(\lambda_1, \lambda_2)$, may be selected from a large menu of well-tested functions in the theory of rubber elasticity. To account for the extension limit, here we adopt the Gent model [30]

$$W_s = -\frac{\mu J_{\lim}}{2} \log\left(1 - \frac{\lambda_1^2 + \lambda_2^2 + \lambda_3^2 - 3}{J_{\lim}}\right), \tag{13}$$

where μ is the shear modulus, and $J_{\lim}$ is a constant characterizing the extension limit. The stretches are restricted as $0 \le (\lambda_1^2 + \lambda_2^2 + \lambda_3^2 - 3)/J_{\lim} < 1$. When $(\lambda_1^2 + \lambda_2^2 + \lambda_3^2 - 3)/J_{\lim} \to 0$, the Gent model recovers the neo-Hookean model, $W_s = (\mu/2)(\lambda_1^2 + \lambda_2^2 + \lambda_3^2 - 3)$. When $(\lambda_1^2 + \lambda_2^2 + \lambda_3^2 - 3)/J_{\lim} \to 1$, the free energy (13) diverges, and the elastomer approaches the extension limit.

The theory is now used to study a membrane of a dielectric elastomer subject to fixed forces $P_1 = P_2 = P$ and $P_3 = 0$, as well as voltage Φ. Write the three stretches as $\lambda_1 = \lambda_2 = \lambda$ and $\lambda_3 = \lambda^{-2}$. Specializing (10), we obtain that

$$\frac{P}{\mu L_2 L_3} = \frac{\lambda - \lambda^{-5}}{1 - \left(2\lambda^2 + \lambda^{-4} - 3\right)/J_{\lim}} - \frac{D_s \lambda \Phi}{\mu L_3} \tanh\left(\frac{\lambda^2 \Phi \varepsilon}{D_s L_3}\right). \tag{14}$$

We may normalize the voltage as $\Phi/\left(L_3\sqrt{\mu/\varepsilon}\right)$, and the force as $P/\left(\mu L_2 L_3\right)$. The extension limit of polymer chains is represented by the dimensionless parameter $J_{\lim}$, and the polarization saturation of dipoles is represented by the dimensionless parameter $D_s/\sqrt{\mu\varepsilon}$.

Fig. 3 plots the voltage-stretch relation at several levels of the applied equal-biaxial forces. When the forces are small, the voltage-stretch curve exhibits a local maximum. As the voltage ramps up, the membrane undergoes the snap-through instability. When the applied forces are large, the local maximum disappears, leading to a monotonic voltage-stretch curve. Before the voltage is applied, the applied forces pull the membrane toward the extension limit, so that the steep stiffening removes the local maximum of the voltage-stretch curve. This mechanism may explain why mechanical forces enhance voltage-induced deformation.[11]

The effect of polarization saturation is appreciated by inspecting the equations of state, (10) and (11). 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, an effect that tends to stabilize the elastomer. This effect is illustrated in Fig. 4a, where the voltage-stretch curves are plotted for elastomers without the extension limit ($J_{\lim} = \infty$) and subject to no applied forces. The local maximum is eliminated when $D_s/\sqrt{\mu\varepsilon}$ is small. Setting $P = 0$ and $J_{\lim} = 0$ in (14), we note that the voltage approaches a limiting value $\Phi_{\lim} = \mu L_3 / D_s$ as $\lambda \to \infty$.

Fig. 4b plots the voltage-stretch curves for elastomers with $J_{\lim} = 100$ and several values of $D_s / \sqrt{\mu\varepsilon}$. Such a diagram suggests various routs to achieve large voltage-induced deformation. For instance, a large value of permittivity both reduces the level of the voltage needed for actuation and stabilizes the voltage-stretch curve. A large shear modulus increases the level of the voltage needed for actuation, but helps to stabilize the voltage-stretch curve. Of course, to achieve large deformation by applying voltage on the verge of the snap-through instability, one must ensure that the voltage will not cause electrical breakdown.[26,27]

In summary, we develop a model of electromechanical coupling to account for nonlinear elastic and dielectric behavior. Both extension limit and polarization saturation can significantly affect the snap-through instability. The model may aid the search for high-performance dielectric elastomer transducers.

This work is supported by NSF (CMMI-0800161) and by MURI (W911NF-09-1-0476). The stay of Liwu Liu and Bo Li at Harvard University was supported by visiting-student scholarships from the Chinese Government. Li also acknowledges National Natural Science Foundation of China (Grant No.10972174) for the support of his research on dielectric elastomers.

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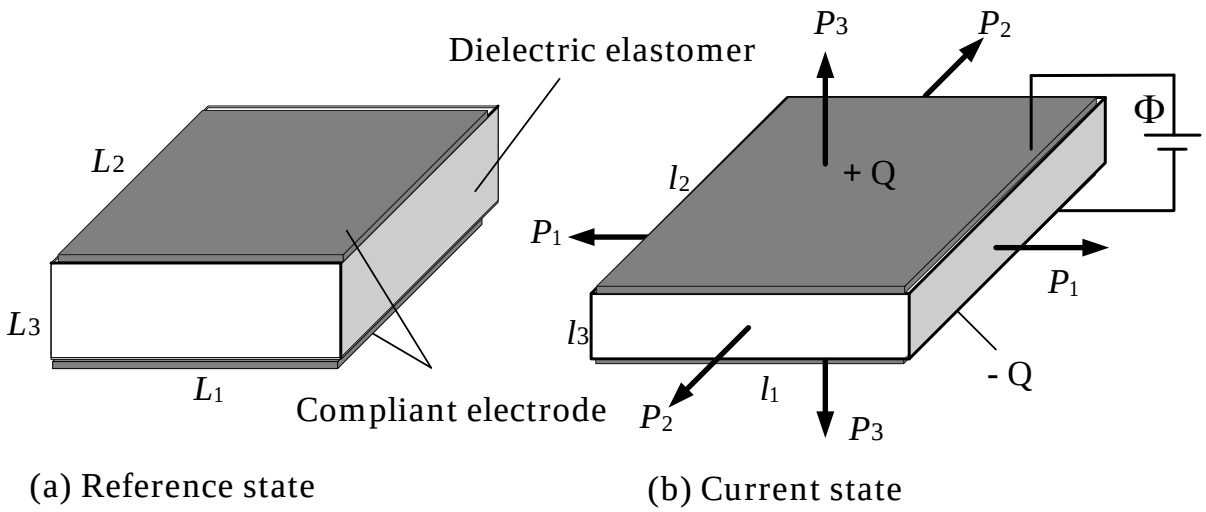

FIG. 1. A membrane of a dielectric elastomer is sandwiched between two compliant electrodes. (a) In the reference state, the dielectric is subject to neither forces nor voltage. (b) In the current state, subject to forces and voltage, the membrane deforms, and charge flows from one electrode to the other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

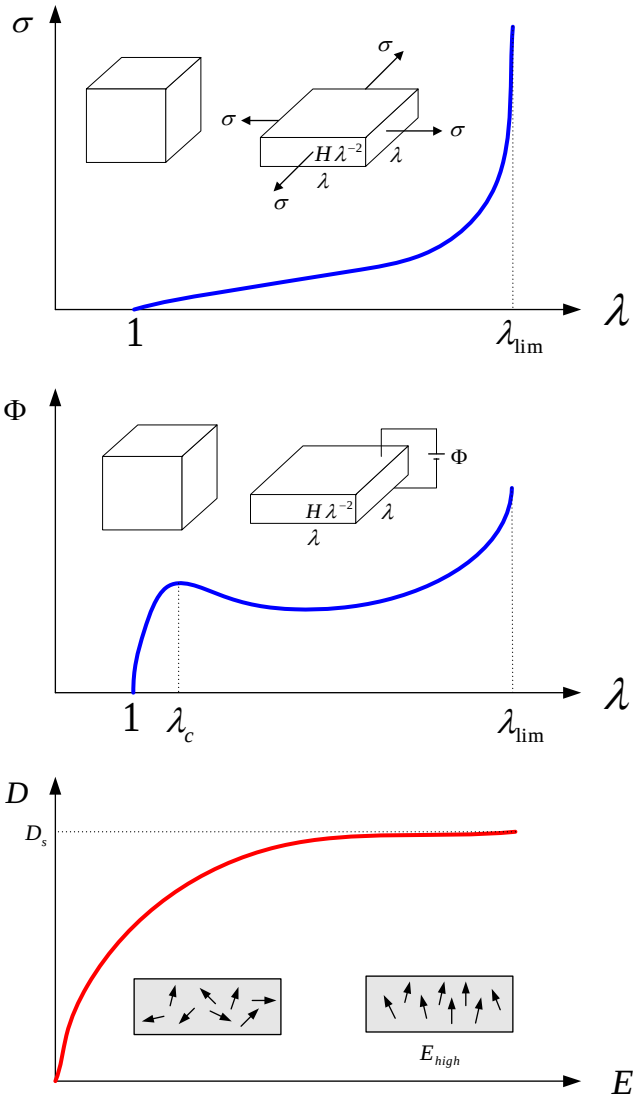

FIG. 2. (a) Stress-stretch curve of a membrane of an elastomer under biaxial stresses. The curve stiffens steeply upon approaching the extension limit. (b) Voltage-stretch curve of a membrane of a dielectric elastomer is typically not monotonic. (c) For a dielectric contains randomly oriented dipoles, as the electric field increases, the dipoles rotate to align with the electric field, and the electric displacement saturates.

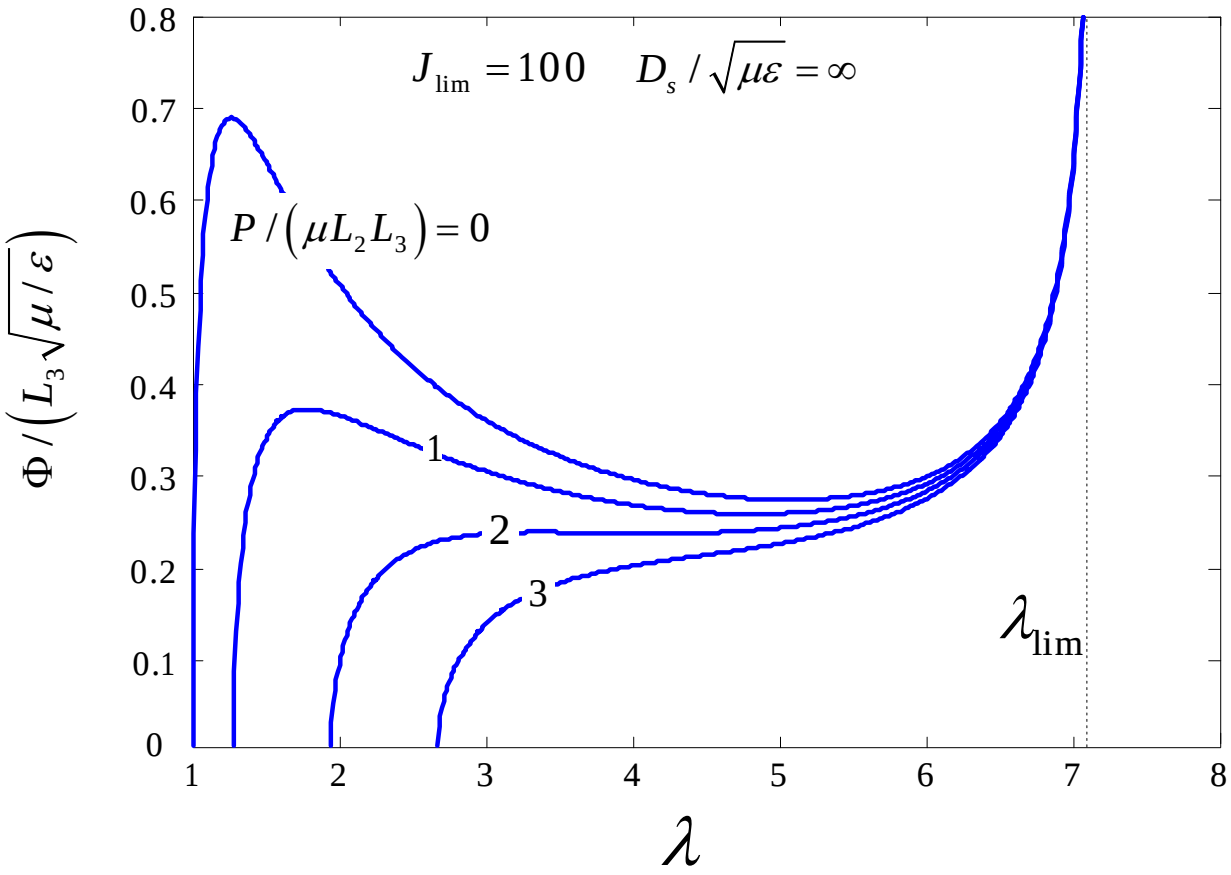

FIG. 3. Voltage-stretch curves of a dielectric elastomer subject to equal-biaxial forces.

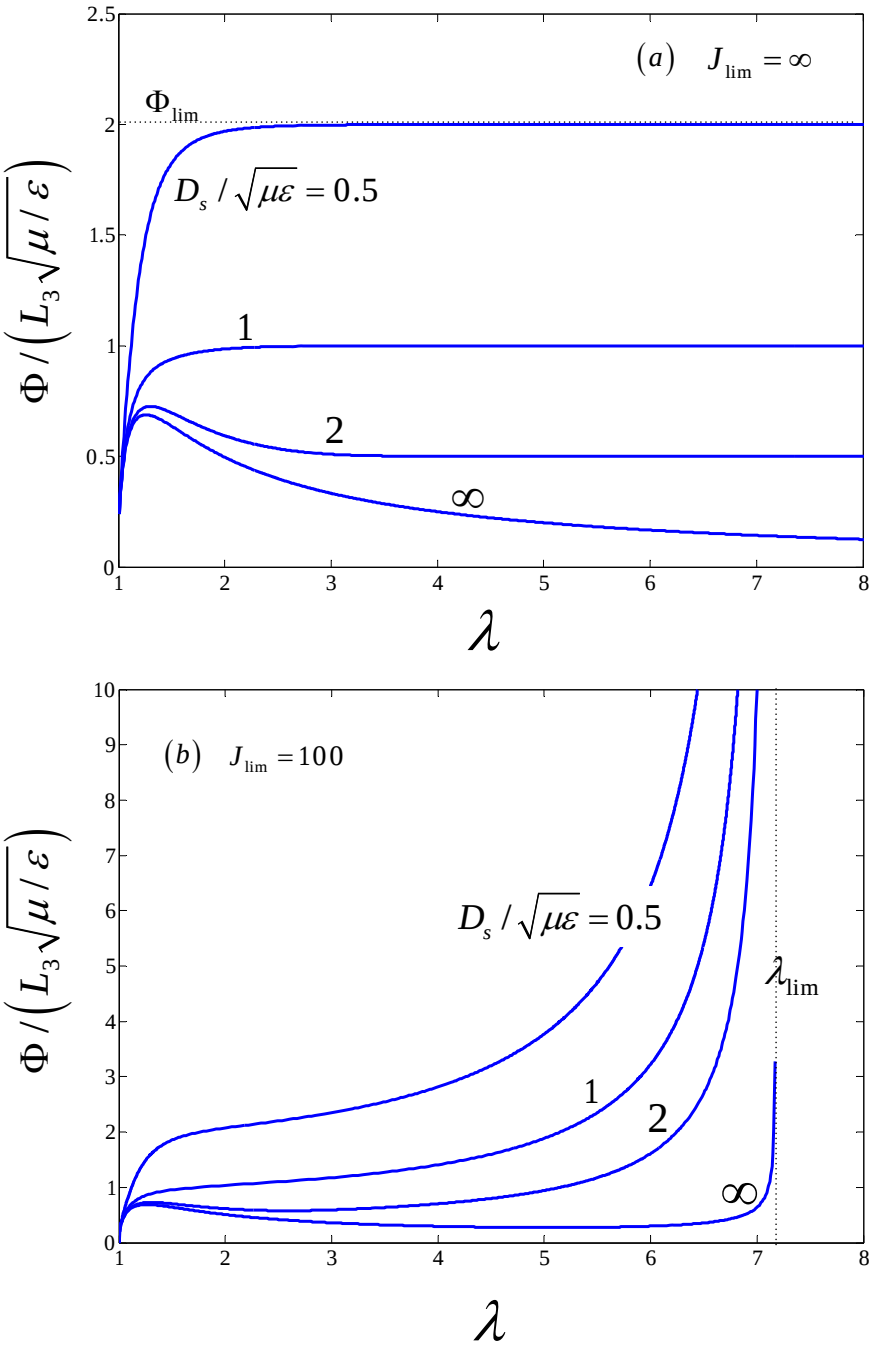

FIG. 4. (a) Voltage-stretch curves for a dielectric elastomer without extension limit ($J_{\lim}=\infty$), but with several levels of polarization saturation. (b) Voltage-stretch curves for a dielectric elastomer with an extension limit ($J_{\lim}=100$), and with several levels of polarization saturation.