True method to analyze electromechanical stability of dielectric elastomers

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A layer of dielectric elastomer can be voltage actuated to behave as actuators, but needs to avoid the electromechanical instability of excessively thin down accompanied with electric breakdown. We develop a true method to analyze the electromechanical stability of dielectric elastomers by adopting the positive definiteness of the true tangential modulus matrix, and demonstrate that the previous method is only valid for the special case of zero prestress. Our new method is applicable for arbitrary prestress cases including zero prestress, with predictions consistent with available experimental measurements. Our theoretical results demonstrate the significant effects of prestress on critical voltage and critical actuation stretches.

The large deformation of dielectric elastomers induced by voltage enables their extensive applications as actuators[1-4]. However, electromechanical instability is often encountered to disable the actuator, which occurs when a threshold voltage is reached, causing excessively thin down of thickness[5]. Theoretically, the electromechanical stability of dielectric elastomers has been extensively investigated[6-11]. However, these theoretical studies are all established on the positive definiteness of the nominal Hessian matrix, which is only valid for the particular case of zero prestress[12-14].

The stability of an electromechanical system at certain load point should be evaluated using its current state in Eulerian coordinates, not the nominal state in Lagrangian coordinates. To judge the occurrence of electromechanical stability, one need to use the true Hessian matrix in

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Eulerian coordinates rather than the nominal Hessian matrix in Lagrangian coordinates. Only when there exists neither stress nor deformation, the nominal Hessian matrix would be identical as the true Hessian matrix. Consequently, the positive definiteness of the nominal Hessian matrix as employed in existing studies dictates only the stability of the initial load route.

This paper attempts to develop a true method for analyzing the electromechanical stability of dielectric elastomers. We present the true electromechanical stability in general case, applicable not only to the initial load route point but also to arbitrary load points. We consider the electromechanical system as shown in Fig. 1, where the layer made of dielectric elastomer is sandwiched between two compliant electrodes and subjected to the combined loading of voltage Φ and biaxial stresses (σ_1, σ_2). The elastomer deforms from its initial (undeformed) state (L_1, L_2, L_3) to the current state (l_1, l_2, l_3) with principal stretches (λ_1, λ_2, λ_3). The elastomer is assumed incompressible, so that $\lambda_1\lambda_2\lambda_3 = 1$.

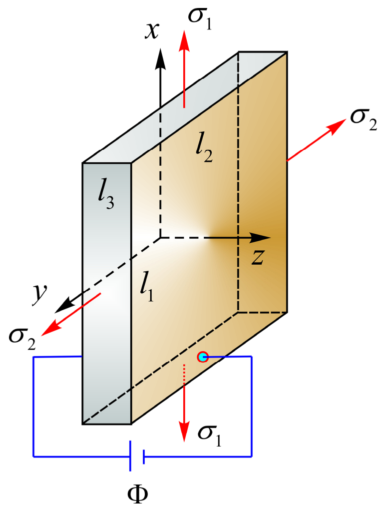


FIG. 1 (Color online) A layer of dielectric elastomer sandwiched between two compliant electrodes is subjected to combined voltage and biaxial stresses. The layer deforms from initial dimensions (L_1, L_2, L_3) to current dimensions (l_1, l_2, l_3) with principal stretches (λ_1, λ_2, λ_3).

With voltage Φ exerted on the two electrodes, the electric field in current state is $E = \Phi\lambda_1\lambda_2/L_3$ and the corresponding charge on either electrode is $Q = L_1L_2\varepsilon\Phi\lambda_1^2\lambda_2^2/L_3$. The elastomer layer needs to bear the Maxwell stress ($-\varepsilon E^2/2$, $-\varepsilon E^2/2$, $\varepsilon E^2/2$), which is the same as stress state ($-\varepsilon E^2$, $-\varepsilon E^2$, 0) since the elastomer is incompressible. For ideal elastomers, the permittivity ε is unaffected by deformation. The free energy function of dielectric elastomers can be expressed using the neo-Hookean model, as[15]

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

where μ is the small-strain shear modulus. The true stresses stemming from elastic deformation can be expressed as $\sigma_{ij}=\frac{F_{iK}}{\det\left(\mathbf{F}\right)}\frac{\partial W_s}{\partial F_{jK}}$, **F** being deformation gradient. These true stresses and the Maxwell stresses are balanced by the Cauchy stresses, as:

$$\sigma_1=\mu\left(\lambda_1^2-\lambda_1^{-2}\lambda_2^{-2}\right)-\varepsilon\left(\frac{\Phi}{L_3}\right)^2\lambda_1^2\lambda_2^2 \tag{2}$$

$$\sigma_2=\mu\left(\lambda_2^2-\lambda_1^{-2}\lambda_2^{-2}\right)-\varepsilon\left(\frac{\Phi}{L_3}\right)^2\lambda_1^2\lambda_2^2 \tag{3}$$

$$Q=\frac{L_1L_2\varepsilon\Phi}{L_3}\lambda_1^2\lambda_2^2 \tag{4}$$

where the principal Cauchy stress σ_i is work conjugate to principal strain ε_i, which is directly related to stretch as $\varepsilon_i=\ln\lambda_i$, while the electric charge Q is work conjugate to voltage Φ. The above constitutive equations describe the behavior of the electromechanical system in true space (*i.e.*, Eulerian coordinates).

A stable electromechanical system requires minimization of its total energy, *i.e.*, the strong ellipticity condition should be satisfied in a finitely deforming elastic body, as:

$$H_{ijkl}b_ib_kN_jN_l>0,\ H_{ijkl}=\frac{\partial^2 W}{\partial F_{ji}\partial F_{lk}} \tag{5}$$

for all arbitrary $\mathbf{b}\otimes\mathbf{N}\neq 0$. Here, H_{ijkl} is the component of the fourth-order elasticity tensor, which is the nominal Hessian matrix. Incorporating the prestresses and $\mathbf{n}=\mathbf{F}^{-\mathrm{T}}\mathbf{N}$, one can rewrite this condition as[12-14]

$$\left(C_{ijkl}+\sigma_{jl}\delta_{ik}\right)b_ib_kn_jn_l>0,\ C_{ijkl}=\frac{1}{J}F_{i\alpha}F_{k\beta}\frac{\partial^2 W}{\partial F_{j\alpha}\partial F_{l\beta}} \tag{6}$$

where σ_{jl} is the Cauchy stress.

Condition (7) can also be stated in terms of the positive definiteness of acoustic tensor $Q_{ik}\left(\mathbf{n}\right)=\left(C_{ijkl}+\sigma_{jl}\delta_{ik}\right)n_jn_l$. Stability of the electromechanical system is then governed by the positive definiteness of true tangential stiffness matrix $c_{ijkl} = \left(C_{ijkl} + \sigma_{jl}\delta_{ik} \right)$. Only for the case of zero stress (*i.e.*, $\boldsymbol{\sigma} = 0$), the strong ellipticity condition can be degraded to the positive definiteness of nominal tangential stiffness matrix (*i.e.*, nominal Hessian matrix), which is called the Born stability[12,14]. As the Born stability based on nominal Hessian matrix is only valid for the limiting case of zero stress, we use the true tangential stiffness matrix to analyze the electromechanical stability of the electromechanical system for non-zero stress cases.

The electromechanical system of Fig. 1 depends on three independent variables, so that its generalized constitutive relation is $\begin{bmatrix} \sigma_1 & \sigma_2 & Q \end{bmatrix}^T = \begin{bmatrix} c_{MN} \end{bmatrix} \begin{bmatrix} \varepsilon_1 & \varepsilon_2 & \Phi \end{bmatrix}^T$, where $c_{MN} = c_{ijkl}$ if $M = ij$ and $N = kl$ are noted. The true tangential stiffness matrix can thence be obtained as:

$$c_{MN} = \begin{bmatrix} \lambda_1 \dfrac{\partial \sigma_1}{\partial \lambda_1} & \lambda_2 \dfrac{\partial \sigma_1}{\partial \lambda_2} & \dfrac{\partial \sigma_1}{\partial \Phi} \\ \lambda_1 \dfrac{\partial \sigma_2}{\partial \lambda_1} & \lambda_2 \dfrac{\partial \sigma_2}{\partial \lambda_2} & \dfrac{\partial \sigma_2}{\partial \Phi} \\ \lambda_1 \dfrac{\partial Q}{\partial \lambda_1} & \lambda_2 \dfrac{\partial Q}{\partial \lambda_2} & \dfrac{\partial Q}{\partial \Phi} \end{bmatrix} \tag{8}$$

Making use of the generalized constitutive equations of (2)-(4), we rewrite the true tangential modulus matrix (*i.e.*, true Hessian matrix) as:

$$c_{MN} = \begin{bmatrix} 2\mu\left(\lambda_1^2 + \lambda_1^{-2}\lambda_2^{-2}\right) - 2\varepsilon\left(\dfrac{\Phi}{L_3}\right)^2 \lambda_1^2\lambda_2^2 & 2\mu\lambda_1^{-2}\lambda_2^{-2} - 2\varepsilon\left(\dfrac{\Phi}{L_3}\right)^2 \lambda_1^2\lambda_2^2 & -2\varepsilon\left(\dfrac{\Phi}{L_3^2}\right)\lambda_1^2\lambda_2^2 \\ 2\mu\lambda_1^{-2}\lambda_2^{-2} - 2\varepsilon\left(\dfrac{\Phi}{L_3}\right)^2 \lambda_1^2\lambda_2^2 & 2\mu\left(\lambda_2^2 + \lambda_1^{-2}\lambda_2^{-2}\right) - 2\varepsilon\left(\dfrac{\Phi}{L_3}\right)^2 \lambda_1^2\lambda_2^2 & -2\varepsilon\left(\dfrac{\Phi}{L_3^2}\right)\lambda_1^2\lambda_2^2 \\ 2\dfrac{L_1L_2\varepsilon\Phi}{L_3}\lambda_1^2\lambda_2^2 & 2\dfrac{L_1L_2\varepsilon\Phi}{L_3}\lambda_1^2\lambda_2^2 & \dfrac{L_1L_2\varepsilon}{L_3}\lambda_1^2\lambda_2^2 \end{bmatrix} \tag{9}$$

which, at equilibrium state, should possess the property of positive definiteness.

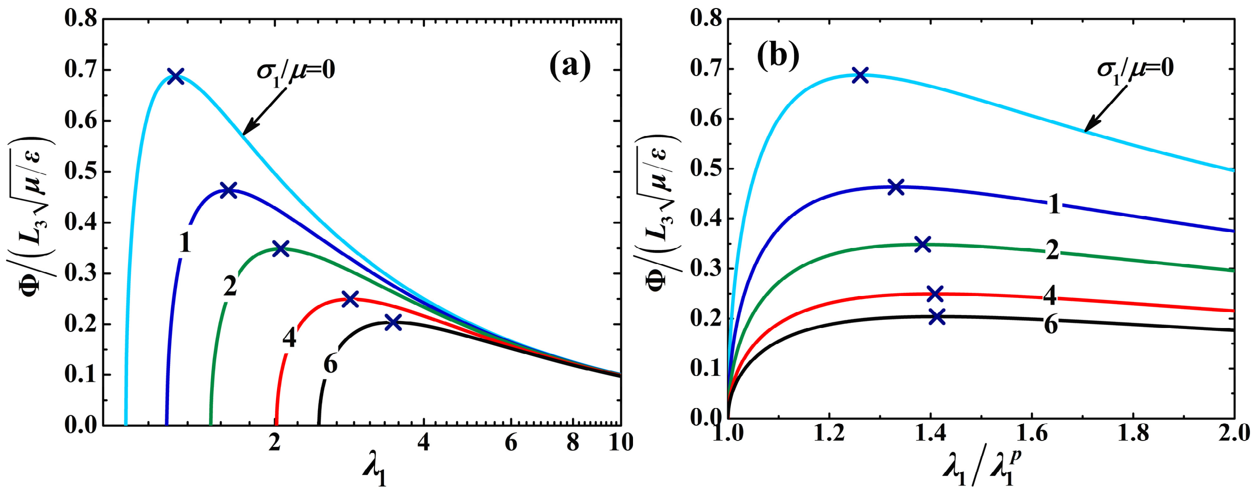
In the presence of prescribed prestresses σ_1 and σ_2, the true tangential modulus matrix is positive definite and the electromechanical system is stable when the exerted voltage is sufficiently small. When the voltage reaches a critical value Φ^c, the true tangential modulus matrix ceases to be definite positive, yielding $\det(\mathbf{c}) = 0$. All the parameters associated with the critical condition can be obtained by solving the constitutive equations and $\det(\mathbf{c}) = 0$.

We consider the general case of unequal biaxial prestresses, *i.e.*, $\sigma_1 \neq \sigma_2$; the stretches thus induced should also be unequal, $\lambda_1 \neq \lambda_2$. Using the constitutive equations, we express the normalized voltage as a function of stretches, as:

$$\frac{\Phi}{L_3\sqrt{\mu/\varepsilon}} = \lambda_1^{-1}\lambda_2^{-1}\sqrt{\left(\lambda_1^2 - \lambda_1^{-2}\lambda_2^{-2}\right) - \frac{\sigma_1}{\mu}},\ \lambda_2 = \sqrt{\lambda_1^2 - \frac{\sigma_1 - \sigma_2}{\mu}} \tag{10}$$

Because the electromechanical stability is voltage-controlled, the above voltage versus stretch relation should be taken into account to consider this stability. For arbitrarily prescribed prestresses, Eq. (10) describes the electromechanical performance of the considered system (Fig. 1). For example, the case of equal-biaxial prestresses is described by $\sigma_1 = \sigma_2$ while the case of uniaxial prestress is described by $\sigma_1 \neq 0$ and $\sigma_2 = 0$.

As previously mentioned, electromechanical stability should be evaluated in the current state. At selected levels of equal-biaxial prestresses, we plot in Fig. 2 the voltage-actuated responses of dielectric elastomer layer. For each prestress level, the voltage versus stretch curve exhibits a peak (marked by cross), which signifies the onset of pull-in instability. The left side of the curve corresponds to positive definiteness of the true tangential modulus matrix, whereas its right side is related to non-positive definiteness of the matrix. At the critical onset point, we have $\det(\mathbf{c}) = 0$. Increasing the prestress reduces the actuation voltage needed to realize the same actuation deformation, which agrees well with experimental observations[1]. Also, as the prestress is increased, the critical actuation stretch increases.



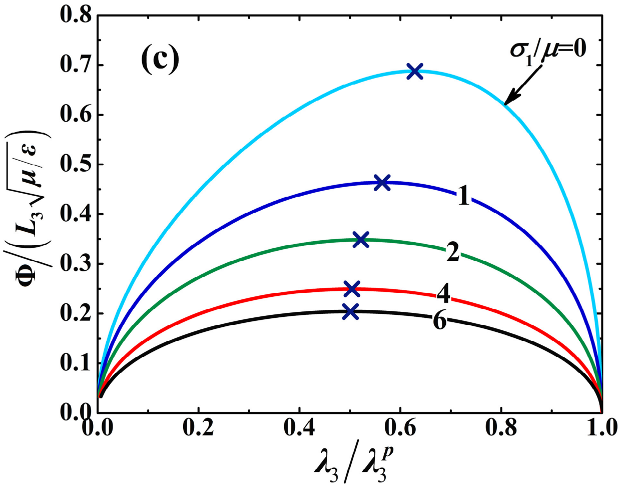


FIG. 2 (Color online) Electromechanical responses of dielectric elastomer actuator to selected equal-biaxial pre-stresses: (a) voltage versus in-plane stretch; (b) voltage versus in-plane actuation stretch; (c) voltage versus out-of-plane actuation stretch. Critical onsets of electromechanical instability are marked by crosses.

In the absence of prestress, it has been experimentally reported that the maximum thickness strain is approximately 40%[2]. Under such conditions, we maximize Eq. (10) to obtain the critical stretch of $\lambda^c \approx 1.2610$, which corresponds to a thickness strain of ~37%. This result is in good agreement with experimental data. The critical voltage is $\Phi^c/\left(L_3\sqrt{\mu/\varepsilon}\right) \approx 0.6874$, resulting in a critical electric field of $\Phi^c/L_3 \approx 10^8$ V/m for typical values of $\mu = 10^6\,\mathrm{N/m^2}$ and $\varepsilon = 4\times10^{-11}$ F/m for dielectric elastomers. This is on the same order of magnitude of the breakdown field that has previously been reported[5]. Similar results were obtained by Zhao and Suo[6], because their Born-type electromechanical stability theory is valid for the special case of zero prestress[14,16,17].

Figures 3(a)-(d) present the effects of unequal-biaxial prestresses on critical voltage, critical in-plane actuation stretches, and critical out-of-plane actuation stretch, with $\sigma_2/\sigma_1 = 0$ representing uniaxial prestress and $\sigma_2/\sigma_1 = 1$ denoting equal-biaxial prestresses. In Fig. 3(a), the critical voltage is seen to decrease with increasing prestress, implying a lower voltage is required to induce large deformation when a suitable prestress is exerted. With fixed σ_1, increasing σ_2 tends to increase the critical in-plane actuation stretch λ_1^c/λ_1^p, decrease the other critical in-plane actuation stretch λ_2^c/λ_2^p, as well as decrease the out-of-plane actuation stretch λ_3^c/λ_3^p.

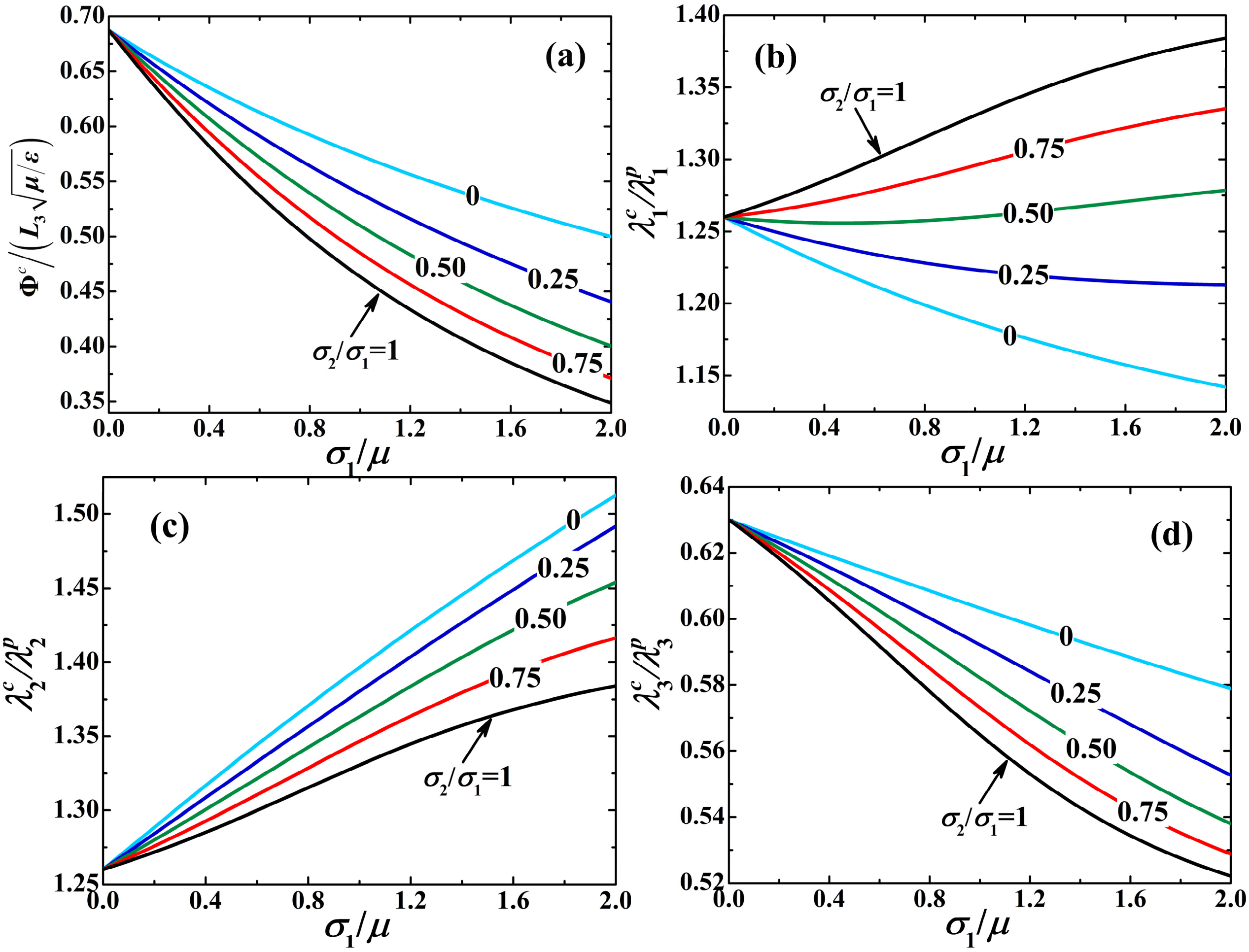


FIG. 3 (Color online) Effect of unequal-biaxial prestresses on (a) critical voltage, (b) and (c) critical in-plane actuation stretches, and (d) critical out-of-plane actuation stretch

In summary, we propose a true method for analyzing the electromechanical stability of dielectric elastomers by generalizing the stability criteria to non-zero prestress cases. We demonstrate that the positive definiteness of the commonly adopted nominal Hessian matrix is only valid for the special case of zero prestress. Our theory is applicable for arbitrary prestresses, thus it can be favorably degraded to consider the zero prestress case. We reveal that the presence of prestress affects significantly the critical voltage and actuation stretches. The neo-Hookean model for dielectric elastomer deformation is applied, which can be extended to consider other types of material. This work should be helpful for actual design of voltage actuated devices.

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