

Electromechanical instability in semicrystalline polymers

Xuanhe Zhao, Zhigang Suo ^a

School of Engineering and Applied Sciences, Harvard University, Cambridge, MA 02138

Abstract

When a layer of a semicrystalline polymer is subject to a tensile force in its plane and a voltage through its thickness, the deformation of the layer is initially homogeneous, but then localizes. The electromechanical instability sets in when the force and the voltage reach critical conditions. The critical conditions are determined in this paper, and are related to two special cases: the Considère condition for the necking instability, and the Stark-Garton condition for the pull-in instability. The general critical conditions show that a tensile force can markedly reduce the critical voltage.

Keywords: polymers, electromechanical instability, Maxwell stress.

^a email: suo@seas.harvard.edu

Semicrystalline polymers are widely used as dielectrics in cables, capacitors and actuators.¹⁻⁴ Such applications require that dielectrics sustain high voltages. A major mode of failure caused by the high voltages is the pull-in instability.⁵⁻⁹ Subject to a voltage, a layer of a polymer thins down, and the electric field increases. The positive feedback may lead to the pull-in instability upon reaching a critical voltage. In various applications, the layer is often subject to a combination of a voltage and a tensile force. Experiments have shown that the tensile force can substantially reduce the critical voltage.¹⁰⁻¹⁶

This paper calculates the critical conditions for the electromechanical instability in a semicrystalline polymer subject to combined electric voltage and mechanical force. The general critical conditions recover the Stark-Garton condition for the pull-in instability⁵, and the Considère condition for the necking instability.^{5,17} Furthermore, the general critical conditions show that a tensile force can indeed reduce the critical voltage.

To focus on essential ideas, consider a model illustrated in Fig 1. A layer of a polymer, thickness H and sides $L \times L$ in the undeformed state, is sandwiched between two compliant electrodes. The polymer is then subject to a voltage Φ through the thickness and a biaxial force P in the plane. The electromechanical instability typically sets in at a large deformation, where the polymer changes its shape more substantially than changes its volume. Following a common practice, we assume that the polymer is incompressible. Consequently, when the thickness reduces to λH , the two sides stretch to $L/\sqrt{\lambda}$.

As illustrated in Fig. 2, the biaxial force P causes a biaxial stress of magnitude

$$\sigma = \frac{P}{HL\sqrt{\lambda}}. \quad (1)$$

Because the polymer is taken to be incompressible, superimposing a state of hydrostatic

compression to the polymer does not change the state of deformation in the polymer. Consequently, the state of deformation in the polymer subject to the biaxial tension in the plane is the same as the state deformation in the polymer subject to a uniaxial compression through the thickness.

The voltage causes an electric field:

$$E = \frac{\Phi}{H\lambda}. \quad (2)$$

This electric field also causes the polymer to deform (Fig. 2). When the deformation is large, the polymer chains change configuration, but the electric polarization may be negligibly affected by the large deformation. As a commonly adopted idealization, the dielectric behavior of the polymer is taken to be liquid-like, such that the permittivity ε is a constant unaffected by the deformation.¹⁸ For an incompressible dielectric with a constant permittivity, the effect of the electric field on deformation can be described by a compressive Maxwell stress, εE^2 .¹⁹

When the polymer is subject to a combination of biaxial force P and voltage Φ , the state of deformation in the polymer is the same as that in the polymer subject to a uniaxial compressive stress of magnitude $\sigma + \varepsilon E^2$. The natural strain in the thickness direction is $\ln \lambda$. We will describe the mechanical behavior of the polymer by using a power-law model, where the uniaxial stress is taken to scale with the natural strain to some power.^{9,17} Consequently, under the combined voltage and force, the effective stress relates to the stretch as

$$\sigma + \varepsilon E^2 = K(-\ln \lambda)^N, \quad (3)$$

where N and K are parameters used to fit experimentally recorded stress-strain relation.

The parameter N measures how steeply the polymer strain-hardens. Stark and Garton assumed the linear elastic behavior, $N = 1$.⁵ However, the stress-strain relations for semicrystalline polymers significantly deviate from the linear elastic behavior, with more realistic values being $N = 0.1-0.6$. The parameter K measures the yield strength of the polymer, and is much smaller than the elastic modulus of the polymer. Consequently, the Stark-Garton model in its original form significantly overestimates the critical voltage. The modified model using the power-law adequately describes the experimentally measured critical voltage.⁹

In an analysis of stability, it is essential to specify the variables independently controlled in experiments. In this paper, we assume that experiments independently vary the force P and the voltage Φ . In terms of the independent variables, (3) becomes

$$\frac{P}{HL\sqrt{\lambda}} + \varepsilon \left(\frac{\Phi}{H\lambda} \right)^2 = K(-\ln \lambda)^N. \quad (4)$$

The equation embodies two competing trends as the polymer deforms: material hardening and geometric softening. Material hardening results from the monotonically increasing stress-strain curve, and is described by the power-law model $K(-\ln \lambda)^N$. Geometric softening results from the thinning of the layer, and is described by the stretch appearing in the denominators of the two terms on the left-hand side of (4).

Figure 3 (a) represents (4) by plotting the normalized force as a function of the stretch at several fixed values of the voltage. The overall shape of the curves reflects the competition between material hardening and geometric softening. When deformation is small, $\lambda \approx 1$, material hardening prevails, and the force must increase to cause further deformation. When deformation is large, $\lambda \ll 1$, geometric softening prevails, and the

force decreases to cause further deformation. For a fixed value of the voltage, as the polymer deforms, the force attains a maximum, corresponding to the critical condition for the instability to set in. For the special case that the polymer is subject to a biaxial tensile force P in the absence of voltage, $\Phi = 0$, Eq. (4) reduces to $P = KHL(-\ln \lambda)^N \lambda^{1/2}$. Maximizing the function $P(\lambda)$, we obtain the critical stretch $\lambda_c = \exp(-2N)$ and the critical stress $\sigma_c = K(2N)^N$. This special case recovers the Considère condition for the necking instability.¹⁷

Alternatively, Fig. 3(b) represents (4) by plotting the normalized voltage as a function of the stretch at several fixed values of the force. The overall shape of the curves also reflects the competition between material hardening and geometric softening. For the special case that the polymer is subject to the voltage Φ in the absence of the force, $P = 0$, Eq. (4) reduces to $\Phi = H\sqrt{K/\varepsilon}(-\ln \lambda)^{N/2} \lambda^2$. Maximizing the function $\Phi(\lambda)$, we obtain the critical stretch $\lambda_c = \exp(-N/2)$ and the critical electric field $E_c = \sqrt{K/\varepsilon}(N/2)^{N/2}$. This special case recovers the modified Stark-Garton condition for the pull-in instability.⁹

Observe that the necking instability sets in at the critical stretch $\lambda_c = \exp(-2N)$, and the pull-in instability sets in at the critical stretch $\lambda_c = \exp(-N/2)$. That is, the necking instability requires more deformation than the pull-in instability. This difference is understood by inspecting the two terms on the left-hand side of (4). The reduction in the thickness of the layer from H to λH causes geometric softening, but λ enters the two terms by different powers: the geometric softening impacts more on the voltage than on the force.

In general, when P is fixed at any value, the voltage is a function $\Phi(\lambda)$ as

determined by (4). Setting $d\Phi(\lambda)/d\lambda = 0$ at a fixed value of P , we obtain that

$$\frac{P}{HL\sqrt{\lambda}} + 4\varepsilon\left(\frac{\Phi}{H\lambda}\right)^2 = 2NK(-\ln \lambda)^{N-1}. \quad (5)$$

This equation is also obtained by regarding (4) as a function $P(\lambda)$ when Φ is fixed at any value, and then setting $dP(\lambda)/d\lambda = 0$.

Eqs. (4) and (5) together determine the critical conditions for the electromechanical instability in a power-law dielectric subject to combined tensile force and voltage. Under the critical conditions, once any one of the three parameters, λ , P and Φ , are prescribed, the other two can be determined by simultaneously solving (4) and (5). Fig. 4 plots the critical conditions for the electromechanical instability on the plane with the force and the voltage as coordinates. The critical voltage is small when the tensile force is large. This trend agrees with experimental observations.¹⁰⁻¹⁶

Recall that the stress and the electric field are related to the force and the voltage, (1) and (2). From (4) and (5) we can solve the stress and the electric field in terms of λ , namely,

$$\frac{\sigma}{K} = \frac{4}{3}(-\ln \lambda)^N - \frac{2N}{3}(-\ln \lambda)^{N-1}, \quad (6)$$

$$\frac{\varepsilon E^2}{K} = -\frac{1}{3}(-\ln \lambda)^N + \frac{2N}{3}(-\ln \lambda)^{N-1}. \quad (7)$$

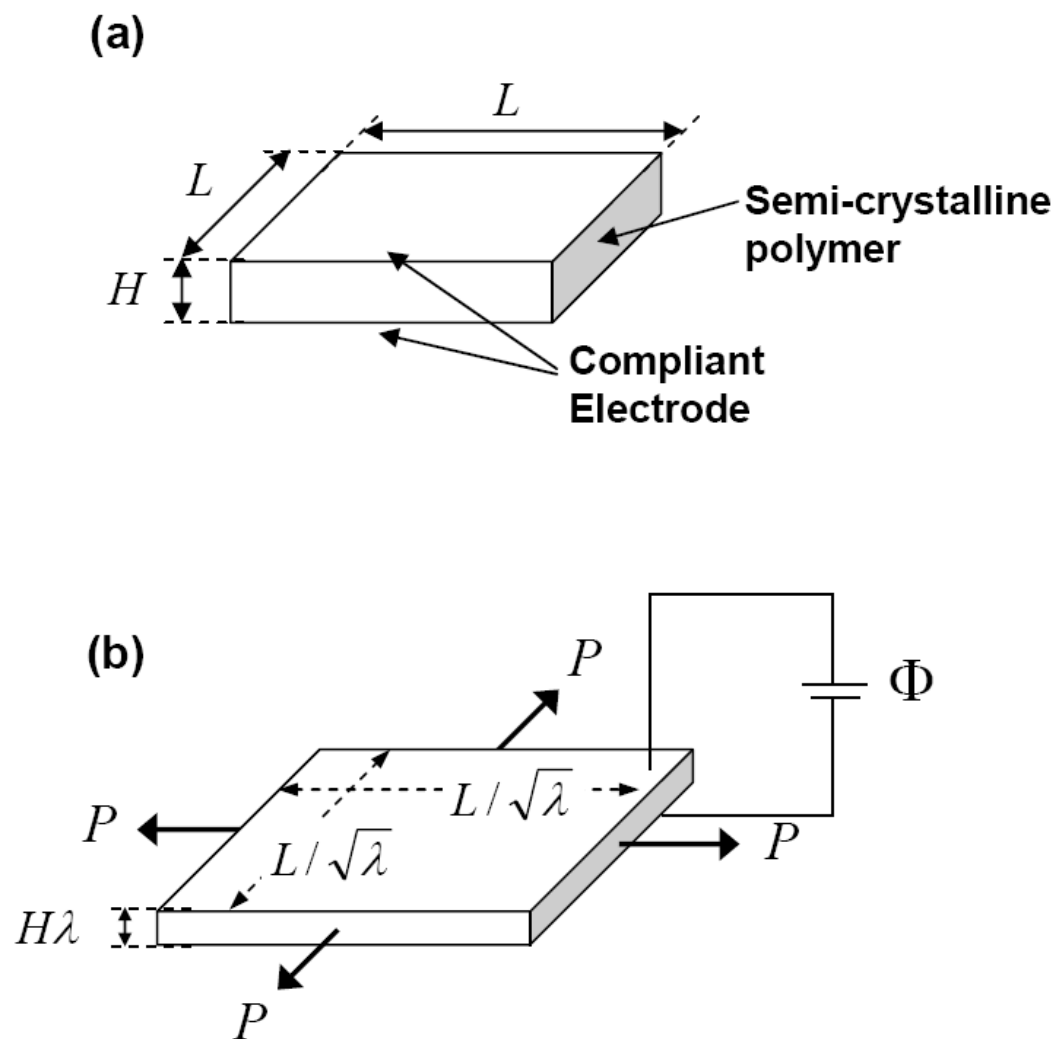
In terms of the parameters σ, E, λ , these two equations express the critical conditions under which the electromechanical instability sets in. The critical conditions (6) and (7) generalize both the Considère condition and the Stark-Garton condition. These equations are plotted in Fig. 5. The electromechanical instability precedes electrical breakdown when the critical electric field determined in Fig. 5 is lower than the electric breakdown strength of

the polymer.

We have calculated the critical conditions for the electromechanical instability of a semicrystalline polymer subject to a combination of mechanical force and electric voltage. To focus on essential ideas, we have restricted to the biaxial force and power-law material model. The method, however, can be extended to other loading conditions²⁰ and other material models²¹. While the model predicts the same trend as reported in the experimental literature,¹⁰⁻¹⁶ we hope that more complete experimental data will become available to ascertain the model.

This work is supported by the National Science Foundation through a grant on Soft Active Materials.

Figures



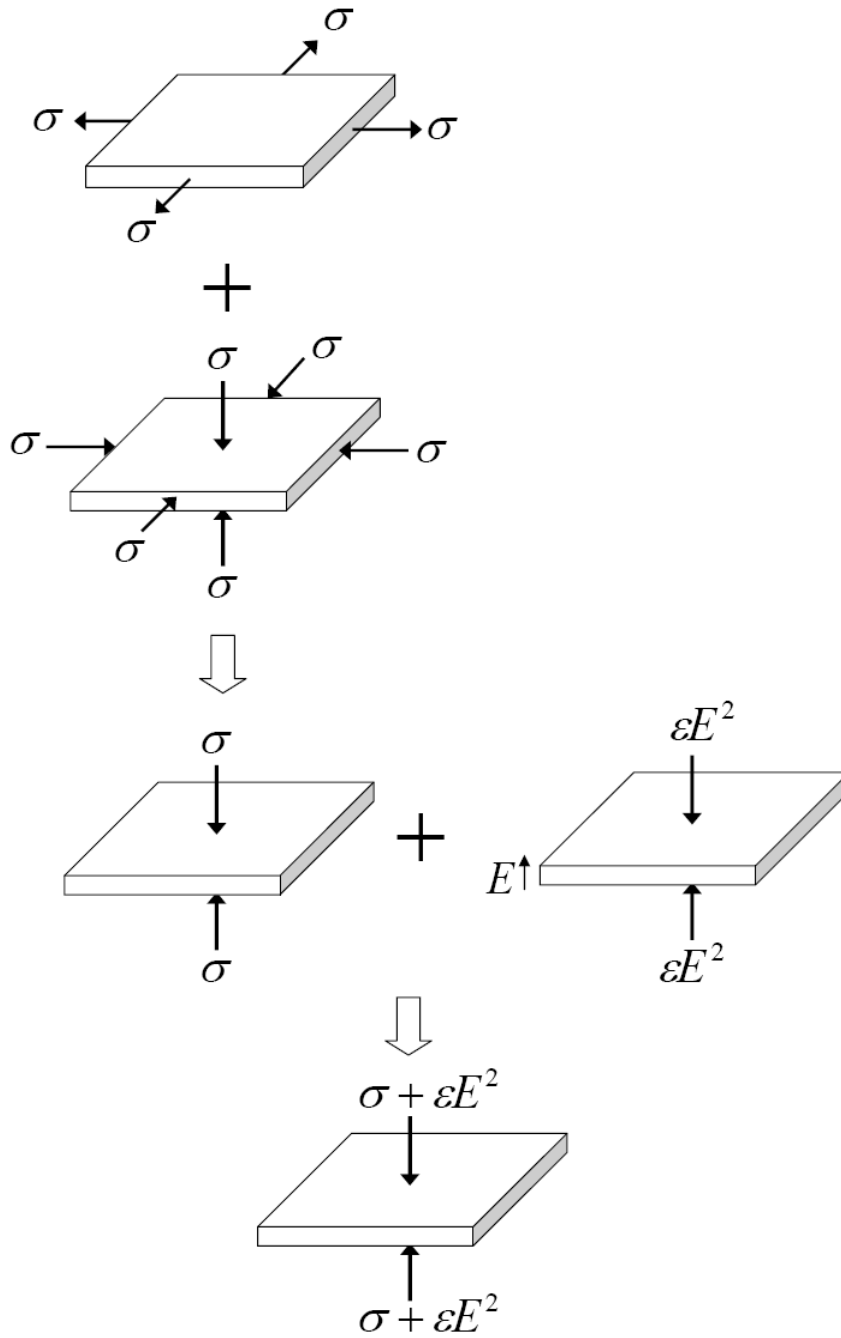


FIG. 2. For an incompressible dielectric, the state of strain caused by a biaxial force and a voltage is the same as that caused by a uniaxial compressive stress of magnitude $\sigma + \epsilon E^2$.

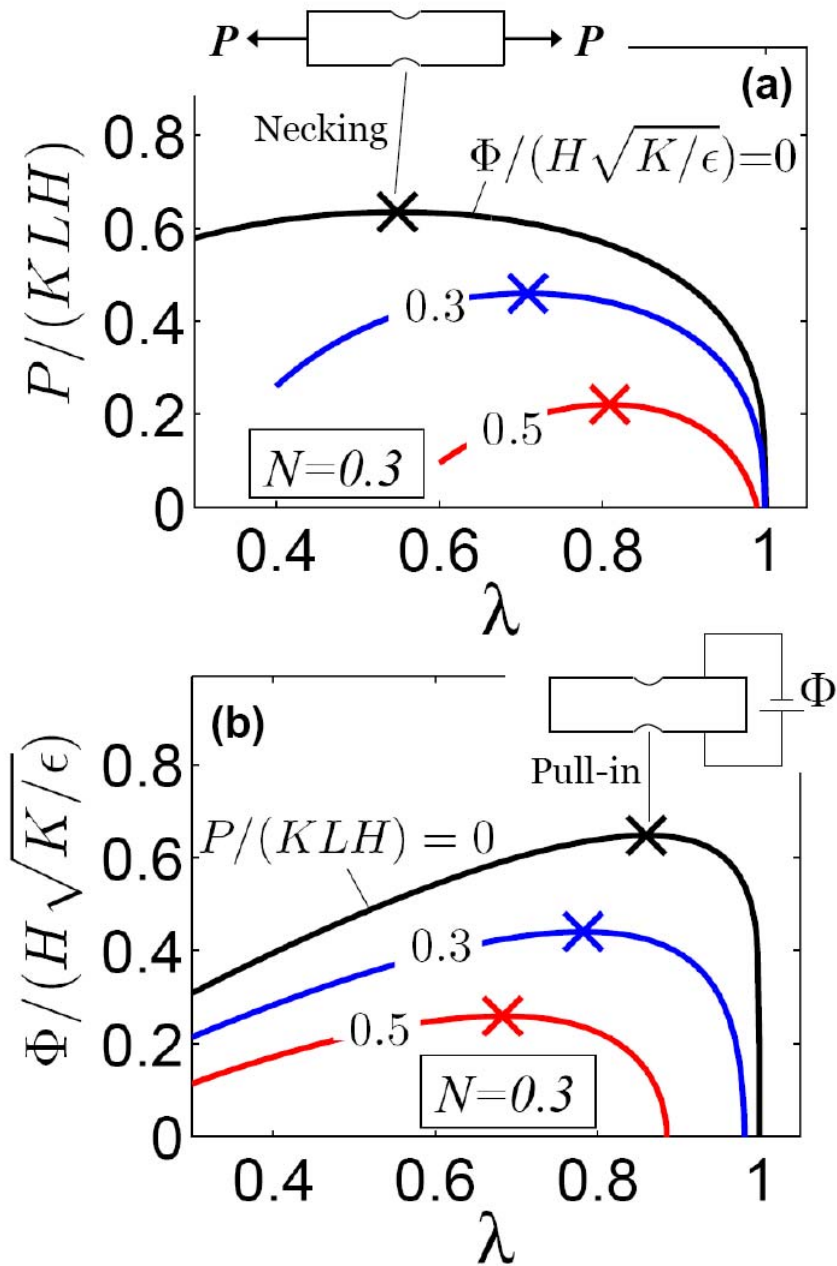


Fig. 3. (a) The force as a function of the stretch for several fixed values of the voltage. (b) The voltage as a function of the stretch for several fixed values of the force.

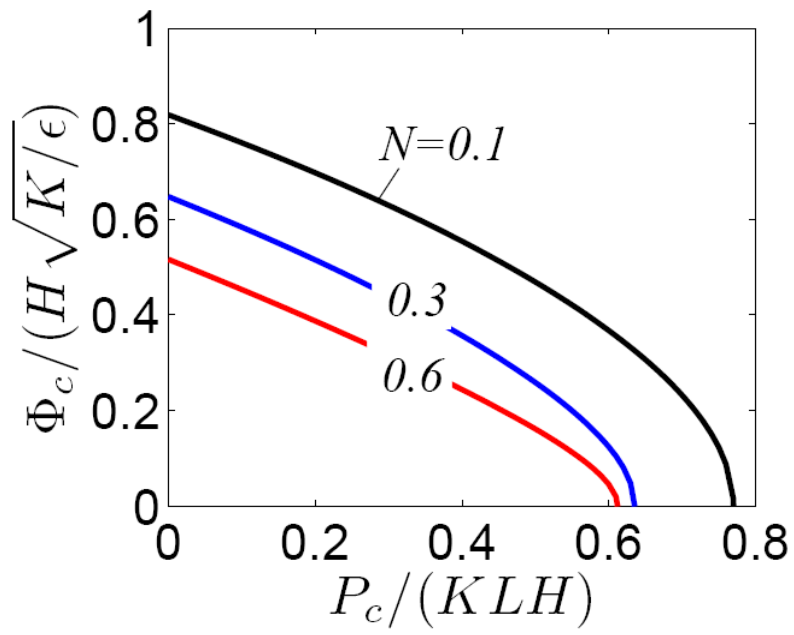


Fig. 4. On the plane with the force and the voltage as coordinates, the critical conditions for the electromechanical instability are plotted for several values of the hardening exponent.

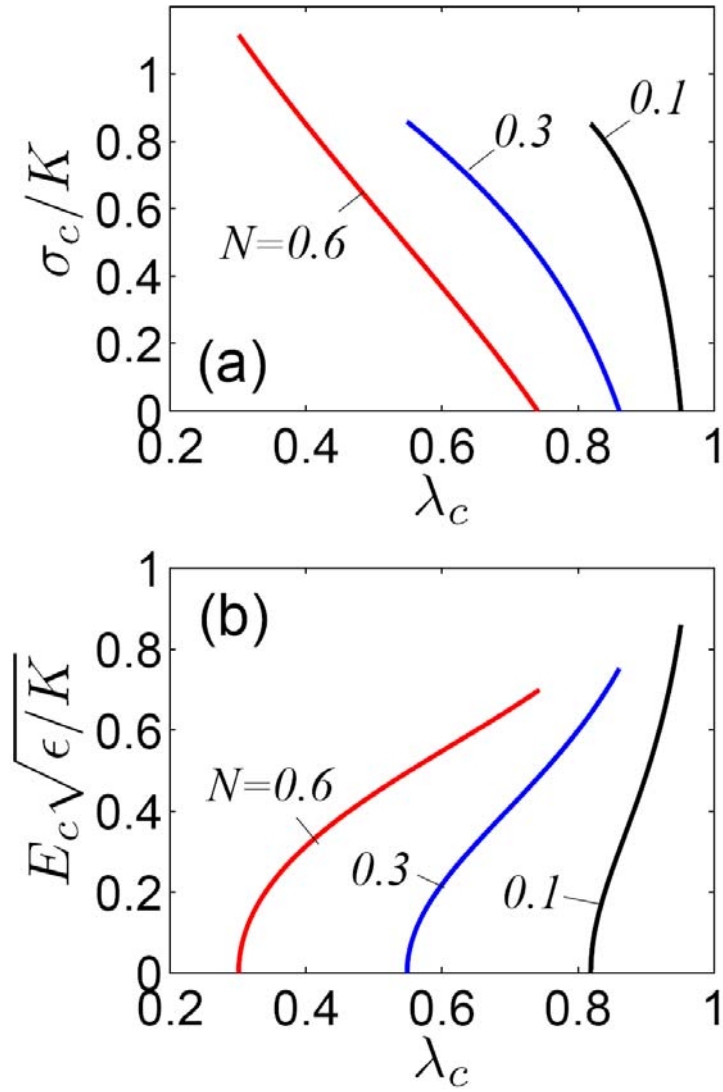


Fig. 5. (a) The critical stress as a function of the critical stretch for several values of the hardening exponent. (b) The critical electric field as a function of the critical stretch for several values of the hardening exponent.

Reference

- 1 T. Takahashi, H. Ohtsuka, H. Takehana, and T. Niwa, *Ieee Transactions on Power Apparatus and Systems* **104**, 1945-1950 (1985).
- 2 M. Rabuffi and G. Picci, *Ieee Transactions on Plasma Science* **30**, 1939-1942 (2002).
- 3 Q. M. Zhang, V. Bharti, and X. Zhao, *Science* **280**, 2101-2104 (1998).
- 4 Q. M. Zhang, H. F. Li, M. Poh, F. Xia, Z. Y. Cheng, H. S. Xu, and C. Huang, *Nature* **419**, 284-287 (2002).
- 5 K. H. Stark and C. G. Garton, *Nature* **176**, 1225-1226 (1955).
- 6 J. Blok and D. G. LeGrand, *Journal of Applied Physics* **40**, 288 (1969).
- 7 H. R. Zeller and W. R. Schneider, *Journal of Applied Physics* **56**, 455 (1984).
- 8 J. C. Fothergill, *IEEE fiansactions on Electrical Insulation* **26**, 1124 (1991).
- 9 X. Zhou, X. H. Zhao, Z. G. Suo, C. Zou, J. Runt, S. Liu, S. H. Zhang, and Q. M. Zhang, *Applied Physics Letters* **94**, 3 (2009).
- 10 S. Mita and K. Yahagi, *Japanese Journal of Applied Physics* **14**, 197-201 (1975).
- 11 C. H. Park, T. Kaneko, M. Hara, and M. Akazaki, *Ieee Transactions on Electrical Insulation* **17**, 234-240 (1982).
- 12 P. Guerin, Hoangthegiam, B. Ai, P. Destruel, L. Deschamps, and J. Perret, *Journal of Applied Physics* **57**, 4805-4807 (1985).
- 13 E. Ilstad and S. T. Hagen, in *Proc. IEEE Int. Symp. Electrical Insulation; Vol. 89* (1992), p. 525-531.
- 14 E. David, J. L. Parpal, and J. P. Crine, in *Proc. IEEE Int. Symp. Electrical Insulation* (1994), p. 170-173.
- 15 N. Amyot, E. David, S. Y. Lee, and I. H. Lee, *Ieee Transactions on Dielectrics and Electrical Insulation* **9**, 458-466 (2002).
- 16 F. H. Zheng, Y. W. Zhang, C. Xiao, J. F. Xia, and Z. L. An, *Ieee Transactions on Dielectrics and Electrical Insulation* **15**, 965-973 (2008).
- 17 J. W. Hutchinson and K. W. Neale, in *Proceedings of the IUTAM Symposium on Finite Elasticity*, Martinus Nijhoff/The Hague, Boston/London, 1979, p. 237.
- 18 X. H. Zhao, W. Hong, and Z. G. Suo, *Physical Review B* **76**, 134113 (2007).
- 19 R. Pelrine, R. Kornbluh, Q. B. Pei, and J. Joseph, *Science* **287**, 836-839 (2000).
- 20 X. H. Zhao and Z. G. Suo, *Applied Physics Letters* **91**, 061921 (2007).
- 21 X. H. Zhao and Z. G. Suo, *Journal of Applied Physics* **104**, 123530 (2008).