A THEORETICAL STUDY OF THE HYPERELASTICITY OF ELECTRO-GELS

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A theoretical study of the hyperelasticity of electro-gels

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The continuum theory of electro-elasticity is used in order to describe the large deformations observed in gels endowed with electric properties, when they are placed in electric fields. The analytical solution of the properly constructed boundary value problem agrees quantitatively with available experimental data.

Keywords: hyperelasticity; electro-gels; biomimetic materials; MEMS; artificial muscles

1. Introduction

Solid metal alloys and plastics have found many applications in medical practise as biomaterials in prosthetics, artificial hearts, valves, stents, etc. They include, among others, piezoelectric and magnetostrictive materials as bone growth stimulators (Polk 1996) and thermosensitive shape memory alloys in (endo-)orthodontics, vascular circulation (stents, valves) or as surgical tools (Lagoudas 2000). There is an increasing need nowadays for new materials in many medical applications, with biomimetic functionalities in eye, ear, muscles and valve operations, drug targeting etc., that combine low cost, high efficiency and minor side effects. Hydrogels with thermo-electromagnetic or chemical properties are good candidates, since they combine better biocompatibility (hydrophilic) along with more efficient actuation or sensory mechanism (large deformation). Experiments on pH sensitive (Qing et al. 2001) and thermosensitive hydrogels (Lindlein et al. 2000) have been performed recently. In a series of experiments Zrinyi et. al. (2000a-b), Filipcsei et. al. (2000) and Fehér et. al. (2001) prepared electric and magnetic gels and tested their response to electromagnetic fields, confirming their capability to mimic muscle contraction (artificial muscles). Our previous research on the physical principles of micromagnetic and magnetoelastic phenomena in solids (Voltairas et. al. 1999a - c; 2000a - d) directed our motivation to examine possible applications of magnetic carriers in medicine. In a recent paper (Voltairas et. al. 2000e) we proposed a phenomenological model in order to estimate if a ferrofluid internal tamponade, along with a semi-solid magnetic silicon band (magnetic scleral buckle) can contribute to retinal re-attachment in retinal detachment surgery. A condition for the elastic stability of the ferrofluid was derived. In an attempt to quantify conditions for magnetic

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drug targeting and delivery, we developed a hydrodynamic model and compared its predictions with available experimental data (Voltairas et. al. 2002).

The present work constitutes the first part in a project aimed at examining under what conditions electro- or ferrogels can replace non-recoverable injured muscles. In this paper we focus on the control of the experimental apparatus that measures the deformation of the electro-gels as a function of the external electric stimulus (Filipcsei et. al. 2000 and Fehér et. al. 2001), by proposing a suitable theory for the phenomenon. The general continuum theory of nonlinear electro-elasticity is summarized in §2. In §3 we introduce the basic model assumptions for: the geometry of the problem, the displacement field (plane deformations), the electric field (collinearity, uniformity) and the material properties (isotropy, homogeneity, etc.). The constitutive laws are derived and comparison with experimental data is performed. Finally in §4 we conclude with the limitations of the model, the possible improvements and future generalizations to magnetic field sensitive gels (ferrogels) and similar biophysical phenomena.

2. General equations and notation

When a polymer hydrogel containing electrically polarized micro- or nanoparticles, from hereafter called electro-qel, is exposed to an electric field deforms. The observed large strains can be measured experimentally as a function of the applied electric field. The process is usually reversible for suitable applied fields and gel densities, thus the elasticity of the electro-gel is analogous to the mechanical deformations of hyperelastic materials. Hence, we will prefer the phenomenological description based on the continuum theory of nonlinear electro-elasticity. In similar problems, with large deformations in human soft tissues, the continuum approach proved successful, for modeling purposes (Holzapfel et. al. 2000). Electro-elasticity theory is well documented (see Eringen 1962; Eringen & Maugin 1989), including also magnetic and thermal effects. Hereafter, bold and double bold characters will denote vector and tensor fields, respectively. We consider that the electro-gel deforms as a continuous body, which in the reference (undeformed) configuration occupies a region $\Omega \subset S$, of the whole space S, inside the closed surface $\partial \Omega$. The material points are identified by their position vectors X in Ω , with Cartesian coordinates X_A (A = 1, 2, 3). After the deformation the electro-gel occupies the region Ω_d , and a point originally denoted by X is deformed to the position x with coordinates x_i (i =1, 2, 3). The deformation gradient $\mathbb{F} \equiv \nabla_X \otimes x(X)$, has Cartesian components given by $F_{iA} = \partial x_i/\partial X_A$. For a uniform static applied electric field, the equilibrium problem is described by the partial differential equations:

$$\nabla_X \cdot S + f_e = 0,$$
 in Ω (2.1)

$$\nabla \cdot D = q_f \, \delta(x), \quad \text{in } \mathcal{S}$$
 (2.2)

$$\nabla \times E = 0$$
, in S (2.3)

and the jump conditions

$$[S^T + J(F^{-1}T_m)^T]N = 0, \text{ on } \partial\Omega$$
 (2.4)

$$[D] \cdot n = \sigma_s$$
, on $\partial \Omega_d$ (2.5)

$$t \cdot ||E|| = 0$$
, on $\partial \Omega_d$. (2.6)

in the reference configuration. Here

$$f_e = \nabla_X \cdot (J \mathbb{F}^{-1} \mathbb{T}_m) = J \nabla \cdot \mathbb{T}_m = J (P \cdot \nabla) E + q_f E,$$
 (2.7)

is the electric body force with

$$T_m = D \otimes E - \varepsilon_0 \frac{E^2}{2} I,$$
 (2.8)

the Maxwell stress tensor, due to Einstein and Laub (see Toupin 1956),

$$S = J \mathbb{F}^{-1} \mathbb{T}, \qquad (2.9)$$

is the nominal stress tensor (Ogden 1997),

$$D = \varepsilon_0 E + P, \qquad (2.10)$$

is the displacement vector, $\delta(x) = 1$ for $x \in \Omega_d$ and $\delta(x) = 0$ for $x \in S - \Omega_d$, $[A] \equiv A_{\text{out}} - A_{\text{in}}$, E is the electric field, P is the polarization vector per unit volume, q_f and σ_s are the free body and surface charges, respectively, $J = \text{det } \mathbb{F}$, \mathbb{T} is the Cauchy stress tensor, ∇_X and ∇ are the gradient vector operators in the reference and present configurations, respectively, ε_0 is the electric permeability of vacuum, I is the identity tensor, \otimes and \cdot denote tensor and inner product, respectively, t is the unit tangential vector on $\partial \Omega_d$ and N and n are the outward unit vectors on $\partial\Omega$ and $\partial\Omega_d$, respectively, with

$$n = J(\mathbb{F}^{-1})^T N. \qquad (2.11)$$

Gravitational effects are neglected due to strong electric forces f_e . The above formulation of the electro-elastic boundary value problem is due to Toupin (1956). The only difference is that we have included the external applied field E_0 in the Maxwell stress tensor,

$$E = E_P + E_0,$$
 (2.12)

with E_P the depolarizing field, which is identical to Toupin's Maxwell-self field $E_{\rm MS}$. The notion of the Lorentz local field $E_{\rm L}=-E$ is frequently used. If we decompose the electric field E in tangential and normal components on $\partial\Omega$, neglect mechanical surface tractions and make use of the definitions (2.7-12) and the jump conditions (2.5-6), the balance of electro-mechanical surface tractions (2.4) reduces to:

$$S^T N = \frac{P_n^2 - \sigma_s^2}{2\varepsilon_0} n + \sigma_s E_0, \quad P_n \equiv P \cdot n, \text{ on } \partial\Omega.$$
 (2.13)

The boundary value problem (2.1-6) is derived from an energy variational principle (Toupin 1956). The derivation is not unique. It depends on the form of the electrostatic energy. For the great controversy regarding the form of the electrostatic energy, or equivalently the form of the Maxwell stress tensor, the reader should consult the footnotes in Eringen (1962). The variational principle imposes also constraints on the form of the constitutive relations, which in our case read:

$$S = S(F, \Pi) \equiv \frac{\partial W}{\partial F},$$
 (2.14)

$$S = S(\mathbb{F}, \Pi) \equiv \frac{\partial W}{\partial \mathbb{F}}, \qquad (2.14)$$

$$E = E(\Pi, \mathbb{F}) \equiv \frac{\partial W}{\partial \Pi}. \qquad (2.15)$$

The free energy depends both on the strain and the polarization:

$$W = W(E, \Pi),$$
 (2.16)

where $\mathbb{E} = \frac{1}{2} (\mathbb{F}^T \mathbb{F} - \mathbb{I})$ is the Green finite strain tensor and $\Pi = P/\rho$ is the polarization per unit mass. ρ is the density in Ω_d and is related to the density ρ_0 in Ω through the equation $\rho_0 = J\rho$. For more rigour about the constrains that both the material symmetry and the second law of thermodynamics impose on the exact form of the constitutive laws (2.14-15) see Eringen & Maugin (1989). For small concentration of the charged micro- or nanoparticles (diluted electro-gel), the polarization contributions to the free energy density W are negligible, thus

$$W \simeq W(\mathbb{E})$$
. (2.17)

Then equations (2.14-15) are replaced by the more traditional uncoupled ones:

$$\mathbb{S} \simeq \mathbb{S}(\mathbb{F}) = \frac{\partial W}{\partial \mathbb{F}},$$
 (2.18)
 $P \simeq P(E).$ (2.19)

$$P \simeq P(E)$$
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Henceforth, we will restrict attention to diluted electro-gels, with constitutive equations of the form (2.18-19) and vanishing mechanical surface traction. In that case, Maxwell' s equations of electrostatics (2.2-3) are no longer coupled with the "mechanical" ones (2.1). Introducing the electrostatic potential Φ in (2.2-3) results in

$$\varepsilon_0 \nabla^2 \Phi = \nabla \cdot P - q_f \delta(x), \quad E \equiv -\nabla \Phi.$$
 (2.20)

If we express the nominal stress tensor $\mathbb S$ in terms of the Biot stresses $t_1^{(1)},\,t_2^{(1)},\,t_3^{(1)}$ (the principal values of the Biot stress tensor (Ogden 1997)):

$$T^{(1)} = (SR + R^TS^T)/2,$$
 (2.21)

where \mathbb{R} is the finite rotation tensor, the derived problem is a complicated but well posed one; it involves four second-order partial differential equations (in $t_i^{(1)}$, (i = $(1, 2, 3), \Phi_{in}$ in the region Ω inside $\partial\Omega$, one second-order partial differential equation (in Φ_{out}) in the region $S - \Omega$ outside $\partial \Omega$, four boundary conditions on $\partial \Omega$, the continuity $[\Phi] = 0$ of the potential on $\partial\Omega$ and the usual regularity conditions at infinity. Even for relatively simple prescribed deformation modes, that satisfy (2.1), the nonlinearities involved in (2.20) due to (2.19), make the resultant potential problem a formidable task. For its solution, in special cases, a number of simplifications must be introduced, guided by the observed geometry of deformation, as well as from physical considerations.

3. Theory and experiment

Consider a homogeneous, isotropic, diluted electro-gel, in the form of a rectangular block of cubic cross-section, of width ζ and height L that is placed at a distance R from the coordinates origin, between the two electrodes of figure 1. Let L_e be the distance between the electrodes and E_0 the uniform static electric field produced by them. We assume that the electro-gel admits plane deformations of the form

$$r = f(X_1), \quad \theta = g(X_2), \quad x_3 = X_3,$$
 (3.1)

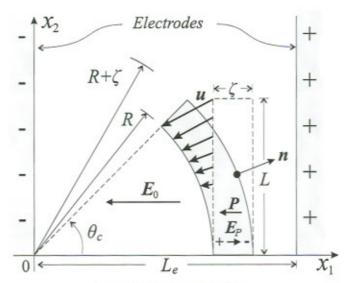


Figure 1. Problem geometry.

shown in figure 1. Then the displacement field,

$$u = X - x(X), \qquad (3.2)$$

corresponds to the bending of the dashed rectangle, of figure 1, into a section of a circular disc, with radius difference $\Delta R = \zeta$. The electro-gel is kept fixed at points X = (R, 0) and $(R + \zeta, 0)$, that is:

$$u(R, 0) = u(R + \zeta, 0) = 0.$$
 (3.3)

Since the electro-gel is considered incompressible, its volume v remains invariant, during the electrically driven deformation, $v(\Omega) = v(\Omega_d)$. This condition determines the maximum deflection angle θ_c in terms of geometrical parameters:

$$\theta_c = 2L/(2R + \zeta). \qquad (3.4)$$

The principal stretches λ_i , i = 1, 2, 3 are given, due to (3.1) by the relations:

$$\lambda_1 = f'(X_1), \quad \lambda_2 = f(X_1) g'(X_2), \quad \lambda_3 = 1,$$
(3.5)

where the prime denotes differentiation with respect to the argument. The incompressibility constraint

$$\lambda_1 \, \lambda_2 \, \lambda_3 = 1 \tag{3.6}$$

results, due to (3.5), after separation of variables, to the solution:

$$r = \sqrt{2B_1 X_1 + B_2}, \quad \theta = X_2/B_1 + B_3.$$
 (3.7)

The unknown constants B_i , i = 1, 2, 3 are determined from the conditions (3.3)

$$B_1 = R + \zeta/2 = L/\theta_c,$$

 $B_2 = -R(R + \zeta) = (\zeta/2)^2 - (L/\theta_c)^2,$ (3.8)
 $B_2 = 0$

In order to treat in a relatively easy manner the electrostatic problem, we assume that the polarization vector P and the electric field E, either inside or outside the electro-gel, are constant and collinear to the external uniform applied field E_0 :

$$E_{in} = \chi_{\epsilon} E_0, \qquad (3.9)$$

$$E_{\text{out}} \simeq E_0$$
, (3.10)

$$P = \varepsilon_0 \chi_{\pi} E_{in} = \varepsilon_0 \chi_{\pi} \chi_{\epsilon} E_0, \qquad (3.11)$$

$$E_0 = -E \hat{E}_1$$
, $E = \text{const.} > 0$, (3.12)

where χ_{ϵ} and χ_{π} are dimensionless functions of the electric field E and the shape of the electro-gel L/ζ , and \hat{E}_i and \hat{e}_i , (i=1,2) the unit vectors in the reference and present configuration, respectively. The above assumptions are valid for the bulk of the electro-gel but not close to the boundary $\partial\Omega$. The solution (3.9-12) satisfies the electrostatic problem (2.2-3), while the jump condition (2.5) reduces to

$$\varepsilon_0(1 - \chi) E_0 \cdot n = \sigma_s$$
, on $\partial \Omega_d$, (3.13)

with χ the electric susceptibility given by:

$$\chi = \chi(E, L/\zeta) = \chi_{\epsilon} (1 + \chi_{\pi}). \qquad (3.14)$$

In general, $E_{\rm in} < E_{\rm out}$, due to the presence of polarization effects, so $0 \le \chi_{\epsilon} \le 1$ and $\chi_{\pi} > 0$. For vanishing free body charges, $q_f = 0$, and due to the assumptions (3.9-12), equation (2.7) gives $f_e = 0$. Then, due to the constitutive laws (2.18-19), the equilibrium problem (2.1) reduces to the purely mechanical one:

$$\nabla_X \cdot S = 0.$$
 (3.15)

Moreover, since the electro-gel is considered isotropic and due to deformation mode considered (3.1-8), the nominal stress tensor S admits, according to Ogden (1997), the decomposition:

$$S = T^{-1}R^T = t_1^{(1)}\hat{E}_1 \otimes \hat{e}_r + t_2^{(1)}\hat{E}_2 \otimes \hat{e}_\theta,$$
 (3.16)

with $t_i^{(1)} \equiv \partial W/\partial \lambda_i$, (i=1,2). If we substitute (3.7-8) into (3.5), we obtain

$$\lambda_1 = 1/\lambda_2 = \lambda = B_1/r = [2\theta_c X_1/L + \zeta^2 \theta_c^2/(4L^2) - 1]^{-1/2},$$
 (3.17)

and the Biot stresses $t_i^{(1)}$, (i = 1, 2) become

$$t_1^{(1)} = \tau(\lambda) = W'(\lambda),$$
 (3.18)
 $t_2^{(1)} = -\lambda^2 \tau(\lambda),$ (3.19)

$$t_2^{(1)} = -\lambda^2 \tau(\lambda),$$
 (3.19)

where prime denotes differentiation with respect to the argument. Then, with the aid of (3.16) and (3.18-19), the mechanical equilibrium equation (3.15) reduces to

$$\lambda \tau'(\lambda) - \tau(\lambda) = 0,$$
 (3.20)

since from (3.17) $\partial t_i^{(1)}/\partial X_2 = 0$. The general solution of (3.20) is

$$\tau(\lambda) = W'(\lambda) = C_1 \lambda, \quad W(\lambda) = C_1 \lambda^2 / 2 + C_2,$$
 (3.21)

where C_i , (i=1,2) are undetermined constants. For vanishing free surface charges $(\sigma_s=0)$, in accordance with our assumption for a diluted electro-gel, the balance of the electro-mechanical surface tractions (2.13) results, due to the solutions (3.9-12) and (3.16), (3.18-19) to $\tau(\lambda) = P_n^2/(2\varepsilon_0)$, or equivalently, due to (3.21) to:

$$2\lambda - e^2 (1 - \chi_{\epsilon})^2 \cos^2 \theta = 0$$
, $e \equiv E/E_s$, $E_s \equiv (2C_1/\epsilon_0)^{1/2}$, (3.22)

on the boundary $X_1=R+\zeta$. The assumption $\sigma_s=0$ implies, from (3.13), that $E_{\rm in}=E_{\rm out}$ or $\chi=1$, since $E_0\cdot n\neq 0$ on $\partial\Omega$. In order to preserve the positive definite character of the strain-energy function $W(\lambda)$ we must have $C_1>0$. Notice that equation (3.22) does not hold for every θ on $\partial\Omega$, but since our primary concern is to model available experimental data, we just have to satisfy (3.22) only for the maximum deflection angle θ_c , since what is measured in the experiments is $u_x=u_x(R+\zeta,L)$ for given E and L/ζ . From (3.17) and (3.22) we obtain, for $X_1=R+\zeta$ and $\theta=\theta_c$:

$$e^{2}(1 - \chi_{\epsilon})^{2}[1 + \zeta \theta_{c}/(2L)]\cos^{2}\theta_{c} = 2.$$
 (3.23)

If we solve (3.23) for θ_c and substitute the result in

$$u \equiv u_x/(R + \zeta) = 1 - \cos \theta_c, \qquad (3.24)$$

we derive the displacement u, as a function of the applied field e and its shape L/ζ , provided that the function $\chi_{\epsilon} = \chi_{\epsilon}(e, L/\zeta)$ will be specified. Fehér et. al. (2001) observed, that u(e=0)=0. Unfortunately, the solution θ_c of (3.23) and thus u of (3.24) are singular at e=0, for a diluted electro-gel, $\chi_{\epsilon}(0, L/\zeta)=0$. This singularity is a consequence of isotropy, collinearity and especially homogeneity introduced in (3.9-12). Since the main physical mechanisms, observed in the experiments, are present in our model, we can overthrow the singularity at e=0, by neglecting (3.4) and replacing the geometrical definition of λ (3.17), with a suitable function of e and L/ζ . Thus, if we substitute,

$$\lambda = \lambda_0 (1 + \chi_{\epsilon})/(1 + \gamma \chi_{\epsilon}), \quad \lambda_0 \equiv e^2 (1 - \chi_{\epsilon})^2 / 2,$$
 (3.25)

with $\gamma \geq 1$, in (3.22) and solve for $\cos \theta_c$, the displacement (3.24) reduces to

$$u \simeq u_x/L_e = 1 - [(1 + \chi_\epsilon)/(1 + \gamma \chi_\epsilon)]^{1/2}$$
. (3.26)

The longer is the surface of the electro-gel, that is exposed to the uniform external field, the larger the depolarization effects induced on it and as a consequence the smaller the total electric field inside. In order to take into account this shape dependence of the constitutive law (3.11) we admit for χ_{ϵ} the simple power law:

$$\chi_{\epsilon} = \alpha (L/\zeta)^{2\beta} e^{\beta},$$
(3.27)

where all α , β and γ are dimensionless constants. Although the form (3.25) has the drawback that $\lambda(e=0)=0$, compared to the expected $\lambda(e=0)=1$, it recovers the observed u(e=0)=0, due to (3.27). According to experiment (see Fehér et. al. (2001)), increase of the height of the electro-gel L resulted in larger deformations for the same externally applied electric field E. This is predicted from our model,

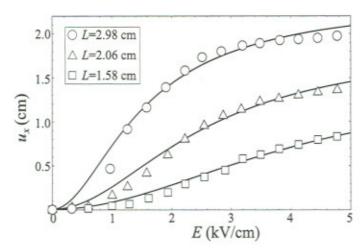


Figure 2. The displacement u_x as a function of applied electric field E, for varying length L. The full lines correspond to equations (3.26-27). Circles, triangles and cubes are experimental data from Filipcsei et. al. (2000).

Table 1. Calculated parameters from experimental data

L(cm)	γ	$\alpha \times 10^{-7}$	$E_s (kV/cm)$
2.98	20.6	0.47351	0.41228
2.06	7.0	1.86657	0.45290
1.58	4.0	3.08580	0.41176

due to shape dependence introduced in (3.27). The comparison between our model (3.26-27) and the experiments of Fehér et. al. (2001), is performed in figure 2, for three values of the length of the electro-gel. All the full lines in figure 2, correspond to $\beta=2$, because in the limit of infinitesimal applied electric fields $(E\to 0)$ the displacement (3.26) reduces to:

$$u_x \simeq A (L/\zeta)^4 E^2$$
, $A = \alpha (\gamma - 1) L_e/(2 E_s^2)$. (3.28)

This law, in the form

$$u_x = D L^4 V^2$$
, $D = 9.1 \times 10^{-3} \text{ kV}^{-2} \text{ cm}^{-3}$, (3.29)

where V is the applied voltage, has been observed by Fehér et. al. (2001). Thus with the substitution $E = V/L_e$ in (3.28) and comparing the result with (3.29), we express E_s , or equivalently C_1 , due to (3.22), in terms of α , γ and D as:

$$C_1 = \alpha \varepsilon_0 (\gamma - 1)/(4 D \zeta^4 L_e). \qquad (3.30)$$

The full curves in figure 2 correspond to the values of the parameters in table 1, with $\beta=2$, $\zeta=0.1\,\mathrm{cm}$ and $L_e=3\,\mathrm{cm}$. Better agreement with the experimental data, for $E\leq 2\,\mathrm{kV/cm}$, could be obtained by allowing β to vary. In order to emphasize on the observed linear dependence (3.29), for $E\to 0$, we present on figure 3 the displacement u_x as a function of $L^4\,V^2$. The dashed line corresponds to (3.29).

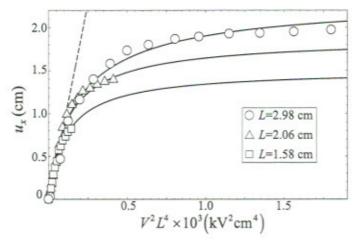


Figure 3. Same as figure 2, but with the displacement u_x as a function of the $L^4 V^2$.

4. Discussion

In this communication, we developed the theoretical framework, for treating large deformations in electric field sensitive gels, based on the continuum theory of electro-elasticity. The simplified model proposed in §3, includes all the information for quantitative interpretation of electric field dependent deformation measurements, since it captures the main physical mechanisms: shape, charged microor nanoparticle concentration of the electro-gel and nonlinearities on constitutive laws. The preservation of the basic features of the geometrical character of the observed deformation (maximum deflection angle, aspect ratio) were essential for the success of the theoretical approach. Drawbacks like the limitation to unit electric susceptibilites ($\chi = 1$), are consistent with diluted electro-gel assumptions and not of major concern, due to bio-compatibility issues. Stability topics, either experimental or theoretical, related to applied electric field strength and external loads (mechanical, hydrodynamical, gravitational, chemical, etc.), should be examined, for future biomedical applications. Electrophoretic forces, due to inhomogeneities in electric field distribution ($f_e \neq 0$) can also be taken into account, with the cost of complicating the solution procedure. Due to its generality, the presented theoretical analysis easily conforms with similar observed mechanisms in biophysics (paraand diamagnetic elastic responses of plants and biological tissues in magnetic fields (see Kuznetsov et. al. 1999)), as well as in prototypes in the rapidly developing field of micro-electro-mechanical systems (MEMS) and their medical counterparts, biomedical microdevices (Bio-MEMS). With minor modifications in the general theory, similar deformation phenomena in ferrogels in external magnetic fields can be explained. Results will be published in the near future.

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