



# A nonlinear acoustomechanical field theory of polymeric gels



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## ABSTRACT

A nonlinear acoustomechanical field theory is presented for polymeric gels undergoing large deformation coupled with diffusion mass transport of solvent molecules in and out of the gel. The theory is developed by combining the acoustic radiation stress theory with the nonlinear elasticity theory of polymeric gels. Explicit velocity and acoustic fields are determined by solving the elastodynamical equations of wave propagation in Eulerian coordinates, which are then employed to determine the distribution of acoustic radiation stresses inside the gel. The nonlinear elasticity of gels is modeled by adopting the Flory–Rehner free energy functions for network stretching and molecules-polymer mixing. For illustration, the developed theory is applied to a layer of polymeric gel immersed in external solvent subjected to two counterpropagating acoustic waves. The acoustically actuated large deformation of the gel is analyzed under three different constraint conditions. Unique acoustomechanical behaviors of polymeric gels are revealed, such as periodical response and nonlinear chaos. This work is expected to enable novel design of ultrasound-triggered sensors and actuators made of polymeric gels, and can also enlighten the application of ultrasonic waves in biomedical engineering.

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## 1. Introduction

Polymeric gel is often immersed in a liquid environment and can be regarded as an assembly of three-dimensional cross-linked networks and small molecule solvent. With the migration of small molecules in and out of the gel when subjected to external stimuli, it is capable of undergoing large and recoverable elastic deformation. This functionality of polymeric gel enables applications such as carriers for drug delivery (Duncan, 2003; Fischel-Ghodsian et al., 1988; Jeong et al., 1997), actuators and sensors in microfluidic devices (Beebe et al., 2000; Calvert, 2009), and tissue engineering matrices (Lee and Mooney, 2001; Luo and Shoichet, 2004). Recently, there is considerable interest in harnessing acoustic waves to actuate large and nonlinear deformation in polymeric gels, with promising applications in medical devices, microfluidic manipulation devices, adaptive robots, etc. (Huebsch et al., 2014; Xin and Lu, 2016b). With regard to large deformation of gels coupled with diffusion mass transport under either mechanical or electric loading, there already exist a great deal of theoretical works (Biot, 1941; Chester and Anand, 2010; Hong et al., 2008; Tanaka and Fillmore, 1979). However, there is yet a comprehensive acoustomechanical field theory of acoustic-triggered deformation and mass transport of polymeric gels. Such a theory is formulated in the present study.

This work should be of paramount importance for gain a fundamental insight into a series of ultrasound techniques and thus can enlighten the widespread application of ultrasound technique in biomedical engineering (De Cock et al., 2016; Zhang et al., 2016).

The Poynting theory says that an acoustic wave propagating in a medium also carries momentum flux in the direction of wave passing on, which gives rise to a back acoustic radiation pressure on the source (Post, 1953, 1960). Generally, the radiation stress is a field concept since it is induced by acoustic momentum transfer between adjacent medium particles and directly deduced from the nonlinearity of acoustic field. Lord Rayleigh firstly gave the expression of acoustic radiation stress in compressional fluids (Rayleigh, 1905), while Brillouin firstly pointed out the second-rank tensor nature of acoustic radiation stress (Beyer, 1978). They all concluded that the radiation stress is proportional to the mean mechanical energy density  $\langle E \rangle$  of medium particle motion. Subsequently, enormous efforts are devoted to investigating radiation forces acting on rigid or compressible spheres (Doinkov, 1994; Hasegawa and Yosioka, 1969), acoustical trapping and tweezers (Caleap and Drinkwater, 2014; Shi et al., 2009), acoustic levitation and contactless handling of matter (Brandt, 2001; Foresti and Poulikakos, 2014), deforming fluid interface and biological tissue (Issenmann et al., 2008; Mishra et al., 2014; Walker, 1999), etc. All these researches demonstrate the fact that acoustic radiation stress generated by focused acoustic input is sufficiently large to levitate a metallic sphere and induce material deformation.

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Since the elastic moduli of polymeric gels typically range from dozens of times Pa to several times kPa, acoustic radiation stress with magnitude of mega pascal can induce large deformation in the gel (Mishra et al., 2014; Xin and Lu, 2016b). As previously mentioned, acoustically triggered gel deformation is attractive for a variety of applications. Therefore, it is necessary to formulate a nonlinear acoustomechanical theory for polymeric gels. In the present study, a nonlinear acoustomechanical field theory is developed in Eulerian coordinates for polymeric gels coupled with diffusion mass transport, by combining the nonlinear elasticity theory of gels and the acoustic radiation stress theory. For illustration, the large deformation and mass transport behavior of gels is separately analyzed under biaxial constraint, uniaxial constraint and freestanding conditions. For convenience, acoustic radiation stress/force will be called as acoustic stress/force. This work provides a theoretical guideline for designing novel acoustically triggered sensors and actuators made of polymeric gels.

## 2. Inhomogeneous field theory of gels

An inhomogeneous field theory of polymeric gels is formulated below in three-dimensional space. The Eulerian coordinates are adopted, since it is more intuitive and convenient for theoretical analyses when all the field quantities are expressed in true values. The polymeric gel is actually an assembly of a three-dimensional cross-linked network of long polymers and one type of small molecules, which is formed when dry networks immerse in and imbibe external solvent. The external solvent is in self-equilibrium and can be regarded as a virtual polymeric gel with networks of vanishing elastic moduli, so that the whole system can be modeled using a uniform field theory. In the field theory, the volume integrals cover both the gel and the solvent, and the surface integrals cover all the interfaces between the gel and the solvent. Let the dry network at undeformed state be the reference configuration. Let each material particle be labeled using its coordinate  $\mathbf{X}$  in the reference configuration, and let each spatial point be labeled using its coordinate  $\mathbf{x}$  in the current configuration. The deformation gradient  $\mathbf{F} = \partial\mathbf{x}/\partial\mathbf{X}$  maps the reference configuration  $\mathbf{X}$  to the current configuration  $\mathbf{x}$ . The Cauchy stress is related to the first Piola–Kirchhoff stress as  $\boldsymbol{\sigma} = \mathbf{s} \cdot \mathbf{F}^T/\det(\mathbf{F})$ . Let  $dV(\mathbf{x})$  be a volume element with mass density  $\rho(\mathbf{x})$  and body force  $\mathbf{f}^b(\mathbf{x}, t)$ . Let  $\mathbf{n}(\mathbf{x})dA(\mathbf{x})$  be a surface element with surface force  $\mathbf{f}^s(\mathbf{x}, t)$ , where  $dA(\mathbf{x})$  is the area of the element and  $\mathbf{n}(\mathbf{x})$  is the unit vector normal to the interface between two materials, labeled as  $-$  for pointing toward outside and  $+$  for pointing toward inside. Force balance of the volume element is represented by  $\partial\boldsymbol{\sigma}/\partial\mathbf{x} + \mathbf{f}^b = \rho\partial^2\mathbf{u}/\partial t^2$ , with force boundary condition  $\boldsymbol{\sigma} \cdot \mathbf{n} = \mathbf{f}^s$  and displacement field  $\mathbf{u}(\mathbf{x}, t)$ .

Now consider an acoustomechanical problem coupled with diffusion mass transport. Essentially, the acoustomechanical problem is a static field problem on the basis of a dynamic field problem. Because the acoustic (radiation) stress is intrinsically a time-averaged stress over one oscillation cycle of an acoustic wave, it is a static stress regardless of time. This static acoustic stress eventually causes static material deformation. Consequently, the acoustomechanical problem is a dynamic field problem for wave propagation but a static field problem for large material deformation. In the present article, all the terms associated with large material deformation are pure static terms without including any inertial terms for wave propagation. In the current configuration, the volume force on an element is  $f_i^b dV$  and the surface force on an element is  $f_i^s dA$ . Force balance dictates that the combination of Cauchy stress and acoustic stress should satisfy:

$$\int (\sigma_{ij} - \tau_{ij}) \frac{\partial \xi_i}{\partial x_j} dV = \int f_i^b \xi_i dV + \int f_i^s \xi_i dA \quad (1)$$

for arbitrary test function  $\xi_i(\mathbf{x})$  where  $\tau_{ij}$  is the acoustic stress (pressure), representing compression when it is positive and tension when it is negative. Here the acoustic (radiation) stress is generated due to the momentum transfer between adjacent particles in the process of ultrasonic wave propagation, which can be calculated from the acoustic fields inside and outside of the material. While the Cauchy stress is generally equal to outside mechanical stress. Following the Gauss divergence theorem, one has:

$$\int (\sigma_{ij} - \tau_{ij}) \frac{\partial \xi_i}{\partial x_j} dV = \int (\sigma_{ij}^- - \sigma_{ij}^+) n_i \xi_i dA - \int (\tau_{ij}^- - \tau_{ij}^+) n_i \xi_i dA - \int \frac{\partial (\sigma_{ij} - \tau_{ij})}{\partial x_j} \xi_i dV \quad (2)$$

The test function  $\xi_i(\mathbf{x})$  is considered to be continuous across the material interface, while the stress can be discontinuous at the interface. The weak form of the force balance condition in Eq. (1) can be rewritten as:

$$\frac{\partial (\sigma_{ij}(\mathbf{x}, t) - \tau_{ij}(\mathbf{x}, t))}{\partial x_j} + f_i^b(\mathbf{x}, t) = 0 \quad (3)$$

in the volume, and

$$\left[ (\sigma_{ij}^-(\mathbf{x}, t) - \sigma_{ij}^+(\mathbf{x}, t)) - (\tau_{ij}^-(\mathbf{x}, t) - \tau_{ij}^+(\mathbf{x}, t)) \right] n_i(\mathbf{x}, t) = f_i^s(\mathbf{x}, t) \quad (4)$$

at the interface. These equations describe the general force balance condition of the acoustomechanical system.

Next consider the diffusion phenomenon of mass transport between polymeric gel and external solvent. The small molecules penetrating in and out of the gel with chemical potential  $\mu(\mathbf{x}, t)$  give rise to the change of the chemical potential and the swelling stress. In the current configuration, let  $r(\mathbf{x}, t)$  be the number of small molecules generated by sources (e.g., chemical reaction) inside the gel per unit time and volume, and let  $i(\mathbf{x}, t)$  be the number of small molecules generated by sources in the interface element per unit time and area. Mass conservation of the small molecules requires:

$$\frac{\partial c(\mathbf{x}, t)}{\partial t} + \frac{\partial j_i(\mathbf{x}, t)}{\partial x_i} = r(\mathbf{x}, t) \quad (5)$$

in the volume, where  $c(\mathbf{x}, t)$  is the concentration of small molecules in a volume element,  $j_i(\mathbf{x}, t)$  is the number flux of small molecules that migrate through unit area of the interface element. Also, at the interface:

$$(j_i^+(\mathbf{x}, t) - j_i^-(\mathbf{x}, t)) n_i(\mathbf{x}, t) = i(\mathbf{x}, t) \quad (6)$$

These diffusion balance conditions can be rewritten in a weak form by applying an arbitrary test function  $\zeta(\mathbf{x})$ . Specifically, multiplying this test function on Eq. (5), integrating over the gel volume and adopting Eq. (6), one obtains:

$$\int \frac{\partial c}{\partial t} \zeta dV = \int j_i \frac{\partial \zeta}{\partial x_i} dV + \int r \zeta dV + \int i \zeta dA \quad (7)$$

This weak form of mass conservation is completely equivalent to Eqs. (5) and (6).

To complement the above field equations expressed in the form of force balance law and mass conservation law, the material law is proposed below by applying the principle of minimization potential energy. Generally, the constitutive relation of hyperelastic materials can be expressed using the Helmholtz free energy function, which is dependent upon the strain or concentration of molecules. With an arbitrary small virtual perturbation, the virtual changes of the free energy can be described as:

$$\delta W_e = \frac{\partial W_e(\boldsymbol{\epsilon})}{\partial \epsilon_{ij}} \delta \epsilon_{ij}, \quad \delta W_a = \frac{\partial W_a(\boldsymbol{\epsilon})}{\partial \epsilon_{ij}} \delta \epsilon_{ij}, \quad \delta W_m = \frac{\partial W_m(c)}{\partial c} \delta c \quad (8)$$

where  $W_e(\boldsymbol{\varepsilon})$  and  $W_a(\boldsymbol{\varepsilon})$  are the elastic strain energy density related to the Cauchy stress and the acoustic stress, respectively, and  $W_m(c)$  is the mixture energy density of polymers and small molecules. The virtual perturbation also leads the external force, the inside source, and the molecules diffusion to do work on the system. With time passing  $\delta t$ , the system has a perturbed displacement field  $\delta \mathbf{u}$ , the number of small molecules per unit volume generated by the inside source is  $\delta R = r(\mathbf{x}, t)\delta t$ , and the number of small molecules diffused into the gel per unit area is  $\delta I = i(\mathbf{x}, t)\delta t$ . That is, the external force does work  $\int f_i^b \cdot \delta u_i dV + \int f_i^s \cdot \delta u_i dA$ , and the molecules do work  $\int \mu \cdot \delta R dV + \int \mu \cdot \delta I dA$ .

The Lagrangian potential energy is:

$$\begin{aligned} II &= U_e + U_a + U_m - W \\ &= \int (W_e + W_a + W_m) dV - \int f_i^b u_i dV \\ &\quad - \int f_i^s u_i dA - \int \mu R dV - \int \mu I dA \end{aligned} \quad (9)$$

where  $U_e$  and  $U_a$  are the strain energy related to the Cauchy stress and the acoustic stress, respectively,  $U_m$  is the mixture energy of polymers and molecules, and  $W$  is the work done by stimuli. The strain energies due to Cauchy stress and acoustic stress are additive, because for an infinitesimal process from any instant of the quasi-static material deformation process, the constant acoustic stress's doing work is additive to the constant Cauchy stress's doing work during this infinitesimal process. Incorporating Eqs. (2), (7) and (8), one can express the virtual change of the Lagrangian potential energy as:

$$\begin{aligned} \delta II &= \delta U_e + \delta U_a + \delta U_m - \delta W \\ &= \int \left( \frac{\partial W_e}{\partial \varepsilon_{ij}} - \sigma_{ij} \right) \delta \varepsilon_{ij} dV \\ &\quad + \int \left( \frac{\partial W_a(\boldsymbol{\varepsilon})}{\partial \varepsilon_{ij}} + \tau_{ij} \right) \delta \varepsilon_{ij} dV \\ &\quad + \int \left( \frac{\partial W_m(c)}{\partial c} - \mu \right) \delta R dV + \int \left( \frac{\partial W_m(c)}{\partial c} - \mu \right) \delta I dA \\ &\quad + \int \frac{\partial}{\partial x_i} \frac{\partial W_m(c)}{\partial c} \delta J_i dV \end{aligned} \quad (10)$$

where  $\delta J_i = j_i \delta t$  is the number of small molecules that migrate through unit area of the interface element. The principle of minimization potential energy requires:

$$\delta II \leq 0 \quad (11)$$

which should hold for any arbitrary  $\delta \varepsilon_{ij}$ ,  $\delta R$ ,  $\delta I$  and  $\delta J_i$ . Generally, the principle of minimization potential energy only requires  $\delta II = 0$  for a static equilibrium state. However, there exists a dynamic flux for small molecules moving out of the gel, resulting in  $\delta II < 0$ . To satisfy the condition of (11), the first, second, third and fourth integrands should all vanish, namely:

$$\begin{aligned} \sigma_{ij} &= \frac{\partial W_e(\boldsymbol{\varepsilon})}{\partial \varepsilon_{ij}} = \frac{F_{i\alpha}}{J} \frac{\partial W_e(\mathbf{F})}{\partial F_{j\alpha}} \\ \tau_{ij} &= -\frac{\partial W_a(\boldsymbol{\varepsilon})}{\partial \varepsilon_{ij}} = -\frac{F_{i\alpha}}{J} \frac{\partial W_a(\mathbf{F})}{\partial F_{j\alpha}} \\ \mu &= \frac{\partial W_m(c)}{\partial c} \end{aligned} \quad (12)$$

The first and second integrand terms lead to the Cauchy stress and the acoustic stress, respectively, which all contribute to the inside migration of small molecules in gel and correspond to the non-volumetric shape reconfiguration. The third and fourth integrand terms lead to the chemical potential owing to the field of inside sources pouring out or generating small molecules. The

fifth term introduces the definition of dynamic diffusion flux, as (Biot, 1941):

$$j_i = -\frac{cD}{kT} \frac{\partial \mu}{\partial x_i} \quad (13)$$

where  $c$  is the concentration of small molecules in the current configuration,  $D$  is the coefficient of diffusion for solvent molecules,  $k$  is the Boltzmann constant, and  $T$  is the absolute temperature. The negative value of the diffusion flux ensures the inequality of (11) is tenable. This diffusion process contributes to the volumetric shape reconfiguration, i.e., the swelling or contraction of gel.

### 3. Acoustomechanical constitutive relation of gels

To further construct the acoustomechanical constitutive relation of polymeric gels, the constraint of molecule incompressibility (Hong et al., 2008) should be taken into account. Because polymeric gels often undergo giant deformation without significant volumetric change, one can assume that the networks and molecules are both individually incompressible. Also, as the condensed matter form of the gel occupies all the space, the molecule incompressibility constraint condition is given by:

$$1 + \nu c \det(\mathbf{F}) = \det(\mathbf{F}), \text{ or } 1 + \nu C = \det(\mathbf{F}) \quad (14)$$

where the true concentration relates to the nominal concentration as  $c = C/\det(\mathbf{F})$ ,  $\nu$  is the volume of each molecule, and  $\nu c \det(\mathbf{F}) (= \nu C)$  is the volume of all small molecules in the gel. The molecular incompressibility can be accounted for by adding a constraint term  $\int p_s (1 + \nu c \det(\mathbf{F}) - \det(\mathbf{F})) dV$  to the Lagrangian energy function, where  $p_s(\mathbf{x}, t)$  is the field of Lagrange multiplier. Accordingly, the Cauchy stress and the true concentration can be obtained as:

$$\sigma_{ij} = \frac{F_{i\alpha}}{\det(\mathbf{F})} \left( \frac{\partial W_e(\mathbf{F})}{\partial F_{j\alpha}} + \frac{\partial W_a(\mathbf{F})}{\partial F_{j\alpha}} \right) - p_s \delta_{ij} \quad (15)$$

$$\mu = \frac{\partial W_m(c)}{\partial c} + p_s \nu \det(\mathbf{F}) = \frac{\partial W_m(C)}{\partial C} \det(\mathbf{F}) + p_s \nu \det(\mathbf{F}) \quad (16)$$

where the Lagrange multiplier  $p_s$  is actually the osmotic pressure or swelling pressure. Here the polymeric gel is considered as a condensed matter composed of networks and solvent, the acoustic stress exerts on this condensed matter quite like mechanical stress, which together balances with the elastic deformation stress of the network and the osmotic pressure of the solvent. The diffusion occurs only when the chemical potential difference between the outside and inside of the gel is large enough to overcome the osmotic pressure. In view of the relationship between the first Piola–Kirchhoff stress and the Cauchy stress  $\mathbf{s} = \det(\mathbf{F})\mathbf{F}^{-T}\boldsymbol{\sigma}$ , the first Piola–Kirchhoff stress and the nominal concentration are obtained as:

$$s_{i\alpha} = \frac{\partial W_e(\mathbf{F})}{\partial F_{i\alpha}} + \frac{\partial W_a(\mathbf{F})}{\partial F_{i\alpha}} - p_s F_{i\alpha}^{-T} \det(\mathbf{F}) \quad (17)$$

$$m = \frac{\partial W_m(C)}{\partial C} + p_s \nu, \quad (18)$$

Eqs. (15)–(18) present the constitutive relation of acoustomechanical polymeric gels coupled with diffusion mass transport in terms of the Helmholtz free energy.

To capture the main physical nature of the acoustomechanical behavior of polymeric gels, explicit Helmholtz free energy functions for relatively simple cases are adopted, which should contain contributions from the elastic stress induced energy, the acoustic stress induced energy, and the mixture of networks and molecules induced energy, as:

$$W(\mathbf{F}, c) = W_e(\mathbf{F}) + W_a(\mathbf{F}) + W_m(c) \quad (19)$$

The free energy density contributed by the elastic stress may be written as (Boyce and Arruda, 2000; Flory and Rehner, 1943):

$$W_e(\mathbf{F}) = \frac{1}{2}NkT(I_1 - 3 - 2 \ln J) \quad (20)$$

where  $I_1 = \text{tr}(\mathbf{F}^T \mathbf{F})$ ,  $J = \det(\mathbf{F})$  and  $N$  is the number of polymer chains.

The free energy density due to the mixture between the networks and molecules may be given by:

$$W_m(c) = \frac{kT}{v} \left[ vJc \ln \left( \frac{vJc}{1+vJc} \right) + \frac{\chi vJc}{1+vJc} \right] \quad (21)$$

where the two terms correspond separately to the entropy and enthalpy of the mixture, and  $\chi$  is the Flory parameter for the interaction between solvent and polymers. The molecules are driven to enter the gel when  $\chi < 0$ , and leave the gel when  $\chi > 0$ .

For the acoustic stress induced energy, consider a thin layer of polymeric gel with initial undeformed dimensions  $(L_1, L_2, L_3)$  and let  $L_3$  represents its thickness. Assume a plane acoustic wave propagates along the thickness direction of the layer and penetrates into it. The layer deforms accordingly to current dimensions  $(l_1, l_2, l_3)$  with principal stretches  $(\lambda_1, \lambda_2, \lambda_3)$ . Let  $\lambda_3$  denote the out-of-plane stretch. The free energy density of the gel induced by acoustic stress is actually the negative work density done by the acoustic stress, which can be expressed as (Xin and Lu, 2016a,b,c,d; Xin et al., 2017):

$$W_a(\mathbf{F}) = -J \ln(\lambda_1^{t_1} \lambda_2^{t_2} \lambda_3^{t_3}) \quad (22)$$

where

$$t_1 = \frac{1}{l_3} \int_0^{l_3} \langle T_{11}(z) \rangle dz, \quad t_2 = \frac{1}{l_3} \int_0^{l_3} \langle T_{22}(z) \rangle dz, \quad (23)$$

$$t_3 = \langle T_{33}^{\text{inside}}(l_3) \rangle - \langle T_{33}^{\text{outside}}(l_3) \rangle \quad (24)$$

$$\langle T_{ij} \rangle = \left[ \frac{\rho_a}{2c_a^2} \left\langle \left( \frac{\partial \phi}{\partial t} \right)^2 \right\rangle - \frac{\rho_a \langle (\nabla \phi)^2 \rangle}{2} \right] \delta_{ij} + \rho_a \left\langle \frac{\partial \phi}{\partial x_i} \frac{\partial \phi}{\partial x_j} \right\rangle \quad (24)$$

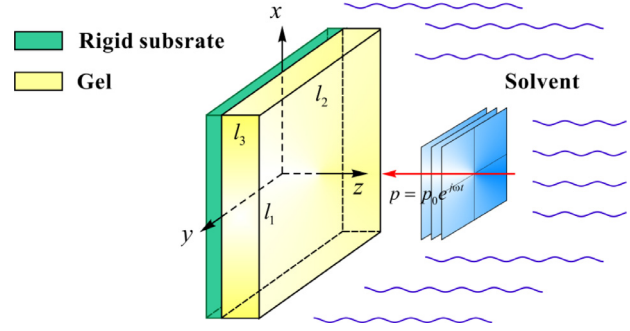
Here,  $\langle T_{ij} \rangle$  is the second-rank acoustic stress tensor (expressed now in Eulerian coordinates),  $\phi$  is the velocity potential (i.e.,  $\mathbf{u} = -\nabla \phi$ ),  $\rho_a$  is the medium density,  $c_a$  is the acoustic speed in the medium,  $\delta_{ij}$  is the Kronecker delta, and  $\langle \cdot \rangle$  denotes the time-average over an oscillation cycle.  $t_1$ ,  $t_2$  and  $t_3$  are the equivalent acoustic stresses exerted on the material interface to cause deformation, with  $l_3$  representing the current thickness of the gel layer. Because the gel is a combination of a three-dimensional cross-linked network and small molecules, it may be taken as a macroscopically homogeneous and isotropic condensed soft matter for wave propagation. Under arbitrary prescribed acoustic inputs and boundary conditions, one can obtain the acoustic and velocity fields and then favorably determine the distribution of acoustic stress inside and outside the gel.

The focus of the present study is placed upon the balance state in which the elastic stress and acoustic stress are balanced with the osmotic pressure. Correspondingly, chemical equilibrium is maintained in and out of the gel, namely,  $\mu = 0$  (or  $m = 0$ ). Adopting the above explicit free energy functions together with Eqs. (15) and (16), one obtains the Cauchy stresses in the gel layer as:

$$\sigma_1 = NkT(\lambda_1 - \lambda_1^{-1})\lambda_2^{-1}\lambda_3^{-1} + \frac{1}{Jv} \frac{dW_m}{dc} - t_1 \quad (25)$$

$$\sigma_2 = NkT(\lambda_2 - \lambda_2^{-1})\lambda_1^{-1}\lambda_3^{-1} + \frac{1}{Jv} \frac{dW_m}{dc} - t_2 \quad (26)$$

$$\sigma_3 = NkT(\lambda_3 - \lambda_3^{-1})\lambda_1^{-1}\lambda_2^{-1} + \frac{1}{Jv} \frac{dW_m}{dc} - t_3 \quad (27)$$



**Fig. 1.** Schematic of a biaxially constrained polymeric gel layer (attached with a rigid substrate), immersed in solvent and subjected to a plane acoustic input  $p = p_0 e^{i\omega t}$ . First, the dry polymer having initial undeformed dimensions  $(L_1, L_2, L_3)$  freely swells to become a gel with dimensions  $(l_1^s, l_2^s, l_3^s)$  when immersed in solvent. Then the gel is fixed to a rigid substrate and struck by an acoustic input, so that it deforms to the current state of dimensions  $(l_1, l_2, l_3)$ , with  $l_1^s = l_1$  and  $l_2^s = l_2$ .

Further, adopting these free energy functions together with Eqs. (17) and (18), one obtains the first Piola–Kirchhoff stresses as:

$$s_1 = NkT(\lambda_1 - \lambda_1^{-1}) + \frac{\lambda_2 \lambda_3}{Jv} \frac{dW_m}{dc} - \lambda_2 \lambda_3 t_1 \quad (28)$$

$$s_2 = NkT(\lambda_2 - \lambda_2^{-1}) + \frac{\lambda_1 \lambda_3}{Jv} \frac{dW_m}{dc} - \lambda_1 \lambda_3 t_2 \quad (29)$$

$$s_3 = NkT(\lambda_3 - \lambda_3^{-1}) + \frac{\lambda_1 \lambda_2}{Jv} \frac{dW_m}{dc} - \lambda_1 \lambda_2 t_3 \quad (30)$$

The above constitutive equations define the mechanical behavior of acoustomechanical polymeric gels coupled with mass transport: (25)–(27) are constitutive relations in true space while (28)–(30) are constitutive relations in nominal space.

Once the boundary conditions are given for a specific problem, one can solve all the state variables in current configuration by employing the constitutive relations of acoustomechanical polymeric gel. Assume the external solvent is in liquid state, so that the work done by atmospheric pressure during the migration process of small molecules into the gel can be neglected. Assume further the liquid solvent is in its equilibrium state to the reference state. With this reference state, the chemical potential of small molecules can be defined as the free energy alteration  $\mu = (p - p_0)v$ , as a result of one molecule migration from reference state to current state.  $p_0$  is the solvent pressure in reference state and  $p$  is the solvent pressure in current state. The mechanical boundary conditions can be given in the form of displacement boundary value  $\mathbf{u}(\mathbf{x}, t)$  or force boundary value  $\mathbf{F}(\mathbf{x}, t)$ . The chemical boundary conditions can be the prescribed boundary value of chemical potential  $\mu(\mathbf{x}, t)$  or molecular number flux  $\mathbf{j}(\mathbf{x}, t)$ .

In the following numerical calculations, representative values of relevant parameters for a typical gel are adopted as: volume per molecule  $v = 10^{-28} \text{m}^3$ , at room temperature  $kT = 4 \times 10^{-21} \text{J}$  and  $kT/v = 4 \times 10^7 \text{Pa}$ . The Flory–Rehner free energy functions of Eqs. (20) and (21) contain two dimensionless parameters  $Nv$  and  $\chi$ . If a dry network has small-strain shear modulus  $NkT = 4 \times 10^4 \text{Pa}$ , then  $Nv = 10^{-3}$ . The dimensionless parameter  $\chi$  is a measure of the enthalpy of mixing, and its typical value falls within the range between 0 and 1.2. Without otherwise statement,  $\chi = 0.5$  is used.

#### 4. Acoustical actuation under biaxial constraint

With reference to Fig. 1, consider a layer of polymeric gel immersed in external solvent. The dry network with initial dimensions  $(L_1, L_2, L_3)$  first freely swells to become a gel with dimensions  $(l_1^s, l_2^s, l_3^s)$  when it equilibrates with the outside solvent. Then



it is attached with a rigid substrate. Assume a plane acoustic wave propagates along the thickness direction of the gel and penetrates into the gel, which will be totally reflected by the rigid substrate and then interacts with the incoming wave to generate a standing-wave field. With wave propagation in the medium governed by  $\nabla \cdot \sigma = \rho \partial^2 \mathbf{u} / \partial t^2$  in Eulerian coordinates and the total reflection boundary condition at the rigid substrate interface, the velocity field and acoustic field of the standing-wave can be expressed as:

$$u_{1z} = jk_{1z} e^{j\omega t} \left[ I e^{-jk_{1z}z} - I e^{jk_{1z}z} \right. \\ \left. \times \frac{e^{jk_{2z}l_3} (k_{1z}k_{2z}\rho_2 - k_{2z}^2\rho_1) + e^{-jk_{2z}l_3} (k_{1z}k_{2z}\rho_2 + k_{2z}^2\rho_1)}{e^{jk_{2z}l_3} (k_{1z}k_{2z}\rho_2 + k_{2z}^2\rho_1) + e^{-jk_{2z}l_3} (k_{1z}k_{2z}\rho_2 - k_{2z}^2\rho_1)} \right] \quad (31)$$

$$p_1 = j\omega\rho_1 e^{j\omega t} \left[ I e^{-jk_{1z}z} + I e^{jk_{1z}z} \right. \\ \left. \times \frac{e^{jk_{2z}l_3} (k_{1z}k_{2z}\rho_2 - k_{2z}^2\rho_1) + e^{-jk_{2z}l_3} (k_{1z}k_{2z}\rho_2 + k_{2z}^2\rho_1)}{e^{jk_{2z}l_3} (k_{1z}k_{2z}\rho_2 + k_{2z}^2\rho_1) + e^{-jk_{2z}l_3} (k_{1z}k_{2z}\rho_2 - k_{2z}^2\rho_1)} \right] \quad (32)$$

$$u_{2z} = \frac{2jI\rho_1 k_{1z} k_{2z}^2 e^{j\omega t} (e^{jk_{2z}(z+l_3)} - e^{jk_{2z}(z-l_3)})}{e^{jk_{2z}l_3} (\rho_2 k_{1z} k_{2z} + \rho_1 k_{2z}^2) + e^{-jk_{2z}l_3} (\rho_2 k_{1z} k_{2z} - \rho_1 k_{2z}^2)} \quad (33)$$

$$p_2 = \frac{2j\omega I \rho_1 \rho_2 k_{1z} k_{2z} e^{j\omega t} (e^{-jk_{2z}(z-l_3)} + e^{jk_{2z}(z-l_3)})}{e^{jk_{2z}l_3} (\rho_2 k_{1z} k_{2z} + \rho_1 k_{2z}^2) + e^{-jk_{2z}l_3} (\rho_2 k_{1z} k_{2z} - \rho_1 k_{2z}^2)} \quad (34)$$

where the subscripts 1 and 2 denote variables related to outside solvent and gel,  $u_z$  and  $p$  are the velocity and pressure,  $\rho$  is the density of medium,  $k_z$  is the wavenumber of acoustic wave,  $l$  is the amplitude of velocity potential, and  $l_3$  is the current thickness of gel. Making use of the above velocity and acoustic fields, one can obtain the acoustic stress distributions in and out of the gel as:

$$\langle T_{11}^{inside} \rangle = \langle T_{22}^{inside} \rangle = \frac{A_{t2} B_{t2}^* e^{-2jk_{2z}z} + A_{t2}^* B_{t2} e^{2jk_{2z}z}}{2\rho_2 c_2^2}, \\ \langle T_{33}^{inside} \rangle = \frac{A_{t2} A_{t2}^* + B_{t2} B_{t2}^*}{2\rho_2 c_2^2} \quad (35)$$

$$\langle T_{11}^{outside} \rangle = \langle T_{22}^{outside} \rangle = \frac{A_{t1} B_{t1}^* e^{-2jk_{2z}z} + A_{t1}^* B_{t1} e^{2jk_{2z}z}}{2\rho_1 c_1^2}, \\ \langle T_{33}^{outside} \rangle = \frac{A_{t1} A_{t1}^* + B_{t1} B_{t1}^*}{2\rho_1 c_1^2} \quad (36)$$

The corresponding equivalent stresses are:

$$t_1 = t_2 = \frac{1}{2\rho_2 c_2^2} \\ \times \left\{ \frac{1}{2jk_{2z}l_3} [A_{t2}^* B_{t2} (e^{2jk_{2z}l_3} - 1) - A_{t2} B_{t2}^* (e^{-2jk_{2z}l_3} - 1)] \right\} \quad (37)$$

$$t_3 = \frac{A_{t2} A_{t2}^* + B_{t2} B_{t2}^*}{2\rho_2 c_2^2} - \frac{A_{t1} A_{t1}^* + B_{t1} B_{t1}^*}{2\rho_1 c_1^2} \quad (38)$$

where the superscript asterisk \* means complex conjugates of the corresponding variables, and:

$$A_{t1} = j\omega\rho_1 I \quad (39)$$

$$B_{t1} = j\omega\rho_1 I \frac{e^{jk_{2z}l_3} (k_{1z}k_{2z}\rho_2 - k_{2z}^2\rho_1) + e^{-jk_{2z}l_3} (k_{1z}k_{2z}\rho_2 + k_{2z}^2\rho_1)}{e^{jk_{2z}l_3} (k_{1z}k_{2z}\rho_2 + k_{2z}^2\rho_1) + e^{-jk_{2z}l_3} (k_{1z}k_{2z}\rho_2 - k_{2z}^2\rho_1)} \quad (40)$$

$$A_{t2} = \frac{2j\omega I \rho_1 \rho_2 k_{1z} k_{2z} e^{jk_{2z}l_3}}{e^{jk_{2z}l_3} (\rho_2 k_{1z} k_{2z} + \rho_1 k_{2z}^2) + e^{-jk_{2z}l_3} (\rho_2 k_{1z} k_{2z} - \rho_1 k_{2z}^2)} \quad (41)$$

$$B_{t2} = \frac{2j\omega I \rho_1 \rho_2 k_{1z} k_{2z} e^{-jk_{2z}l_3}}{e^{jk_{2z}l_3} (\rho_2 k_{1z} k_{2z} + \rho_1 k_{2z}^2) + e^{-jk_{2z}l_3} (\rho_2 k_{1z} k_{2z} - \rho_1 k_{2z}^2)} \quad (42)$$

Under the acoustic input, the gel can be stretched along its thickness direction. However, it has fixed in-plane dimensions ( $l_1^0$ ,  $l_2^0$ ) and fixed in-plane stretch  $\lambda_1 = \lambda_2$ . The normalized Cauchy stress in the out-of-plane direction can be given by:

$$\frac{\sigma_3 v}{kT} = Nv(\lambda_3 - \lambda_3^{-1})\lambda_1^{-2} \\ + [\ln(1 - \lambda_1^{-2}\lambda_3^{-1}) + \lambda_1^{-2}\lambda_3^{-1} + \chi\lambda_1^{-4}\lambda_3^{-2}] - \frac{vt_3}{kT} \quad (43)$$

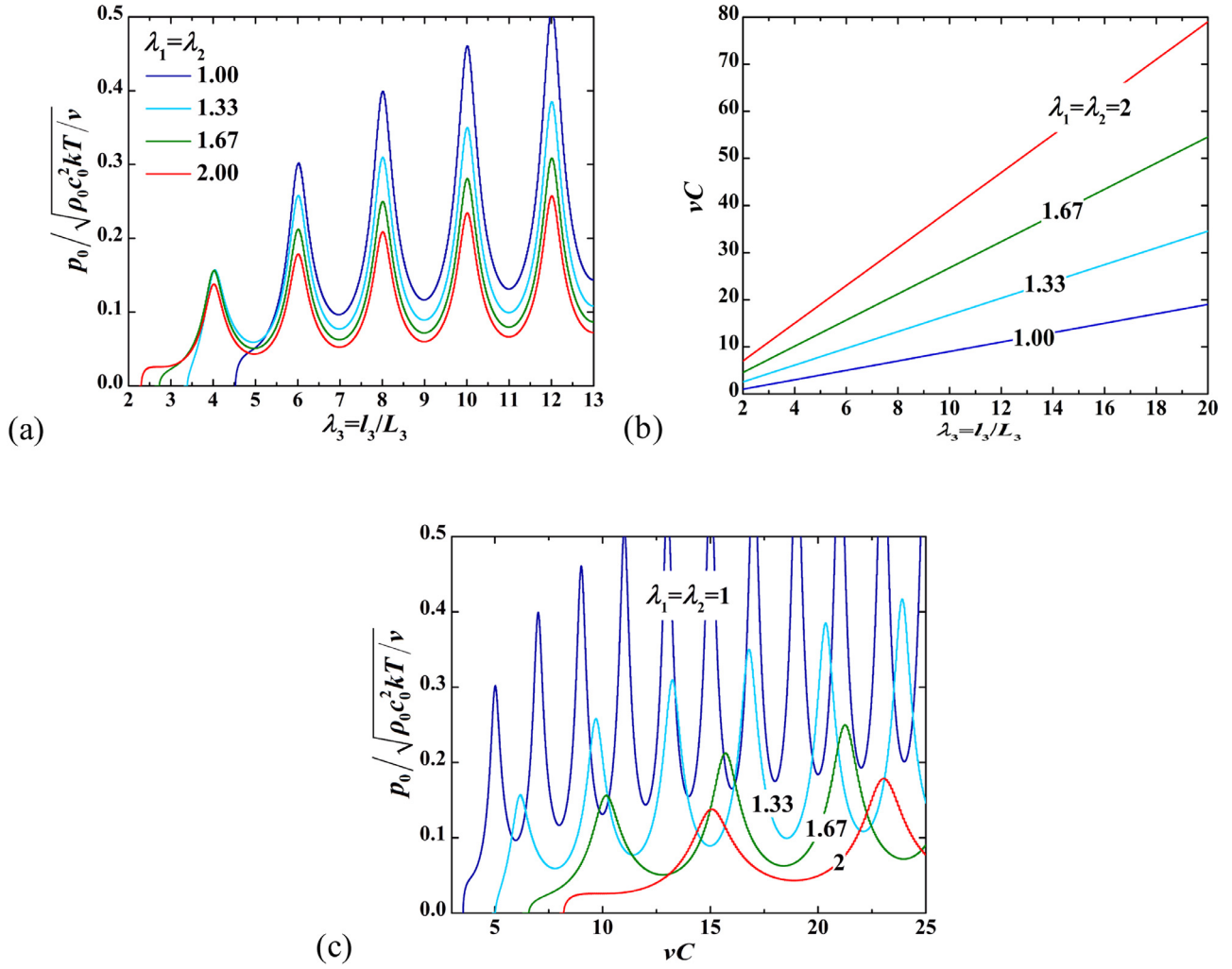
In the absence of external mechanical force, the Cauchy stress should equal to zero. With Eq. (43), the normalized acoustic input  $p_0/\sqrt{\rho_0 c_0^2 kT/v}$  can be extracted as a function of out-of-plane stretch  $\lambda_3$ , where  $p_0$  is the amplitude of the acoustic input and  $\rho_0 c_0$  is the acoustic impedance of the solvent.

Fig. 2 presents the acoustomechanical responses of polymeric gel coupled with diffusion mass transport under prescribed biaxial constraints of  $\lambda_1 = \lambda_2 = 1, 1.33, 1.67$  and  $2$ . As the extent of biaxial constraint is varied, the deformation of gel is acoustically actuated from different initial out-of-plane stretches when  $p_0/\sqrt{\rho_0 c_0^2 kT/v} = 0$ , as shown in Fig. 2(a) and (c). This is because the gel can further swell to equilibrium with the outside solvent even after the gel is biaxially constrained to the rigid substrate. The acoustic input-stretch curves show remarkable periodic properties when the gel is further acoustically deformed after the initial swelling stretch, which is attributed to the fact that the acoustic stress  $t_3$  varies periodically as a function of the out-of-plane stretch. The larger the biaxial constraint is, the smaller the acoustic input required to maintain the same deformation is. In other words, the in-plane prestretch is able to amplify the acoustomechanical response of polymeric gels. Due to the molecule incompressibility condition of  $1 + vC = \lambda_1 \lambda_2 \lambda_3$ , the ratio of molecules volume over dry polymers  $vC$  is a linear function of the out-of-plane stretch, as shown in Fig. 2(b). The larger the biaxial constraint is, the larger the ratio  $vC$  at the same stretch is. As the biaxial constraint is varied, the acoustic input versus volume ratio curves also display periodical feature but have different periods (Fig. 2(c)).

## 5. Acoustical actuation under uniaxial constraint

Consider a layer of polymeric gel fixed between two rigid substrates with a prescribed swelling stretch of  $\lambda_1 = \lambda_p$ , which is further subjected to two counterpropagating acoustic waves along its thickness direction, as sketched in Fig. 3. As acoustic wave propagation obeys the elastodynamical equations, superposition of the two opposing velocity fields and acoustic fields leads to:

$$u_{1z} = jk_{1z} I e^{j\omega t} \left[ e^{-jk_{1z}z} - e^{jk_{1z}z} \right. \\ \left. \times \frac{j \sin(k_{2z}l_3) (\rho_2^2 k_{1z}^2 - \rho_1^2 k_{2z}^2) + 2\rho_1 \rho_2 k_{1z} k_{2z}}{2\rho_1 \rho_2 k_{1z} k_{2z} \cos(k_{2z}h) + j(\rho_1^2 k_{2z}^2 + \rho_2^2 k_{1z}^2) \sin(k_{2z}h)} \right] \quad (44)$$



**Fig. 2.** Nonlinear acoustomechanical deformation of a polymeric gel layer under prescribed biaxial constraints of  $\lambda_1 = \lambda_2 = 1, 1.33, 1.67$  and  $2$ : (a) acoustic input  $p_0 / \sqrt{\rho_0 c_0^2 k T / \nu}$  plotted as a function of out-of-plane stretch  $\lambda_3$ ; (b) ratio of molecules volume over dry polymers  $\nu C$  plotted as a function of  $\lambda_3$ ; (c)  $p_0 / \sqrt{\rho_0 c_0^2 k T / \nu}$  plotted as a function of  $\nu C$ .

$$p_1 = j\omega\rho_1 l e^{j\omega t} \left[ e^{-jk_{1z}z} + e^{jk_{1z}z} \right. \\ \left. \times \frac{j \sin(k_{2z}l_3)(\rho_2^2 k_{1z}^2 - \rho_1^2 k_{2z}^2) + 2\rho_1\rho_2 k_{1z}k_{2z}}{2\rho_1\rho_2 k_{1z}k_{2z} \cos(k_{2z}h) + j(\rho_1^2 k_{2z}^2 + \rho_2^2 k_{1z}^2) \sin(k_{2z}h)} \right] \quad (45)$$

$$\langle T_{11}^{outside} \rangle = \langle T_{22}^{outside} \rangle = \frac{A_{c1} B_{c1}^* e^{-2jk_{2z}z} + A_{c1}^* B_{c1} e^{2jk_{2z}z}}{2\rho_1 c_1^2}, \langle T_{33}^{outside} \rangle \\ = \frac{A_{c1} A_{c1}^* + B_{c1} B_{c1}^*}{2\rho_1 c_1^2} \quad (49)$$

The corresponding equivalent stresses are:

$$u_{2z} = \frac{2l\rho_1 k_{1z} k_{2z} e^{j\omega t} [\rho_1 k_{2z} \sin(k_{2z}(z-l_3)) + \rho_1 k_{2z} \sin(k_{2z}z) + j\rho_2 k_{1z} \cos(k_{2z}(z-l_3)) - j\rho_2 k_{1z} \cos(k_{2z}z)]}{2\rho_1\rho_2 k_{1z}k_{2z} \cos(k_{2z}h) + j(\rho_1^2 k_{2z}^2 + \rho_2^2 k_{1z}^2) \sin(k_{2z}h)} \quad (46)$$

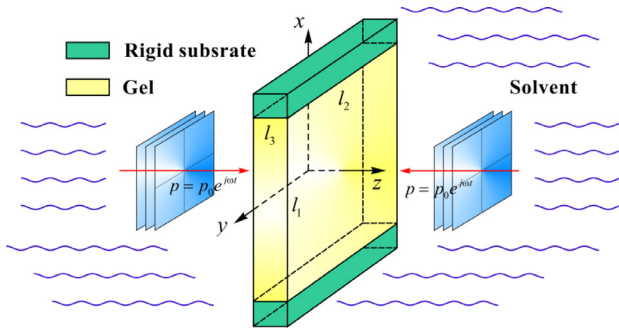
$$p_2 = \frac{2\omega l \rho_1 \rho_2 k_{1z} e^{j\omega t} [\rho_2 k_{1z} \sin(k_{2z}(z-l_3)) - \rho_2 k_{1z} \sin(k_{2z}z) + j\rho_1 k_{2z} \cos(k_{2z}(z-l_3)) + j\rho_1 k_{2z} \cos(k_{2z}z)]}{2\rho_1\rho_2 k_{1z}k_{2z} \cos(k_{2z}h) + j(\rho_1^2 k_{2z}^2 + \rho_2^2 k_{1z}^2) \sin(k_{2z}h)} \quad (47)$$

Applying the above solutions for velocity and acoustic fields, one obtains the acoustic stresses as:

$$\langle T_{11}^{inside} \rangle = \langle T_{22}^{inside} \rangle = \frac{A_{c2} B_{c2}^* e^{-2jk_{2z}z} + A_{c2}^* B_{c2} e^{2jk_{2z}z}}{2\rho_2 c_2^2}, \langle T_{33}^{inside} \rangle \\ = \frac{A_{c2} A_{c2}^* + B_{c2} B_{c2}^*}{2\rho_2 c_2^2} \quad (48)$$

$$t_1 = t_2 = \frac{1}{2\rho_2 c_2^2} \left\{ \frac{1}{2jk_{2z}l_3} [A_{c2}^* B_{c2} (e^{2jk_{2z}l_3} - 1) - A_{c2} B_{c2}^* (e^{-2jk_{2z}l_3} - 1)] \right\} \quad (50)$$

$$t_3 = \frac{A_{c2} A_{c2}^* + B_{c2} B_{c2}^*}{2\rho_2 c_2^2} - \frac{A_{c1} A_{c1}^* + B_{c1} B_{c1}^*}{2\rho_1 c_1^2} \quad (51)$$



**Fig. 3.** Schematic of a uniaxially constrained polymeric gel layer (attached with two rigid substrates) immersed in solvent and subjected to two counterpropagating acoustic waves  $p = p_0 e^{i k x}$ . The dry polymer with initial undeformed dimensions ( $L_1, L_2, L_3$ ) freely swells to become a gel with dimensions ( $l_1^i, l_2^i, l_3^i$ ) when immersed in solvent. Then the gel is fixed to the rigid substrates and struck by an acoustic input, deforming thereby to current state of dimensions ( $l_1, l_2, l_3$ ), with  $l_1^i = l_1$ .

where

$$A_{c1} = j\omega\rho_1 l e^{j k_{1z} l_3} \times \left( \frac{j(\rho_2^2 k_{1z}^2 - \rho_1^2 k_{2z}^2) \sin(k_{2z} l_3) + 2\rho_1 \rho_2 k_{1z} k_{2z}}{2\rho_1 \rho_2 k_{1z} k_{2z} \cos(k_{2z} l_3) + j(\rho_1^2 k_{2z}^2 + \rho_2^2 k_{1z}^2) \sin(k_{2z} l_3)} \right) \quad (52)$$

$$B_{c1} = j\omega\rho_1 l e^{-j k_{1z} l_3} \quad (53)$$

$$A_{c2} = j\omega\rho_1 \rho_2 k_{1z} l \times \left( \frac{e^{j k_{2z} l_3} (\rho_2 k_{1z} + \rho_1 k_{2z}) + (\rho_1 k_{2z} - \rho_2 k_{1z})}{2\rho_1 \rho_2 k_{1z} k_{2z} \cos(k_{2z} l_3) + j(\rho_1^2 k_{2z}^2 + \rho_2^2 k_{1z}^2) \sin(k_{2z} l_3)} \right) \quad (54)$$

$$B_{c2} = j\omega\rho_1 \rho_2 k_{1z} l \times \left( \frac{e^{-j k_{2z} l_3} (\rho_1 k_{2z} - \rho_2 k_{1z}) + (\rho_2 k_{1z} + \rho_1 k_{2z})}{2\rho_1 \rho_2 k_{1z} k_{2z} \cos(k_{2z} l_3) + j(\rho_1^2 k_{2z}^2 + \rho_2^2 k_{1z}^2) \sin(k_{2z} l_3)} \right) \quad (55)$$

Given the acoustic input and the prescribed uniaxial constraint of  $\lambda_1 = \lambda_p$ , the polymeric gel not only swells further but also acoustically deforms in the  $y$ - and  $z$ -directions (Fig. 3). Correspondingly, the normalized Cauchy stresses are given by:

$$\frac{\sigma_2 v}{kT} = Nv(\lambda_2 - \lambda_2^{-1})\lambda_1^{-1}\lambda_3^{-1} + [\ln(1 - \lambda_1^{-1}\lambda_2^{-1}\lambda_3^{-1}) + \lambda_1^{-1}\lambda_2^{-1}\lambda_3^{-1} + \chi\lambda_1^{-2}\lambda_2^{-2}\lambda_3^{-2}] - \frac{t_2 v}{kT} \quad (56)$$

$$\frac{\sigma_3 v}{kT} = Nv(\lambda_3 - \lambda_3^{-1})\lambda_1^{-1}\lambda_2^{-1} + [\ln(1 - \lambda_1^{-1}\lambda_2^{-1}\lambda_3^{-1}) + \lambda_1^{-1}\lambda_2^{-1}\lambda_3^{-1} + \chi\lambda_1^{-2}\lambda_2^{-2}\lambda_3^{-2}] - \frac{t_3 v}{kT} \quad (57)$$

In the absence of external mechanical loading, the normalized acoustic input  $p_0/\sqrt{\rho_0 c_0^2 kT/v}$  can be extracted as a function of the stretches  $\lambda_2$  and  $\lambda_3$  by setting  $\sigma_2 v/kT = 0$  and  $\sigma_3 v/kT = 0$ . The relationship between  $\lambda_2$  and  $\lambda_3$  is given in an implicit form as:

$$Nv[t_3(\lambda_2^2 - 1) - t_2(\lambda_3^2 - 1)] + \lambda_1 \lambda_2 \lambda_3 (t_3 - t_2) [\ln(1 - \lambda_1^{-1} \lambda_2^{-1} \lambda_3^{-1}) + \lambda_1^{-1} \lambda_2^{-1} \lambda_3^{-1} + \chi \lambda_1^{-2} \lambda_2^{-2} \lambda_3^{-2}] = 0 \quad (58)$$

Finally, the acoustomechanical response of the polymeric gel can be obtained by solving the nonlinear equations of (56)–(58).

Fig. 4 plots the nonlinear acoustomechanical deformation of polymeric gel under prescribed uniaxial constraints of  $\lambda_1 = 1, 2, 3$

and 4. Under different uniaxial constraints, the gel exhibits different initial swelling stretches in the two unconstrained directions when  $p_0/\sqrt{\rho_0 c_0^2 kT/v} = 0$ . As the stretch is increased, the acoustic input-stretch curves show periodical variations but slightly different extremal points under different uniaxial constraints. This is attributable to the nonlinear relationship between the stretches in Eq. (58), as presented in Fig. 4(b). The quasi-periodical variation of stretch  $\lambda_2$  with stretch  $\lambda_3$  is induced by the periodical variation of acoustic stress with stretch  $\lambda_3$ . The larger the prescribed stretch  $\lambda_1$ , the smaller the stretch  $\lambda_2$  at a given stretch  $\lambda_3$ . The molecules volume ration  $vC$  plotted in Fig. 4(c) as a function of  $\lambda_3$  also shows quasi-periodical variation. The larger the prescribed stretch  $\lambda_1$ , the more the solvent molecules that the polymers imbibe at the same stretch  $\lambda_3$ . The variation of acoustic input with molecules volume ratio is presented in Fig. 4(d), which exhibits a nonlinear chaos behavior for molecule migration under acoustic loading. Moreover, this chaos behavior is sensitive to the initial molecules volume ratio. The periodical trend of the acoustic input-stretch curve and the quasi-periodical trend of the  $vC$ - $\lambda_3$  curve contribute to the repeated and increased loop configuration of the chaos curve.

## 6. Unconstrained acoustical actuation

As a final example, consider a freestanding polymeric gel layer immersed in solvent and subjected to two counterpropagating acoustic waves, as illustrated in Fig. 5. The gel initially has isotropic swelling stretches since no constraint is enforced. When subjected to the acoustic inputs, the gel deforms further due to acoustic stressing. The wave propagation governed by elastodynamical equations satisfies the same boundary conditions as in the uniaxial constraint case. Therefore, the velocity and acoustic fields as well as the acoustic stresses have the same expressions as those presented in Section 5.

Upon acoustical actuation, the polymeric gel can imbibe more solvent molecules and swell to a larger deformation since the acoustic stress can help overcome the osmotic pressure. When the acoustic stress and elastic stress balance with the osmotic pressure, the gel reaches a steady state. This acoustically actuated swelling behavior is governed by:

$$\frac{\sigma_1 v}{kT} = Nv(\lambda_1 - \lambda_1^{-1})\lambda_2^{-1}\lambda_3^{-1} + [\ln(1 - \lambda_1^{-1}\lambda_2^{-1}\lambda_3^{-1}) + \lambda_1^{-1}\lambda_2^{-1}\lambda_3^{-1} + \chi\lambda_1^{-2}\lambda_2^{-2}\lambda_3^{-2}] - \frac{t_1 v}{kT} \quad (59)$$

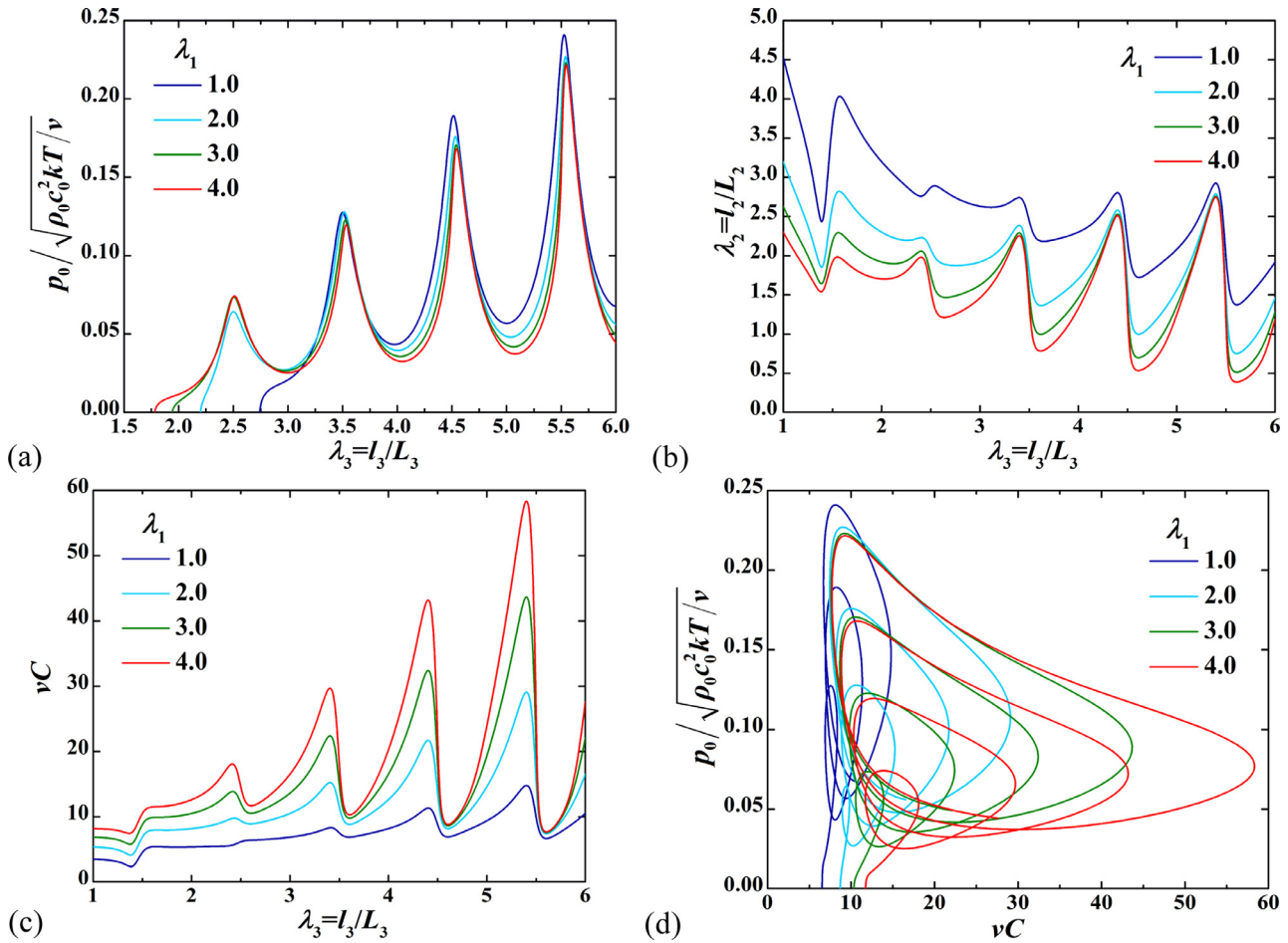
$$\frac{\sigma_2 v}{kT} = Nv(\lambda_2 - \lambda_2^{-1})\lambda_1^{-1}\lambda_3^{-1} + [\ln(1 - \lambda_1^{-1}\lambda_2^{-1}\lambda_3^{-1}) + \lambda_1^{-1}\lambda_2^{-1}\lambda_3^{-1} + \chi\lambda_1^{-2}\lambda_2^{-2}\lambda_3^{-2}] - \frac{t_2 v}{kT} \quad (60)$$

$$\frac{\sigma_3 v}{kT} = Nv(\lambda_3 - \lambda_3^{-1})\lambda_1^{-1}\lambda_2^{-1} + [\ln(1 - \lambda_1^{-1}\lambda_2^{-1}\lambda_3^{-1}) + \lambda_1^{-1}\lambda_2^{-1}\lambda_3^{-1} + \chi\lambda_1^{-2}\lambda_2^{-2}\lambda_3^{-2}] - \frac{t_3 v}{kT} \quad (61)$$

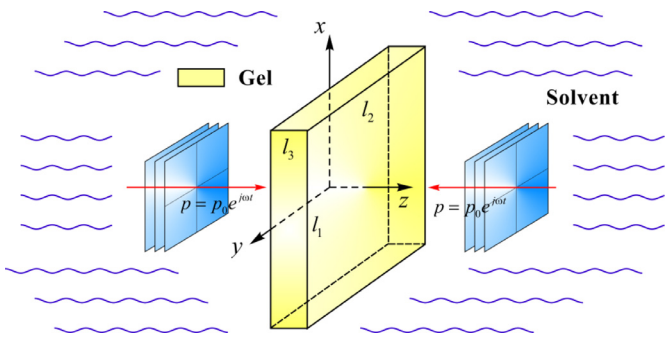
where the gel stretches satisfy:

$$Nv[t_3(\lambda_1^2 - 1) - t_1(\lambda_3^2 - 1)] + [\lambda_1^2 \lambda_3 \ln(1 - \lambda_1^{-2} \lambda_3^{-1}) + 1 + \chi \lambda_1^{-2} \lambda_3^{-1}](t_3 - t_1) = 0, \lambda_1 = \lambda_2 \quad (62)$$

Since no external mechanical force is applied,  $\sigma_1 v/kT = 0$ ,  $\sigma_2 v/kT = 0$  and  $\sigma_3 v/kT = 0$ . The normalized acoustic input  $p_0/\sqrt{\rho_0 c_0^2 kT/v}$  can thence be extracted as a function of gel stretches. Fig. 6 presents the nonlinear acoustomechanical gel response for selected values of mixing parameter:  $\chi = 0.2, 0.3, 0.4$  and  $0.5$ . As shown in Fig. 6(a), as the mixing parameter changes, the gel exhibits different initial swelling stretches when  $p_0/\sqrt{\rho_0 c_0^2 kT/v} = 0$ . The larger the mixing parameter is, the smaller



**Fig. 4.** Nonlinear acoustomechanical deformation of a polymeric gel layer under selected uniaxial constraints of  $\lambda_1=1, 2, 3$  and  $4$ : (a) acoustic input  $p_0/\sqrt{\rho_0 c_0^2 kT/\nu}$  plotted as a function of out-of-plane stretch  $\lambda_3$ ; (b) in-plane stretch  $\lambda_2$  plotted as a function of  $\lambda_3$ ; (c) ratio of molecules volume over dry polymers  $\nu C$  plotted as a function of  $\lambda_3$ ; (d)  $p_0/\sqrt{\rho_0 c_0^2 kT/\nu}$  plotted as a function of  $\nu C$ .



**Fig. 5.** Schematic of a freestanding polymeric gel layer immersed in solvent and subjected to two counterpropagating acoustic waves  $p=p_0 e^{i\omega t}$ . When immersed in solvent, the dry polymer with initial undeformed dimensions  $(L_1, L_2, L_3)$  freely swells to become a gel with dimensions  $(l_1^s, l_2^s, l_3^s)$ . Then the gel is struck by an acoustic input and deforms to the current state of dimensions  $(l_1, l_2, l_3)$ .

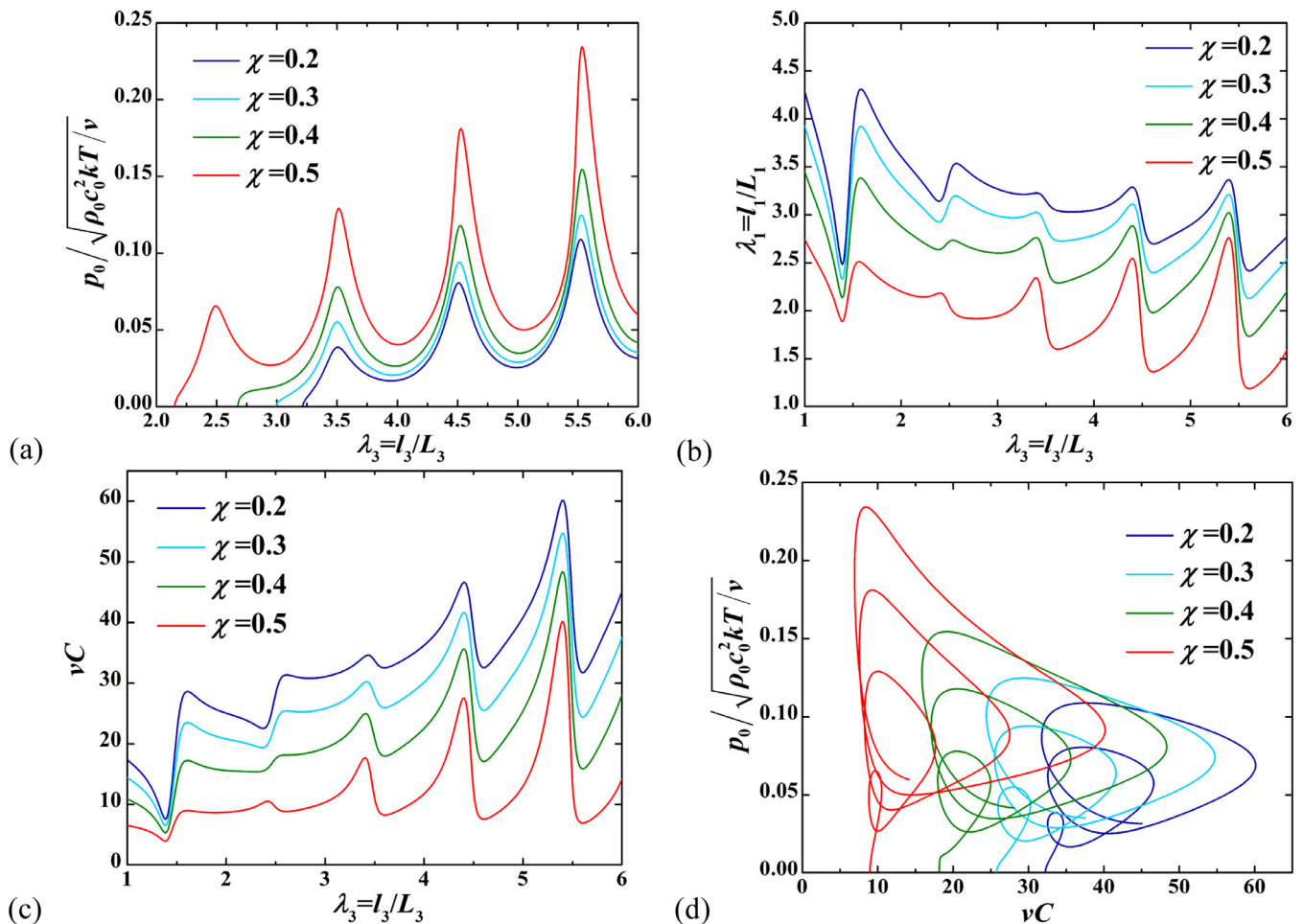
the initial swelling stretch is, because the enthalpy of mixing with a larger mixing parameter tends to motivate the molecules to leave the gel. The acoustic input-stretch curves display periodical variations when the stretch is increased, as the acoustic stress is a periodical function of the stretch. It is also found that the larger the mixing parameter, the smaller the acoustic input needed to maintain the same stretch. As illustrated in Fig. 6(b) and (c), both the stretch  $\lambda_1$  and molecule volume ratio  $\nu C$  are quasi-periodical functions of the stretch  $\lambda_3$ . At the same stretch  $\lambda_3$ , the larger the

mixing parameter is, the smaller the stretch  $\lambda_1$  and molecule volume ratio  $\nu C$  are. As observed in Fig. 6(d), the variation of acoustic input as a function of molecules volume ratio shows a nonlinear chaos behavior, which is sensitive to the mixing parameter or initial swelling stretch. The periodical feature of the acoustic input-stretch curve and the quasi-periodical feature of the  $\nu C$ - $\lambda_3$  curve play a dominant role in the nonlinear chaos behavior, namely, the repeated and increased loop configuration of the curves.

## 7. Concluding remarks

A nonlinear acoustomechanical field theory is developed for polymeric gels immersed in external solvent by combining the acoustic radiation stress theory and the nonlinear elasticity theory of gels, with diffusion mass transport of solvent molecules in and out of the gel accounted for. For convenience of solving the elastodynamical equations that govern acoustic wave propagation, this field theory is established in Eulerian coordinates. Therefore, all the state variables are presented with their own true values, which can be readily transformed to their nominal values. Nonlinear gel elasticity is accounted for by utilizing the Flory–Rehner free energy functions for network stretching and molecule-polymer mixing. The developed field theory is applied to first calculate the velocity and acoustic fields as well as the distribution of acoustic radiation stresses inside and outside the gel, and then analyze its nonlinear acoustomechanical behavior. Specifically, biaxial constraint, uniaxial constraint and freestanding cases are considered.





**Fig. 6.** Nonlinear acoustomechanical deformation of a freestanding polymeric gel layer for selected mixing parameters: (a) acoustic input  $p_0/\sqrt{\rho_0 c_0^2 kT/\nu}$  plotted as a function of out-of-plane stretch  $\lambda_3$ ; (b) in-plane stretch  $\lambda_1$  plotted as a function of  $\lambda_3$ ; (c) ratio of molecules volume over dry polymers  $vC$  plotted as a function of  $\lambda_3$ ; (d)  $p_0/\sqrt{\rho_0 c_0^2 kT/\nu}$  plotted as a function of  $vC$ .

The periodical response and nonlinear chaos behaviors of acoustically actuated polymeric gels are revealed. Results presented in this study are helpful for innovative design of acoustically triggered sensors and actuators made of polymeric gels, and can enlighten the application of ultrasound in biomedical engineering.

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