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A unified model for piezocomposites with non-piezoelectric matrix and piezoelectric ellipsoidal inclusions

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Abstract

In this paper, the closed-form solutions of the electroelastic Eshelby's tensors of a piezoelectric ellipsoidal inclusion in an infinite non-piezoelectric matrix are obtained via the Green's function technique. Based on the generalized Budiansky's energy-equivalence framework and the closed-form solutions of the electroelastic Eshelby's tensors, a unified model for multiphase piezocomposites with the non-piezoelectric matrix and piezoelectric inclusions is set up. The closed-form solutions of the effective electroelastic moduli of piezocomposites are also obtained. The unified model has a rigorous but simple form, which can describe the multiphase piezocomposites with different connectivities, such as 0–3, 1–3, 2–2, 2–3, 3–3 connectivities, etc. It can also describe the effects of non-interaction and interaction among the inclusions. As examples, the closed-form solutions of the effective electroelastic moduli are given by means of the dilute solution for the 0-3 piezocomposite with transversely isotropic piezoelectric spherical inclusions and by means of the dilute solution and the Mori-Tanaka's method for the 1-3 piezocomposite with two kinds of transversely isotropic piezoelectric cylindrical inclusions. The predicted results are compared with experimental data, which shows that the theoretical curves calculated by means of the Mori-Tanaka's method agree quite well with the experimental values, but the theoretical curves obtained by the dilute solution agree well with the experimental values only when the volume fraction of the ceramic inclusion is less than 0.3. The results in this paper can be used to analyze and design the multiphase piezocomposites. © 1999 Elsevier Science Ltd. All rights reserved.

1. Introduction

With the development of information industry and the appearance of smart materials and smart structures (Jiang et al., 1994), material science has entered a new era. Conventional materials are not able to satisfy modern engineering requirements. Since piezocomposites provide material properties superior to conventional piezoelectric materials, piezocomposites have become attractive

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candidates for use in many fields. In the past few years, much work has been done in the analysis and prediction of the effective properties of piezocomposites based on mesomechanics.

The micromechanical characterization and analysis of piezocomposites were launched by the Newnham et al.'s (1978) connectivity theory, which is based on the combination of mechanics of materials type parallel and series models. These analyses were extended by Banno (1983) to consider discontinuous reinforcement through a cubes approach. A different route was taken by Smith and Auld (1991) in the analysis of continuous fiber-reinforced piezoelectric composites. A more rigorous treatment of the coupled electroelastic fields in a piezoelectric concentric cylinder geometry was performed by Grekov et al. (1989). Benveniste (1992, 1993a, b, 1994) obtained the effective eigenstress and spontaneous for multiphase composites with the arbitrary phase geometry and the effective properties for fibrous piezocomposites based on the use of virtual work theorems. But the closed-form solutions of the effective electroelastic properties for multiphase piezocomposites with the arbitrary phase geometry have not been obtained yet.

Wang (1992) studied the problem via the Green's function technique and obtained integral expressions for the constraint–strain and constraint–electric–field of a spherical inclusion in an infinite piezoelectric matrix, but the integral expressions are very complicated, thus the closed-form solutions of the constraint–strain and constraint–electric–field are too difficult to obtain even though the matrix is non-piezoelectric. Although Wang (1992) gave the closed-form solutions of the constraint–electric–field of a transversely isotropic cylindrical inclusion in an infinite non-piezoelectric matrix and the dilute solutions of the effective electroelastic moduli of 1–3 piezocomposites, the solutions considered the interaction among inclusions were not obtained. Based on the contour integral representation of the Green's function derived by Deeg (1980), Dunn and Taya (1993a, b) made a significant contribution to the analysis of the effective behavior of the piezocomposites when the interaction among reinforcements was considered, but the closed-form solutions of the effective electroelastic moduli of the piezocomposites when the interaction among reinforcements was considered, but the closed-form solutions of the effective electroelastic moduli of the piezocomposites with ellipsoidal inclusions have not been given due to the fact that the electroelastic Eshelby's tensors are given by a complicated integral form.

From the view of application, many piezocomposites are made up of piezoelectric inclusions and the non-piezoelectric matrix. For example, the sensors made of piezoelectric materials, which are widely used in smart materials or smart structures (Jiang et al., 1994), are generally embedded in a non-piezoelectric matrix in which the elastic fields and the electric fields are decoupled. As another example, some kinds of very important materials, such as relaxor ferroelectric materials, are made up of a ferroelectric phase and a paraelectric phase, which can be considered as composites with the inclusions and matrix. That is, the inclusions are the ferroelectric phase that has electromechanical coupling behavior and the matrix is the paraelectric phase which has no electromechanical coupling behavior. In this case, the problem can be simplified significantly.

This paper describes a unified model for multiphase piezocomposites with non-piezoelectric matrix and piezoelectric ellipsoidal inclusions and obtains the closed-form solutions of the effective electroelastic properties of this kind of piezocomposites. The unified model has a rigorous but simple form, which can describe the multiphase piezocomposites with different connectivities, such as 0-3, 1-3, 2-2, 2-3, 3-3 connectivities, etc. It can also describe the non-interaction and interaction among the inclusions.

The present paper is divided into five sections:

(1) The closed-form solutions of the electroelastic Eshelby's tensors of a piezoelectric ellipsoidal

inclusion in an infinite non-piezoelectric matrix are obtained via a Green's function technique.

- (2) Based on the generalized Budiansky's energy-equivalence framework (Budiansky, 1965) and the results of the section (1), a unified model for the multiphase piezocomposites is set up and the closed-form solutions of the effective electroelastic moduli of the multiphase piezocomposites with ellipsoidal inclusions are given.
- (3) The predicted results are compared with experimental data, which shows that the theoretical values agree quite well with the experimental ones.
- (4) It is analyzed in detail for the 1–3 piezocomposite with two kinds of transversely isotropic piezoelectric cylindrical inclusions, in which one of the inclusions is aligned in the positive direction and another is in the negative direction.
- (5) In the appendices, the closed-form solutions of the effective electroelastic moduli are given by means of the dilute solution for the 0–3 piezocomposite with the transversely isotropic piezoelectric spherical inclusion and by the dilute solution and the Mori–Tanaka's method for the 1–3 piezocomposite with two kinds of transversely isotropic piezoelectric cylindrical inclusions.

2. The single inclusion and inhomogeneity

There is a region Ω (the linear piezoelectric inclusion) in an infinite homogeneous linear nonpiezoelectric medium *D*, the far fields are exposed with the uniform strain and electric field. It is asked what the elastic state and the electric field of the inclusion are.

We shall solve this problem with the help of the Green's function technique. The constitutive equations for a linear piezoelectric inclusion is (Maugin, 1988)

$$\boldsymbol{\sigma} = \boldsymbol{C}^* : \nabla \boldsymbol{u} + (\boldsymbol{e}^*)^T \cdot \nabla \boldsymbol{\phi} \quad \text{in } \Omega \tag{1}$$

$$\boldsymbol{D} = \boldsymbol{e}^* : \nabla \boldsymbol{u} - \boldsymbol{k}^* \cdot \nabla \phi \quad \text{in } \Omega \tag{2}$$

where C^* is the elastic moduli tensor, e^* the piezoelectric moduli tensor and k^* the dielectric permittivity, respectively. The superscript '*' refers to the material properties of the piezoelectric inclusion. ϕ and u are the electric potential and elastic displacement. D and σ are the electric displacement and the elastic stress. ∇ is Laplacian operator and $(e^T)_{klp} = e_{pkl}$. Tensors and vectors are denoted by italic bold face letters, inner products by the notation: $\mathbf{a} \cdot \mathbf{x} = a_{ij}x_j$ and $A : \mathbf{a} = A_{ijkl}a_{kl}$, and tensor products by notation: $\mathbf{ab} = a_i b_i$.

Bearing in mind that there is no electromechanical coupling in the matrix, one has the constitutive equations in the matrix as

$$\boldsymbol{\sigma} = \boldsymbol{C} : \nabla \boldsymbol{u} \quad \text{in } \boldsymbol{D} - \boldsymbol{\Omega} \tag{3}$$

$$\boldsymbol{D} = -\boldsymbol{k} \cdot \nabla \phi \quad \text{in } \boldsymbol{D} - \boldsymbol{\Omega} \tag{4}$$

where C and k are the elastic moduli tensor and the dielectric permittivity of the matrix. Because the matrix is non-piezoelectric, we have e = 0. By introducing the following characteristic function

$$h(\mathbf{x}) = \begin{cases} 1 & \mathbf{x} \in \Omega \\ 0 & \text{otherwise} \end{cases}$$
(5)

the elastic, piezoelectric tensors and the dielectric permittivity of the inclusion can be written as

$$C^* = C + C^{\circ}h(x), \quad e^* = e + e^{\circ}h(x) = e^*h(x), \quad k^* = k + k^{\circ}h(x)$$
(6)

where

$$C^{\circ} = C^* - C, \quad e^{\circ} = e^* - e = e^*, \quad k^{\circ} = k^* - k$$
 (7)

From eqns (6), (7), the eqns (1), (3) and (2), (4) can be written as

$$\boldsymbol{\sigma} = \boldsymbol{C} : \nabla \boldsymbol{u} + [\boldsymbol{C}^{\circ} : \nabla \boldsymbol{u} + (\boldsymbol{e}^{\circ})^{T} \cdot \nabla \phi] h(\boldsymbol{x}) \quad \text{in } D$$
(8)

$$\boldsymbol{D} = -\boldsymbol{k} \cdot \nabla \phi + [\boldsymbol{e}^{\circ} : \nabla \boldsymbol{u} - \boldsymbol{k}^{\circ} \cdot \nabla \phi] h(\boldsymbol{x}) \quad \text{in } D$$
(9)

If the free charges and body forces do not exist, the equilibrium equations for both the inclusion and the matrix are

$$\nabla \cdot \boldsymbol{\sigma} = \boldsymbol{0} \tag{10}$$

$$\nabla \cdot \boldsymbol{D} = 0 \tag{11}$$

Substitution of eqns (8) and (9) into eqns (10) and (11) yields

$$C_{ijkl}u_{k,lj} = -[C^0_{ijkl}u_{k,l} + e^0_{mij}\phi_{,m})h(\mathbf{x})]_{,j}$$
(12)

$$k_{im}\phi_{,mi} = [(e_{ikl}^{0}u_{k,l} - k_{im}^{0}\phi_{,m})h(\mathbf{x})]_{,i}$$
(13)

The right terms of eqns (12) and (13) can be regard as body forces and free charges which are applied on the matrix. Since the matrix is non-piezoelectric, two Green's functions G^{μ} and G^{ϕ} are introduced as follows

$$C_{ijkl}G^{u}_{kp,lj}(\boldsymbol{x}-\boldsymbol{x}') = -\delta_{ip}\delta(\boldsymbol{x}-\boldsymbol{x}')$$
(14)

$$k_{im}G^{\phi}_{,im}(\mathbf{x}-\mathbf{x}') = \delta(\mathbf{x}-\mathbf{x}') \tag{15}$$

The Green's function in eqn (14) is that of the perfectly elastic problem which can be obtained as follows (Mura, 1987)

$$G_{ij}^{u}(\mathbf{x} - \mathbf{x}') = (2\pi)^{-3} \int_{-\infty}^{+\infty} N_{ij}(\xi) D^{-1}(\xi) \exp\{i\xi \cdot (\mathbf{x} - \mathbf{x}')\} d\xi$$
(16)

where N_{ij} are cofactors of K_{ij} , $D(\xi)$ is the determinant of K_{ij} and $K_{ik}(\xi) = C_{ijkl}\xi_j\xi_l$.

The Green's function in eqn (15) is that of the perfectly dielectric problem which can be given by (Fan, 1995)

$$G^{\phi}(\mathbf{x} - \mathbf{x}') = -(2\pi)^{-3} \int_{-\infty}^{+\infty} (k_{ij}\xi_i\xi_j)^{-1} \exp\left\{i\xi \cdot (\mathbf{x} - \mathbf{x}')\right\} d\xi$$
(17)

Equations (12) and (13) can be expressed by means of the Green's functions in the forms

$$u_{p} = u_{p}^{0} + \int_{-\infty}^{+\infty} G_{pi}^{u}(\mathbf{x} - \mathbf{x}') [(C_{ijkl}^{0}u_{k,l} + e_{mij}^{0}\phi_{,m})h(\mathbf{x}')]_{,j'} d\mathbf{x}'$$

$$= u_{p}^{0} + \int_{\Omega} G_{pi,j}^{u}(\mathbf{x} - \mathbf{x}') [C_{ijkl}^{0}u_{k,l} + e_{mij}^{0}\phi_{,m}] d\mathbf{x}'$$
(18)

$$\phi = \phi^{0} + \int_{-\infty}^{+\infty} G^{\phi}(\mathbf{x} - \mathbf{x}') [(e^{0}_{ikl}u_{k,l} - k^{0}_{ij}\phi_{,j})h(\mathbf{x}')]_{,j'} d\mathbf{x}'$$

= $\phi^{0} + \int_{\Omega} G^{\phi}_{,i}(\mathbf{x} - \mathbf{x}') [e^{0}_{ikl}u_{k,l} - k^{0}_{ij}\phi_{,j}] d\mathbf{x}'$ (19)

where u_p^0 and ϕ^0 are homogeneous solutions of eqns (12) and (13). In the derivation of eqns (18) and (19), the properties of the characteristic function and the relations

$$G_{p_{i,j}}^{u}(\mathbf{x} - \mathbf{x}') = -G_{p_{i,j}}^{u}(\mathbf{x} - \mathbf{x}')$$
(20)

$$G^{\phi}_{,i'}(\mathbf{x} - \mathbf{x}') = -G^{\phi}_{,i}(\mathbf{x} - \mathbf{x}') \tag{21}$$

are used. By differentiating eqns (18) and (19), the equations of the elastic strain field and electric field can be obtained in the form

$$\varepsilon_{ij} = \frac{1}{2} (u_{i,j} + u_{j,i}) = \varepsilon_{ij}^0 + \frac{1}{2} \int_{\Omega} \left[G^u_{im,nj} (\mathbf{x} - \mathbf{x}') + G^u_{jm,ni} (\mathbf{x} - \mathbf{x}') \right] \times (C^0_{mnkl} u_{k,l} - e^0_{pmn} E_p) \mathrm{d}\mathbf{x}'$$
(22)

$$E_{i} = -\phi_{,i} = E_{i}^{0} - \int_{\Omega} G_{,ji}^{\phi}(\mathbf{x} - \mathbf{x}') (e_{jkl}^{0} u_{k,l} + k_{jp}^{0} E_{p}) \mathrm{d}\mathbf{x}'$$
(23)

In the derivation of eqns (22) and (23), $C_{ijkl}u_{k,l} = C_{ijkl}\varepsilon_{kl}$ and $E_i = -\phi_{,i}$ are used.

Wang (1992), Chen (1993, 1994), Dunn and Taya (1993a) demonstrated that if a linear piezoelectric ellipsoidal inclusion in an infinite linear piezoelectric matrix is subjected to the far fields with the uniform strain and electric field, the constraint strain and the constraint electric field inside the inclusion remain uniform. The problem in this paper is the special case of the above general problem and it can be demonstrated that the result is the same as that of the above general problem. Equations (22) and (23) are applicable to all over the domain *D*. Let us consider the case when point \mathbf{x} is inside the ellipsoidal inclusion, i.e. $\mathbf{x} \in \Omega$. When ε^0 and \mathbf{E}^0 are uniform, ε^1 and \mathbf{E}^1 are also uniform. The superscript 'I' refers to the field values for interior points. Equations (22) and (23) can be written as

$$(\boldsymbol{I} + \boldsymbol{S} : \boldsymbol{C}^{-1} : \boldsymbol{C}^{0}) : \boldsymbol{\varepsilon}^{\mathrm{I}} = \boldsymbol{\varepsilon}^{0} + \boldsymbol{S} : \boldsymbol{C}^{-1} : (\boldsymbol{e}^{0})^{T} \cdot \boldsymbol{E}^{\mathrm{I}}$$
(24)

$$(\mathbf{i} + \mathbf{s} \cdot \mathbf{k}^{-1} \cdot \mathbf{k}^0) \cdot \mathbf{E}^{\mathrm{I}} = \mathbf{E}^0 - \mathbf{s} \cdot \mathbf{k}^{-1} \cdot \mathbf{e}^0 : \varepsilon^{\mathrm{I}}$$
⁽²⁵⁾

where

$$S_{ijkl} = -\frac{1}{2} \int_{\Omega} C_{mnkl} [G^{u}_{im,nj}(\mathbf{x} - \mathbf{x}') + G^{u}_{jn,mi}(\mathbf{x} - \mathbf{x}')] d\mathbf{x}'$$
(26)

is the Eshelby's tensor of the perfectly elastic inclusion problem (Mura, 1987) and

$$s_{ij} = \int_{\Omega} k_{mj} G^{\phi}_{,im}(\mathbf{x} - \mathbf{x}') \mathrm{d}\mathbf{x}'$$
(27)

is the Eshelby's tensor of the perfectly dielectric inclusion problem (Fan and Qin, 1995). Where I and i are four-order and two-order unit tensors, respectively. $(\cdot)^{-1}$ represents the inverse of the indicated quantity.

From eqns (24) and (25), we can obtain the relations among ε^0 , E^0 , ε^I and E^I

$$\boldsymbol{\varepsilon}^{\mathrm{I}} = \mathscr{H}^{1} : \boldsymbol{\varepsilon}^{0} + \mathscr{H}^{2} \cdot \boldsymbol{E}^{0}, \quad \boldsymbol{E}^{\mathrm{I}} = \mathscr{H}^{3} : \boldsymbol{\varepsilon}^{0} + \mathscr{H}^{4} \cdot \boldsymbol{E}^{0}$$
⁽²⁸⁾

where

$$\mathscr{H}^{1} = (\boldsymbol{I} + \boldsymbol{\alpha} \cdot \boldsymbol{\beta})^{-1} : \boldsymbol{A}, \quad \mathscr{H}^{2} = (\boldsymbol{I} + \boldsymbol{\alpha} \cdot \boldsymbol{\beta})^{-1} : \boldsymbol{\alpha} \cdot \boldsymbol{B}$$
(29a)

$$\mathscr{H}^{3} = -(\mathbf{i} + \boldsymbol{\beta} : \boldsymbol{\alpha})^{-1} \cdot \boldsymbol{\beta} : \boldsymbol{A}, \quad \mathscr{H}^{4} = (\mathbf{i} + \boldsymbol{\beta} : \boldsymbol{\alpha})^{-1} \cdot \boldsymbol{B}$$
(29b)

and

$$A = (I + S: C^{-1}: C^{0})^{-1}, \quad B = (i + s \cdot k^{-1} \cdot k^{0})^{-1}$$
(30a)

$$\boldsymbol{\alpha} = \boldsymbol{A} : \boldsymbol{S} : \boldsymbol{C}^{-1} : (\boldsymbol{e}^*)^T, \quad \boldsymbol{\beta} = \boldsymbol{B} \cdot \boldsymbol{s} \cdot \boldsymbol{k}^{-1} \cdot \boldsymbol{e}^*$$
(30b)

where A is the strain concentration-tensor of the perfectly elastic inclusion problem and B is the electric field concentration-tensor of the perfectly dielectric inclusion problem. Equations (29a) can also be written as

$$\mathscr{H}^{1} = \mathbf{A} + \boldsymbol{\alpha} \cdot \mathscr{H}^{3}, \quad \mathscr{H}^{2} = \boldsymbol{\alpha} \cdot \mathscr{H}^{4}$$
(29c)

Equations (29), (30) are the relations among the electroelastic Eshelby's tensors, the perfectly elastic Eshelby's tensors and the perfectly dielectric Eshelby's tensors. Equations (28) are the closed-form solutions of the constraint strain field and the constraint electric field inside the inclusion. Since the perfectly elastic Eshelby's tensors and the perfectly dielectric Eshelby's tensors have been obtained already by means of theory of elasticity and electrodynamics, respectively, the closed-form electroelastic Eshelby's tensors of a piezoelectric ellipsoidal inclusion in an infinite non-piezoelectric matrix can be determined easily.

It is known from the above equations that the most important parameters are α and β in the electromechanical coupling problem. α , three-order tensor whose dimension is m/V, is related to the elastic properties of the inclusion and the matrix, the piezoelectric property and the shape of the inclusion. β , three-order tensor whose dimension is V/m, is concerned with the dielectric properties of the inclusion and the matrix, the piezoelectric property and the shape of the inclusion.

When the inclusion material is non-piezoelectric, i.e. $e^* = 0$, \mathcal{H}^1 , and \mathcal{H}^4 become *A* and *B*, respectively and \mathcal{H}^2 and \mathcal{H}^3 are zero. Now, the problem is separated to be two problems of the perfectly elastic inhomogeneous inclusion and the perfectly dielectric inhomogeneous inclusion. If

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the material is homogeneous and non-piezoelectric, \mathcal{H}^1 and \mathcal{H}^4 become four-order and two-order unit tensors, I and i, respectively.

It should be emphasized that the perfectly elastic and the perfectly dielectric Eshelby's tensors in eqns (29), (30) are relative only to the properties of the matrix, not to those of the inclusion. Generally, since the matrix is isotropic, many problems can be greatly simplified.

3. A unified model for multiphase piezocomposites with non-piezoelectric matrix and piezoelectric ellipsoidal inclusions

Budiansky (1965) established the energy-equivalence framework for composite materials, but the analysis is limited only to the self-consistent method. Huang et al. (1995) developed a unified energy approach based on the Budiansky's energy-equivalence framework to be suitable for several micromechanics models of composite materials, such as the dilute solution, self-consistent method, generalized self-consistent method, and Mori–Tanaka's method. A unified energy approach which can be used in piezocomposites is developed in this paper.

Consider a large cube of a multiphase piezocomposite composed of a coherent non-piezoelectric mixture of several piezoelectric materials. The spatial distributions of the phases are assumed to be such that the piezocomposite is homogeneous. There are total of N phases of piezoelectric inclusions embedded in the non-piezoelectric matrix. The constitutive equations in the inclusions and the matrix are equations (1)–(4), but the material constants in the *n*-th inclusion should be replaced by $C^{(n)}$, $e^{(n)}$, $k^{(n)}$ (n = 1, ..., N). The superscript 'n' refers to the material constants of the *n*-phase. The volume concentrations of the individual phases and the matrix are f_n (n = 1, ..., N) and

$$1-\sum_{n=1}^{N}f_{n},$$

respectively. The piezocomposite can be characterized by the following constitutive relations

$$\boldsymbol{\sigma} = \boldsymbol{\bar{C}} : \boldsymbol{\varepsilon} - (\boldsymbol{\bar{e}})^T \cdot \boldsymbol{E}$$
(31)

$$\boldsymbol{D} = \boldsymbol{\bar{e}} : \boldsymbol{\varepsilon} + \boldsymbol{\bar{k}} \cdot \boldsymbol{E} \tag{32}$$

To determine \bar{C} , \bar{e} and \bar{k} , apply a uniform strain ϵ^0 and a uniform electric field E^0 to the surface of the cube of the piezocomposite. So the boundary conditions can be expressed by the following form

$$\boldsymbol{u}(S) = \boldsymbol{\varepsilon}^0 \cdot \boldsymbol{x}, \quad \boldsymbol{\phi}(S) = -\boldsymbol{E}^0 \cdot \boldsymbol{x} \tag{33}$$

where x is the Cartesian coordinate system and S is the surface of the cube. When the independent variables are the strain and the electric field, the energy function should be electric Gibbs' energy which can be defined by (Lines and Glass, 1977)

$$\mathscr{G}(\boldsymbol{\varepsilon}, \boldsymbol{E}) = \mathscr{G}^{1}(\boldsymbol{\varepsilon}, \boldsymbol{E}) - \mathscr{G}^{2}(\boldsymbol{\varepsilon}, \boldsymbol{E})$$
(34)

where

$$\mathscr{G}^{1}(\boldsymbol{\varepsilon}, \boldsymbol{E}) = \frac{1}{2V} \int_{V} \boldsymbol{\varepsilon} : \boldsymbol{\sigma}(\boldsymbol{\varepsilon}, \boldsymbol{E}) \mathrm{d}V$$
(35)

$$\mathscr{G}^{2}(\boldsymbol{\varepsilon}, \boldsymbol{E}) = \frac{1}{2V} \int_{V} \boldsymbol{E} \cdot \boldsymbol{D}(\boldsymbol{\varepsilon}, \boldsymbol{E}) \mathrm{d}V$$
(36)

and V is the total volume of the piezocomposite.

For $\mathscr{G}^1(\varepsilon, \mathbf{E})$, we have

$$2V\mathscr{G}^{1}(\boldsymbol{\varepsilon},\boldsymbol{E}) = \int_{V} \sigma_{ij}\varepsilon_{ij} \, \mathrm{d}V = \int_{V} \sigma_{ij}u_{j,i} \, \mathrm{d}V = \int_{V} (\sigma_{ij}u_{j})_{,i} \, \mathrm{d}V = \oint_{S} \sigma_{ij}u_{j}n_{i} \, \mathrm{d}S$$
$$= \oint_{S} \sigma_{ij}\varepsilon_{jm}^{0}x_{m}n_{i} \, \mathrm{d}S = \varepsilon_{jm}^{0} \oint_{S} \sigma_{ij}x_{m}n_{i} \, \mathrm{d}S = \varepsilon_{jm}^{0} \int_{V} (\sigma_{ij}x_{m})_{,i} \, \mathrm{d}V$$
$$= \varepsilon_{jm}^{0} \int_{V} \sigma_{ij}x_{m,i} \, \mathrm{d}V = \varepsilon_{jm}^{0} \int_{V} \sigma_{ij}\delta_{mi} \, \mathrm{d}V = \varepsilon_{ij}^{0} \int_{V} \sigma_{ij} \, \mathrm{d}V \tag{37}$$

where *n* is the outward unit normal on the surface *S* of the cube. The strain–displacement relation, $\varepsilon_{ij} = (u_{i,j} + u_{j,i})/2$, symmetry of the stress, $\sigma_{ij} = \sigma_{ji}$, equilibrium of stress, $\sigma_{ij,i} = 0$, the boundary condition (33) and the divergence theorem are used in eqn (37).

For identity, the stress σ is decomposed as

$$\boldsymbol{\sigma} = \boldsymbol{C} : \boldsymbol{\varepsilon} + (\boldsymbol{\sigma} - \boldsymbol{C} : \boldsymbol{\varepsilon}) \tag{38}$$

where C is the elastic moduli tensor of the matrix, and ε is the strain in various phases and is thus non-uniform in volume. $\mathscr{G}^{1}(\varepsilon, E)$ is then

$$2V\mathscr{G}^{1}(\boldsymbol{\varepsilon},\boldsymbol{E}) = \boldsymbol{\varepsilon}^{0}:\boldsymbol{C}: \int_{V} \boldsymbol{\varepsilon} \,\mathrm{d}V + \boldsymbol{\varepsilon}^{0}: \int_{V} (\boldsymbol{\sigma} - \boldsymbol{C}:\boldsymbol{\varepsilon}) \mathrm{d}V$$
(39)

The first integral can be rearranged to

$$\int_{V} \varepsilon_{kl} dV = \frac{1}{2} \int_{V} (u_{k,l} + u_{l,k}) dV = \frac{1}{2} \oint_{S} (u_{k}n_{l} + u_{l}n_{k}) dS$$
$$= \frac{1}{2} \oint_{S} (\varepsilon_{km}^{0} x_{m}n_{l} + \varepsilon_{ln}^{0} x_{n}n_{k}) dS = \frac{1}{2} \left(\varepsilon_{km}^{0} \oint_{S} x_{m}n_{l} dS + \varepsilon_{ln}^{0} \oint_{S} x_{n}n_{k} dS \right)$$
$$= \frac{1}{2} \left(\varepsilon_{km}^{0} \int_{V} x_{m,l} dV + \varepsilon_{ln}^{0} \int_{V} x_{n,k} dV \right) = \frac{1}{2} \left(\varepsilon_{km}^{0} \delta_{ml} + \varepsilon_{ln}^{0} \delta_{nk} \right) \int_{V} dV = \varepsilon_{kl}^{0} V$$
(40)

The integrated in the second integral is zero in the matrix because of the matrix constitutive eqn (3). Thus, the second integral becomes

$$\sum_{n=1}^{N} \int_{V_n} (\boldsymbol{\sigma}^{(n)} - \boldsymbol{C} : \boldsymbol{\varepsilon}^{(n)}) dV = \sum_{n=1}^{N} \int_{V_n} [\boldsymbol{C}^{(n)} : \boldsymbol{\varepsilon}^{(n)} - (\boldsymbol{e}^{(n)})^T \cdot \boldsymbol{E}^{(n)} - \boldsymbol{C} : \boldsymbol{\varepsilon}^{(n)}] dV$$

$$= \sum_{n=1}^{N} (\boldsymbol{C}^{(n)} - \boldsymbol{C}) : \int_{V_n} \boldsymbol{\varepsilon}^{(n)} dV - \sum_{n=1}^{N} (\boldsymbol{e}^{(n)})^T \cdot \int_{V_n} \boldsymbol{E}^{(n)} dV$$

$$= \sum_{n=1}^{N} f_n [\mathcal{C}^{(n)} : \boldsymbol{\varepsilon}^{(n)} - \boldsymbol{e}^{(n)})^T \cdot \boldsymbol{\bar{E}}^{(n)}] V$$
(41)

where $V_n = f_n V$ is the total volume of the *n*-phase and $\mathscr{C}^{(n)} = \mathbf{C}^{(n)} - \mathbf{C}$. $\mathbf{\bar{\epsilon}}^{(n)} = \int_{V_n} \mathbf{\bar{\epsilon}}^{(n)} dV/V_n$ and $\mathbf{\bar{E}}^{(n)} = \int_{V_n} \mathbf{E}^{(n)} dV/V_n$ are the average strain and the average electric field in the *n*-th phase, respectively. The constitutive eqn (1) is used in the *n*-th phases (n = 1, ..., N). $\mathscr{G}^1(\mathbf{\epsilon}, \mathbf{E})$ now becomes

$$\mathscr{G}^{1}(\varepsilon, \boldsymbol{E}) = \frac{1}{2} \left\{ \boldsymbol{\varepsilon}^{0} : \boldsymbol{C} : \boldsymbol{\varepsilon}^{0} + \boldsymbol{\varepsilon}^{0} : \sum_{n=1}^{N} f_{n} [\mathscr{C}^{(n)} : \boldsymbol{\overline{\varepsilon}}^{(n)} - (\boldsymbol{e}^{(n)})^{T} \cdot \boldsymbol{\overline{E}}^{(n)} \right\}$$
(42)

For $\mathscr{G}^2(\varepsilon, E)$, we have

$$2V\mathscr{G}^{2}(\boldsymbol{\epsilon}, \boldsymbol{E}) = \int_{V} D_{i}E_{i} \, \mathrm{d}V = -\int_{V} D_{i}\phi_{,i} \, \mathrm{d}V = -\int_{V} (D_{i}\phi)_{,i} \, \mathrm{d}V = -\oint_{S} D_{i}\phi n_{i} \, \mathrm{d}S$$
$$= \oint_{S} D_{i}(-E_{m}^{0}x_{m})n_{i} \, \mathrm{d}S = E_{m}^{0}\oint_{S} D_{i}x_{m}n_{i} \, \mathrm{d}S = E_{m}^{0}\int_{V} (D_{i}x_{m})_{,i} \, \mathrm{d}V$$
$$= E_{m}^{0}\int_{V} D_{i}x_{m,i} \, \mathrm{d}V = E_{m}^{0}\delta_{mi}\int_{V} D_{i} \, \mathrm{d}V = E_{i}^{0}\int_{V} D_{i} \, \mathrm{d}V$$
(43)

In the derivation of eqn (43), the relation $E_i = -\phi_{,i}$, equilibrium of electric displacement, $D_{i,i} = 0$, boundary condition (33) and divergence theorem are used.

The **D** can be decomposed as

$$\boldsymbol{D} = \boldsymbol{k} \cdot \boldsymbol{E} + (\boldsymbol{D} - \boldsymbol{k} \cdot \boldsymbol{E}) \tag{44}$$

where k is the dielectric permittivity of the matrix and E is the electric field in various phases and is thus non-uniform in volume. $\mathscr{G}^{2}(\varepsilon, E)$ is then

$$2V\mathscr{G}^{2}(\boldsymbol{\varepsilon},\boldsymbol{E}) = \boldsymbol{E}^{0} \cdot \boldsymbol{k} \cdot \int_{V} \boldsymbol{E} \, \mathrm{d}V + \boldsymbol{E}^{0} \cdot \int_{V} (\boldsymbol{D} - \boldsymbol{k} \cdot \boldsymbol{E}) \mathrm{d}V$$
(45)

The first integral can be rearranged to

$$\int_{V} E_{m} \, \mathrm{d}V = -\int_{V} \phi_{,m} \, \mathrm{d}V = -\oint_{S} \phi_{n_{m}} \, \mathrm{d}S = -\oint_{S} (-E_{k}^{0} x_{k}) n_{m} \, \mathrm{d}S = E_{k}^{0} \oint_{S} x_{k} n_{m} \, \mathrm{d}S$$
$$= E_{k}^{0} \int_{V} x_{k,m} \, \mathrm{d}V = E_{k}^{0} \delta_{km} \int_{V} \mathrm{d}V = E_{m}^{0} V$$
(46)

The integration in the second integral is zero in the matrix because of the matrix constitutive eqn (4). Thus, the second integral becomes

$$\sum_{n=1}^{N} \int_{V_n} (\boldsymbol{D}^{(n)} - \boldsymbol{k} \cdot \boldsymbol{E}^{(n)}) \, \mathrm{d}V = \sum_{n=1}^{N} \int_{V_n} (\boldsymbol{e}^{(n)} : \boldsymbol{\varepsilon}^{(n)} + \boldsymbol{k}^{(n)} \cdot \boldsymbol{E}^{(n)} - \boldsymbol{k} \cdot \boldsymbol{E}^{(n)}) \, \mathrm{d}V$$

$$= \sum_{n=1}^{N} \boldsymbol{e}^{(n)} : \int_{V_n} \boldsymbol{\varepsilon}^{(n)} \, \mathrm{d}V + \sum_{n=1}^{N} (\boldsymbol{k}^{(n)} - \boldsymbol{k}) \cdot \int_{V_n} \boldsymbol{E}^{(n)} \, \mathrm{d}V$$

$$= \sum_{n=1}^{N} f_n [\boldsymbol{e}^{(n)} : \boldsymbol{\overline{\varepsilon}}^{(n)} + \mathcal{K}^{(n)} \cdot \boldsymbol{\overline{E}}^{(n)}] V$$
(47)

where $\mathscr{K}^{(n)} = \mathbf{k}^{(n)} - \mathbf{k}$ and the constitutive relation (2) are used in the *n*-th phase (n = 1, ..., N). $\mathscr{G}^2(\varepsilon, \mathbf{E})$ becomes

$$\mathscr{G}^{2}(\boldsymbol{\varepsilon},\boldsymbol{E}) = \frac{1}{2} \left\{ \boldsymbol{E}^{0} \cdot \boldsymbol{k} \cdot \boldsymbol{E}^{0} + \boldsymbol{E}^{0} \cdot \sum_{n=1}^{N} f_{n} [\mathscr{K}^{(n)} \cdot \boldsymbol{\bar{E}}^{(n)} + \boldsymbol{e}^{(n)} : \boldsymbol{\bar{\varepsilon}}^{(n)}] \right\}$$
(48)

Now $\mathscr{G}(\varepsilon, E)$ can be obtained by $\mathscr{G}^1(\varepsilon, E)$ and $\mathscr{G}^2(\varepsilon, E)$ as follows

$$\mathscr{G}(\boldsymbol{\varepsilon}, \boldsymbol{E}) = \frac{1}{2} \left\{ \boldsymbol{\varepsilon}^{0} : \boldsymbol{C} : \boldsymbol{\varepsilon}^{0} + \boldsymbol{\varepsilon}^{0} : \sum_{n=1}^{N} f_{n} [\mathscr{C}^{(n)} : \boldsymbol{\varepsilon}^{(n)} - (\boldsymbol{e}^{(n)})^{T} \cdot \boldsymbol{\bar{E}}^{(n)}] - \boldsymbol{E}^{0} \cdot \boldsymbol{k} \cdot \boldsymbol{E}^{0} - \boldsymbol{E}^{0} \cdot \sum_{n=1}^{N} f_{n} [\mathscr{K}^{(n)} \cdot \boldsymbol{\bar{E}}^{(n)} + \boldsymbol{e}^{(n)} : \boldsymbol{\varepsilon}^{(n)}] \right\}$$
(49)

According to the constitutive equations (31) and (32), the electric Gibbs' energy of the piezocomposite can be given exactly as

$$\mathscr{G}(\boldsymbol{\varepsilon}, \boldsymbol{E}) = \frac{1}{2} \left\{ \boldsymbol{\varepsilon}^{0} : \boldsymbol{\bar{C}} : \boldsymbol{\varepsilon}^{0} - 2\boldsymbol{E}^{0} \cdot \boldsymbol{\bar{e}} : \boldsymbol{\varepsilon}^{0} - \boldsymbol{E}^{0} \cdot \boldsymbol{\bar{k}} \cdot \boldsymbol{E}^{0} \right\}$$
(50)

A comparison between eqns (49) and (50) leads to

$$\boldsymbol{\varepsilon}^{0}: \boldsymbol{\bar{C}}: \boldsymbol{\varepsilon}^{0} - 2\boldsymbol{E}^{0} \cdot \boldsymbol{\bar{e}}: \boldsymbol{\varepsilon}^{0} - \boldsymbol{E}^{0} \cdot \boldsymbol{\bar{k}} \cdot \boldsymbol{E}^{0} = \boldsymbol{\varepsilon}^{0}: \boldsymbol{C}: \boldsymbol{\varepsilon}^{0} - \boldsymbol{E}^{0} \cdot \boldsymbol{k} \cdot \boldsymbol{E}^{0} + \boldsymbol{\varepsilon}^{0}: \sum_{n=1}^{N} f_{n}[\boldsymbol{\mathscr{C}}^{(n)}: \boldsymbol{\bar{\varepsilon}}^{(n)} - (\boldsymbol{e}^{(n)})^{T} \cdot \boldsymbol{\bar{E}}^{(n)}] - \boldsymbol{E}^{0} \cdot \sum_{n=1}^{N} f_{n}[\boldsymbol{\mathscr{K}}^{(n)} \cdot \boldsymbol{\bar{E}}^{(n)} + \boldsymbol{e}^{(n)}: \boldsymbol{\bar{\varepsilon}}^{(n)}]$$
(51)

It should be emphasized that eqn (51) is exact presentations of energy equivalence. The approximation nature of various approaches, such as the dilute solution and the Mori–Tanaka's method, comes solely from the evaluation of the average strains, $\bar{\boldsymbol{\varepsilon}}^{(n)}$, and the average electric field $\bar{\boldsymbol{E}}^{(n)}$, for each individual phase. Once a method for the evaluation of $\bar{\boldsymbol{E}}^{(n)}$ and $\bar{\boldsymbol{E}}^{(n)}$ are chosen, there exists a linear relationship among $\bar{\boldsymbol{\varepsilon}}^{(n)}$, $\bar{\boldsymbol{\varepsilon}}^{(n)}$, $\boldsymbol{\varepsilon}^{0}$ and \boldsymbol{E}^{0} , i.e.

$$\bar{\boldsymbol{\varepsilon}}^{(n)} = \mathscr{M}^{(n)} : \boldsymbol{\varepsilon}^0 + \mathscr{N}^{(n)} \cdot \boldsymbol{E}^0, \quad \bar{\boldsymbol{E}}^{(n)} = \mathscr{P}^{(n)} : \boldsymbol{\varepsilon}^0 + \mathscr{Q}^{(n)} \cdot \boldsymbol{E}^0$$
(52)

where the tensors $\mathcal{M}^{(n)}$, $\mathcal{N}^{(n)}$, $\mathcal{P}^{(n)}$ and $\mathcal{Q}^{(n)}$ depend not only on the *n*-th phase and the matrix, but also on the method used. Substitution of eqn (52) into (51) leads to

$$\boldsymbol{\varepsilon}^{0}: \boldsymbol{\bar{C}}: \boldsymbol{\varepsilon}^{0} - 2\boldsymbol{E}^{0} \cdot \boldsymbol{\bar{e}}: \boldsymbol{\varepsilon}^{0} - \boldsymbol{E}^{0} \cdot \boldsymbol{k} \cdot \boldsymbol{E}^{0} = \boldsymbol{\varepsilon}^{0}: \left\{ \boldsymbol{C} + \sum_{n=1}^{N} f_{n} [\mathcal{C}^{(n)}: \mathcal{M}^{(n)} - (\boldsymbol{e}^{(n)})^{T} \cdot \boldsymbol{\mathcal{P}}^{(n)}] \right\}: \boldsymbol{\varepsilon}^{0} \\ - \boldsymbol{E}^{0} \cdot \left\{ \sum_{n=1}^{N} f_{n} [\mathcal{H}^{(n)} \cdot \boldsymbol{\mathcal{P}}^{(n)} - (\mathcal{C}^{(n)}: \mathcal{N}^{(n)})^{T} + (\mathcal{Q}^{(n)})^{T} \cdot \boldsymbol{e}^{(n)} + \boldsymbol{e}^{(n)}: \mathcal{M}^{(n)}] \right\}: \boldsymbol{\varepsilon}^{0} \\ - \boldsymbol{E}^{0} \cdot \left\{ \boldsymbol{k} + \sum_{n=1}^{N} f_{n} [\boldsymbol{e}^{(n)}: \mathcal{N}^{(n)} + \mathcal{H}^{(n)} \cdot \boldsymbol{\mathcal{Q}}^{(n)}] \right\} \cdot \boldsymbol{E}^{0}$$
(53)

Because ε^0 and E^0 are arbitrary, the \overline{C} , \overline{e} and \overline{k} can be obtained as

$$\bar{\boldsymbol{C}} = \boldsymbol{C} + \sum_{n=1}^{N} f_n \operatorname{sym} \left[\mathscr{C}^{(n)} : \mathscr{M}^{(n)} - (\boldsymbol{e}^{(n)})^T \cdot \mathscr{P}^{(n)} \right]$$
(54)

$$\bar{\boldsymbol{k}} = \boldsymbol{k} + \sum_{n=1}^{N} f_n \operatorname{sym} \left[\boldsymbol{e}^{(n)} : \mathcal{N}^{(n)} + \mathcal{K}^{(n)} \cdot \mathcal{Q}^{(n)} \right]$$
(55)

$$\bar{\boldsymbol{e}} = \frac{1}{2} \sum_{n=1}^{N} f_n [\mathscr{K}^{(n)} \cdot \mathscr{P}^{(n)} - (\mathscr{C}^{(n)} : \mathscr{N}^{(n)})^T + (\mathscr{Q}^{(n)})^T \cdot \boldsymbol{e}^{(n)} + \boldsymbol{e}^{(n)} : \mathscr{M}^{(n)}]$$
(56)

where 'sym A' represents the symmetry part of A, i.e. sym $A = (A_{ijkl} + A_{klij})/2$.

Now, the analytical solutions of the effective electroelastic moduli of the piezocomposites are obtained. It is known from the above equations that the solutions can be expressed by the material constants of the inclusions $C^{(n)}$, $e^{(n)}$, $k^{(n)}$, the material constants of the matrix C, k, the Eshelby's tensors of the perfectly elastic inclusion problem $S^{(n)}$, the Eshelby's tensors of the perfectly dielectric inclusion problem, $s^{(n)}$, and the volume concentrations of the individual phase f_n (n = 1, ..., N). Since $S^{(n)}$ and $s^{(n)}$ were obtained by means of the perfectly inclusion problem and the perfectly dielectric inclusion problem, respectively, the analytical solutions of the effective electroelastic moduli of the piezocomposites can be gained conveniently.

4. The dilute solution and the Mori-Tanaka's method for piezocomposites

4.1. The dilute solution or the non-interacting solution

The average strain and electric field in the dilute solution or the non-interacting solution is approximated by the constraint strain and the constraint electric field that would occur in an isolated piezoelectric inclusion embedded in an infinite non-piezoelectric matrix, whose closed-form solutions can be expressed by equations (28)–(30). When the interaction among the inclusions is not considered, $\mathcal{M}^{(n)}$, $\mathcal{P}^{(n)}$ and $\mathcal{Q}^{(n)}$ are given by

$$\mathscr{M}^{(n)} = \mathscr{H}^{1(n)}, \quad \mathscr{N}^{(n)} = \mathscr{H}^{2(n)}, \quad \mathscr{P}^{(n)} = \mathscr{H}^{3(n)}, \quad \mathscr{Q}^{(n)} = \mathscr{H}^{4(n)}$$
(57)

where $\mathscr{H}^{1(n)}$, $\mathscr{H}^{2(n)}$, $\mathscr{H}^{3(n)}$ and $\mathscr{H}^{4(n)}$ are electroelastic Eshelby's tensors of the *n*-th inclusion in an infinite non-piezoelectric matrix and can be determined by eqn (29). Substitution of eqn (57)

into (54)–(56) gives the effective electroelastic moduli of the multiphase piezocomposites with piezoelectric ellipsoidal inclusions by means of the dilute solution.

4.2. The Mori–Tanaka's method

The Mori–Tanaka's method is shown in this section to correspond to one special way of evaluating the average strain and electric fields of inclusions and thus to have a clear physical description, and to fall into the Budiansky's (1965) energy-equivalence framework, or eqn (51). Details of the Mori–Tanaka's method can be found in Weng (1984) and Benveniste (1987), while a quick derivation for the Mori–Tanaka's method based on the physical description is provided in this section.

The aim of the Mori–Tanaka's method is to account for interactions among inclusions better than the dilute solution. In this method, the inclusions are embedded in an infinite matrix and the matrix is subjected to the average matrix strain, $\bar{\epsilon}^m$, and the average matrix electric field, \bar{E}^m , in the composite system rather than the applied strain, ϵ^0 , and electric field E^0 . The average matrix strain $\bar{\epsilon}^m$ and electric field \bar{E}^m reasonably characterize the interaction among inclusions because each inclusion in the composite material is surrounded by its adjacent matrix and is matched with neighboring inclusions and corresponding matrices through the average matrix strain $\bar{\epsilon}^m$ and electric field \bar{E}^m . Hence, the *n*-th inclusion strain and electric field are obtained by replacing the applied strain ϵ^0 and electric field E^0 , in the dilute solution, eqns (28), with the average matrix strain, $\bar{\epsilon}^m$, and the average electric field \bar{E}^m , i.e.

$$\boldsymbol{\varepsilon}^{(n)} = \mathscr{H}^{1(n)} : \boldsymbol{\bar{\varepsilon}}^m + \mathscr{H}^{2(n)} \cdot \boldsymbol{\bar{E}}^m, \quad \boldsymbol{E}^{(n)} = \mathscr{H}^{3(n)} : \boldsymbol{\bar{\varepsilon}}^m + \mathscr{H}^{4(n)} \cdot \boldsymbol{\bar{E}}^m$$
(58)

where

$$\bar{\boldsymbol{\varepsilon}}^{m} = \langle \boldsymbol{\varepsilon} \rangle_{m} = \frac{1}{V_{m}} \int_{V_{m}} \boldsymbol{\varepsilon} \, \mathrm{d}V, \quad \bar{\boldsymbol{E}}^{m} = \langle \boldsymbol{E} \rangle_{m} = \frac{1}{V_{m}} \int_{V_{m}} \boldsymbol{E} \, \mathrm{d}V$$
(59)

where V_m is the total volume of the matrix. From the definition of the average strain and electric field, one has

$$\boldsymbol{\varepsilon}^{0} = \langle \boldsymbol{\varepsilon} \rangle_{c} = \frac{1}{V} \int_{V} \boldsymbol{\varepsilon} \, \mathrm{d}V = \frac{1}{V} \int_{V_{m}} \boldsymbol{\varepsilon} \, \mathrm{d}V + \frac{1}{V} \sum_{n=1}^{N} \int_{V_{n}} \boldsymbol{\varepsilon} \, \mathrm{d}V = \left[1 - \sum_{n=1}^{N} f_{n} \right] \langle \boldsymbol{\varepsilon} \rangle_{m} + \sum_{n=1}^{N} f_{n} \langle \boldsymbol{\varepsilon} \rangle_{n}$$
(60)

$$\boldsymbol{E}^{0} = \langle \boldsymbol{E} \rangle_{c} = \frac{1}{V} \int_{V} \boldsymbol{E} \, \mathrm{d}V = \frac{1}{V} \int_{V_{m}} \boldsymbol{E} \, \mathrm{d}V + \frac{1}{V} \sum_{n=1}^{N} \int_{V_{n}} \boldsymbol{E} \, \mathrm{d}V = \left[1 - \sum_{n=1}^{N} f_{n}\right] \langle \boldsymbol{E} \rangle_{m} + \sum_{n=1}^{N} f_{n} \langle \boldsymbol{E} \rangle_{n}$$
(61)

In the derivation of the above equations, the relations (40), (46), (59) and (78) are used. It is demonstrated that if a linear piezoelectric ellipsoidal inclusion in an infinite linear non-piezoelectric matrix is subjected to the uniform remote strain, ε^0 , and electric field, E^0 , the constraint strain and electric field inside the inclusion remain uniform. So, one has

$$\langle \boldsymbol{\varepsilon} \rangle_n = \boldsymbol{\varepsilon}^{(n)}, \quad \langle \boldsymbol{E} \rangle_n = \boldsymbol{E}^{(n)}$$
 (62)

Substitution of equations (58), (59) and (62) into (60) and (61), gives

$$\boldsymbol{\varepsilon}^{0} = \boldsymbol{H}^{1} : \boldsymbol{\overline{\varepsilon}}^{m} + \boldsymbol{H}^{2} \cdot \boldsymbol{\overline{E}}^{m}, \quad \boldsymbol{E}^{0} = \boldsymbol{H}^{3} : \boldsymbol{\overline{\varepsilon}}^{m} + \boldsymbol{H}^{4} \cdot \boldsymbol{\overline{E}}^{m}$$
(63)

where

$$\boldsymbol{H}^{1} = \mathbf{I} - \sum_{n=1}^{N} f_{n} [\mathbf{I} - \mathscr{H}^{1(n)}], \quad \boldsymbol{H}^{2} = \sum_{n=1}^{N} f_{n} \mathscr{H}^{2(n)}$$
(64a)

$$\boldsymbol{H}^{3} = \sum_{n=1}^{N} f_{n} \mathscr{H}^{3(n)}, \quad \boldsymbol{H}^{4} = \mathbf{i} - \sum_{n=1}^{N} f_{n} [\mathbf{i} - \mathscr{H}^{4(n)}]$$
(64b)

From equations (63), one can obtain

$$\bar{\boldsymbol{\varepsilon}}^{m} = \boldsymbol{M} : \boldsymbol{\varepsilon}^{0} + \boldsymbol{N} \cdot \boldsymbol{E}^{0}, \quad \bar{\boldsymbol{E}}^{m} = \boldsymbol{P} : \boldsymbol{\varepsilon}^{0} + \boldsymbol{Q} \cdot \boldsymbol{E}^{0}$$
(65)

where

$$M = [H^{1} - H^{2} \cdot (H^{4})^{-1} \cdot H^{3}]^{-1}, \quad N = -M : H^{2} \cdot (H^{4})^{-1}$$
(66a)

$$P = -(H^4)^{-1} \cdot H^3 : M, \quad Q = (H^4)^{-1} \cdot [i - H^3 : N]$$
(66b)

Substitution of equations (65) into (58) gives the average strain $\bar{\mathbf{\epsilon}}^{(n)}$ and electric field $\bar{\mathbf{E}}^{(n)}$ as

$$\boldsymbol{\bar{\varepsilon}}^{(n)} = \boldsymbol{\varepsilon}^{(n)} = \mathcal{M}^{(n)} : \boldsymbol{\varepsilon}^0 + \mathcal{N}^{(n)} \cdot \boldsymbol{E}^0$$
(67)

$$\bar{\boldsymbol{E}}^{(n)} = \boldsymbol{E}^{(n)} = \mathscr{P}^{(n)} : \boldsymbol{\varepsilon}^0 + \mathscr{Q}^{(n)} \cdot \boldsymbol{E}^0$$
(68)

where

$$\mathscr{M}^{(n)} = \mathscr{H}^{1(n)} : \boldsymbol{M} + \mathscr{H}^{2(n)} \cdot \boldsymbol{P}, \quad \mathscr{N}^{(n)} = \mathscr{H}^{1(n)} : \boldsymbol{N} + \mathscr{H}^{2(n)} \cdot \boldsymbol{Q}$$
(69a)

$$\mathscr{P}^{(n)} = \mathscr{H}^{3(n)} : \boldsymbol{M} + \mathscr{H}^{4(n)} \cdot \boldsymbol{P}, \quad \mathscr{Q}^{(n)} = \mathscr{H}^{3(n)} : \boldsymbol{N} + \mathscr{H}^{4(n)} \cdot \boldsymbol{Q}$$
(69b)

Substitution of equations (29), (30) and (69) into (54)–(56) gives the effective electroelastic moduli of the multiphase piezocomposites with piezoelectric ellipsoidal inclusions by means of the Mori–Tanaka's method.

5. Comparison between predicted and experimental results

Chan and Unsworth (1989) gave the experimental results of 1–3 PZT-7A/Araldite D piezocomposite and Furukawa et al. (1978) obtained the experimental results of 0–3 PZT-5A/Epoxy piezocomposites. In Appendix A and Appendix B, the closed-form solutions of the effective electroelastic moduli of the 1–3 and 0–3 piezocomposites shown in Fig. 1 are given. The experimental results and the theoretical curves calculated by the closed-form solutions are plotted in Fig. 2 and Fig. 3, while 1–3 piezocomposite has only the cylindrical inclusion aligned in the positive direction of 3-axis. The electroelastic moduli of PZT-7A and PZT-5A used in subsequent computations are obtained from Chan and Unsworth (1989), Furukawa et al. (1978), Jaffe (1971) and Dunn and Taya (1993), and the parameters are given in Table 1 where the well-known two types of index notations have been adopted.

The parameters \tilde{M} , \tilde{k} and \tilde{d} in Fig. 2 (c)–(e) and Fig. 3 are elastic compliance moduli, dielectric permittivity and charge constants, respectively, which can be determined by the following equations (Lines and Glass, 1977)



Fig. 1. (a) The 1-3 piezocomposite with two kinds of transversely isotropic piezoelectric cylindrical inclusions. (b) The 0-3 piezocomposite with transversely isotropic piezoelectric spherical inclusion.



Fig. 2. Experimental verification of the variation of relative electroelastic moduli of piezocomposites with unidirectional transversely isotropic cylindrical inclusion with volume fraction of ceramic and the experimental values are from Chan et al. (1989). — theoretical results, \bullet experimental results.

$$\tilde{\boldsymbol{M}} = (\boldsymbol{\bar{C}})^{-1}, \quad \tilde{\boldsymbol{d}} = \boldsymbol{\bar{e}} : \boldsymbol{\tilde{M}}, \quad \boldsymbol{\tilde{k}} = \boldsymbol{\bar{k}} + \boldsymbol{\tilde{d}} : (\boldsymbol{\bar{e}})^T$$
(70)

where \bar{C} , \bar{e} and \bar{k} are obtained from equations (54)–(56).

The parameters k_t and k_p in Fig. 2 (a)–(b) are the thickness coupling constant and the planar coupling constant, respectively, which can be calculated as follows (Jaffe, 1971)

$$k_p = \sqrt{\frac{2(\tilde{d}_{311})^2}{\tilde{k}_{33}(\tilde{M}_{1111} + \tilde{M}_{1122})}}, \quad k_t = \frac{B - Ak_p}{\sqrt{1 - A^2}\sqrt{1 - k_p^2}}$$
(71)

where



Fig. 3. Experimental verification of the variation of relative electroelastic moduli of piezocomposites with transversely isotropic spherical inclusion with volume fraction of ceramic and the experimental values come from Furukawa et al. (1976). — theoretical results, \bullet experimental results.

Table 1 Electroelastic material properties

	<i>C</i> ₁₁ (GPa)	С ₁₂ (GPa)	С ₁₃ (GPa)	<i>C</i> ₃₃ (GPa)	С ₄₄ (GPa)	e_{31} (C/m ²)	<i>e</i> ₃₃ (C/m ²)	e_{15} (C/m ²)	k_{11}/k_0*	k ₃₃ /k ₀ *
PZT-7A	148	76.2	74.2	131	25.4	-2.1	9.5	9.2	460	235
PZT-5A	121	75.4	75.2	111	21.1	-5.4	15.8	12.3	916	830
Araldite D	8.0	4.4	4.4	8.0	1.8	0	0	0	4.0	4.0
Epoxy	8.0	4.4	4.4	8.0	1.8	0	0	0	4.2	4.2

* $k_0 = 8.85 \times 10^{-12} \text{ C/Vm}^2$.

$$A = \sqrt{\frac{2(\tilde{M}_{1133})^2}{\tilde{M}_{3333}(\tilde{M}_{1111} - \tilde{M}_{1122})}}, \quad B = \sqrt{\frac{(\tilde{d}_{333})^2}{\tilde{k}_{33}\tilde{M}_{3333}}}$$
(72)

Figures 2 and 3 show that the theoretical curves calculated by means of the Mori-Tanaka's method agree quite well with the experimental values, but the theoretical curves obtained by the dilute solution agree well with the experimental values only when the volume fraction of the ceramic inclusion is smaller than 0.3.

6. The analysis of the 1–3 piezocomposites with two kinds of transversely isotropic piezoelectric cylindrical inclusions

In this section, it is analyzed in detail for the 1–3 piezocomposites with two kinds of transversely isotropic piezoelectric cylindrical inclusions shown in Fig. 1(a), in which one phase is in the positive



Fig. 4. Variations of effective electroelastic moduli of 1–3 piezocomposite of two kinds of transversely isotropic cylindrical inclusions in (σ , E) space with f_0 and C_f .

direction of 3-axis with a volume fraction f^+ and the other phase is in the negative direction of the 3-axis with a volume fraction f^- . The variations of the effective electroelastic properties of this kind of the piezocomposite calculated by the Mori–Tanaka's method with f_0 and C_f are shown in Figs 4–7. f_0 is the total volume fraction, i.e. $f_0 = f^+ + f^-$, and $C_f = f^-/f_0$. Thus, f^+ and f^- can be expressed by f_0 and C_f as follows

$$f^{+} = (1 - C_f)f_0, \quad f^{-} = C_f f_0 \tag{73}$$

It can be shown from the closed-form solutions of the effective electroelastic moduli of this kind of piezocomposite and from Figs 4–7 that



- (1) Except \bar{C}_{1313} and \tilde{M}_{1313} , the other components of elastic moduli \bar{C} and elastic compliance moduli \tilde{M} do not vary with C_f . In other words \bar{C}_{1313} and \tilde{M}_{1313} depend not only on elastic properties but also on piezoelectric and dielectric properties of the inclusions and matrix. The other components of \bar{C} and \tilde{M} depend only on elastic properties of the inclusions and the matrix.
- (2) \tilde{e} and \tilde{d} are zero when $C_f = 0.5$ i.e. $f^+ = f^-$. In this case, the composite does not have any electric-mechanical coupling.
- (3) \bar{k}_{11} and \bar{k}_{33} reach their greatest values at $f_0 = 1$ and $C_f = 0.5$. \tilde{k}_{11} and \tilde{k}_{33} approach to their greatest values when $f_0 = 1$ and $C_f = 0$ or $C_f = 1$.
- (4) k_p reaches its greatest value when $f_0 1$ and $C_f = 0$ or $C_f = 1$ and its smallest one when $f_0 = 1$ and $C_f = 0.5$. k_t approaches to its smallest value while $f_0 = 1$ and $C_f = 0.5$ and its greatest one while f_0 is about 0.8 and $C_f = 0$ or $C_f = 1$.



Fig. 5. Variations of effective electroelastic moduli of 1–3 piezocomposite of two kinds of transversely isotropic cylindrical inclusions in (ε , E) space with f_0 and C_f .

7. Conclusions

In this paper, the closed-form solutions of the electroelastic Eshelby's tensors of a piezoelectric ellipsoidal inclusion in an infinite non-piezoelectric matrix are obtained via the Green's function technique. Based on the generalized Budiansky's energy-equivalence framework and the closed-form solutions of the electroelastic Eshelby's tensors, a unified model for multiphase piezocomposites of non-piezoelectric matrix and piezoelectric inclusions has been set up. The unified model can describe the multiphase piezocomposites with different connectivities. The closed-form solutions of the effective electroelastic moduli of piezocomposites are also obtained. The relations can be expressed by the material constants of the inclusions $C^{(n)}$, $e^{(n)}$, $k^{(n)}$, the material constants of the perfectly elastic inclusion problem $S^{(n)}$, the Eshelby's tensors of the perfectly dielectric inclusion problem of the volume concentrations of the electroelastic inclusion problem, $s^{(n)}$, and the volume concentrations of the



individual phase f_n (n = 1, ..., N). Since $S^{(n)}$ and $s^{(n)}$ can be obtained by means of the perfectly elastic inclusion problem and the perfectly dielectric inclusion problem, respectively, the closed-form solutions of the effective electroelastic moduli of the piezocomposites can be gained conveniently. The results in this paper can be used to analyze and design the multiphase piezo-composites.

Appendix A: The closed-form solutions of the effective moduli of the 1–3 piezocomposite with two kinds of transversely isotropic piezoelectric cylindrical inclusions and non-piezoelectric matrix

As an important example, the 1-3 piezocomposite with two kinds of piezoelectric cylindrical inclusions shown in Fig. 1(a) is considered in detail. In which one phase is aligned in the positive direction of 3-axis and another phase is in the negative direction of 3-axis. The closed-form



Fig. 6. Variations of \tilde{d}_h and \bar{e}_h with f_0 and C_f . Where $\tilde{d}_h = \tilde{d}_{333} + 2\tilde{d}_{311}$ and $\bar{e}_h = \bar{e}_{333} + 2\bar{e}_{311}$.

relations of the effective electroelastic moduli of this kind of piezocomposite are given. It is assumed that the piezoelectric inclusions are transversely isotropic and the matrix is isotropic.

For the inclusion, its non-zero material constants are

$$C_{1111}^{*} = C_{2222}^{*}, \quad C_{1133}^{*} = C_{2233}^{*}, \quad C_{3333}^{*}, \quad C_{1313}^{*} = C_{2323}^{*}, \quad C_{1212}^{*} = \frac{1}{2}(C_{1111}^{*} - C_{1122}^{*})$$

$$e_{311}^{*} = e_{322}^{*}, \quad e_{333}^{*}, \quad e_{113}^{*} = e_{223}^{*}, \quad k_{11}^{*} = k_{22}^{*}, \quad k_{33}^{*}$$
(A1)

where 3-axis is the symmetric axis.

The material constants of the matrix are

$$C_{ijkl}^{-1} = \frac{1}{C_{1111} - C_{1122}} \quad I_{ijkl} - \frac{C_{1122}}{(C_{1111} - C_{1122})(C_{1111} + 2C_{1122})} \delta_{ij}\delta_{kl} \quad k_{ij} = k\delta_{ij}$$
(A2)

In order to simplify the equations, several parameters are introduced by



Fig. 7. Variations of planar coupling constant k_p and thickness coupling constant k_t with f_0 and C_f .

$$b_1 = \frac{C_{1111}^0}{C_{1111}}, \quad b_2 = \frac{C_{1122}^0}{C_{1111}}, \quad b_3 = \frac{C_{1133}^0}{C_{1111}}, \quad b_4 = \frac{C_{1313}^0}{C_{1111}}, \quad b_5 = \frac{C_{1212}^0}{C_{1111}}, \quad b_6 = \frac{C_{3333}^0}{C_{1111}}$$
(A3)

$$a_1 = \frac{C_{1122}}{C_{1111}}, \quad c_1 = \frac{k_{11}^0}{k_{11}}, \quad c_2 = \frac{k_{33}^0}{k_{11}}, \quad h_1 = \frac{(e_{113}^*)^2}{k_{11}C_{1111}}, \quad h_2 = \frac{(e_{311}^*)^2}{k_{11}C_{1111}}$$
(A4)

where

$$C^0 = C^* - C, \quad k^0 = k^* - k$$
 (A5)

Appendix AI: The dilute solution or non-interaction solution

The non-zero components of $m{ar{C}}$ are

$$\bar{C}_{1111} + \bar{C}_{1122} = C_{1111} \left\{ 1 + a_1 + \frac{2f_0(b_1 + b_2)}{2 + b_1 + b_2} \right\}$$
(AI1a)

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$$\bar{C}_{1111} - \bar{C}_{1122} = C_{1111} \left\{ 1 - a_1 + \frac{4f_0(1 - a_1)(b_1 - b_2)}{4(1 - a_1) + (3 - a_1)(b_1 - b_2)} \right\}$$
(AI1b)

$$\bar{C}_{3333} = C_{1111} \left\{ 1 + f_0 \left[b_6 - \frac{2b_3^2}{2 + b_1 + b_2} \right] \right\}$$
(AI1c)

$$\bar{C}_{1133} = \bar{C}_{2233} = C_{1111} \left\{ a_1 + \frac{f_0 b_3}{2 + b_1 + b_2} \right\}$$
 (AI1d)

$$\bar{C}_{1313} = \bar{C}_{2323} = C_{1111}(1-a_1) \left\{ \frac{1}{2} + \frac{f_0[h_1 + (2+c_1)b_4]}{h_1 + (2+c_1)(1-a_1+b_4)} \right\}$$
(AI1e)

$$\bar{C}_{1212} = C_{1111}(1-a_1) \left\{ \frac{1}{2} + \frac{2f_0 b_5}{2(1-a_1) + (3-a_1) b_5} \right\}$$
$$\bar{C}_{ijkl} = \bar{C}_{ijlk} = \bar{C}_{klij}$$
(AI1f)

The non-zero components of \bar{k} are

$$\bar{k}_{11} = \bar{k}_{22} = k_{11} \left\{ 1 + \frac{2f_0[h_1 + (1 - a_1 + b_4)c_1]}{h_1 + (2 + c_1)(1 - a_1 + b_4)} \right\}$$
(AI2a)

$$\bar{k}_{33} = k_{11} \left\{ 1 + f_0 \left[c_2 + \frac{2h_2}{2 + b_1 + b_2} \right] \right\}$$
(AI2b)

The non-zero components of \bar{e} are

$$\bar{e}_{113} = \bar{e}_{233} = \frac{2(f^+ - f^-)(1 - a_1)e_{113}^*}{h_1 + (2 + c_1)(1 - a_1 + b_4)}$$
(AI3a)

$$\bar{e}_{311} = \bar{e}_{322} = \frac{2(f^+ - f^-)e_{311}^*}{2 + b_1 + b_2}$$
(AI3b)

$$\bar{e}_{333} = (f^+ - f^-) \left(e^*_{333} - \frac{2b_3 e^*_{311}}{2 + b_1 + b_2} \right)$$

$$\bar{e}_{pij} = \bar{e}_{pji}$$
(AI3c)

where f_0 is the total volume fraction of the piezoelectric inclusions; f^+ and f^- are the volume concentrations of the piezoelectric inclusion in the positive direction of 3-axis and in the negative direction of 3-axis, respectively. The relation among them is $f^+ + f^- = f_0$.

Appendix AII: The Mori–Tanaka's method

The non-zero components of \bar{C} are

$$\bar{C}_{1111} + \bar{C}_{1122} = C_{1111} \left\{ 1 + a_1 + \frac{2f_0(b_1 + b_2)}{2 + (1 - f_0)(b_1 + b_2)} \right\}$$
(AII1a)

$$\bar{C}_{1111} - \bar{C}_{1122} = C_{1111} \left\{ 1 - a_1 + \frac{4f_0(1 - a_1)(b_1 - b_2)}{4(1 - a_1) + (1 - f_0)(3 - a_1)(b_1 - b_2)} \right\}$$
(AII1b)

$$\bar{C}_{3333} = C_{1111} \left\{ 1 + f_0 \left[b_6 - \frac{2(1 - f_0)b_3^2}{2 + (1 - f_0)(b_1 + b_2)} \right] \right\}$$
(AII1c)

$$\bar{C}_{1133} = \bar{C}_{2233} = C_{1111} \left\{ a_1 + \frac{2f_0 b_3}{2 + (1 - f_0)(b_1 + b_2)} \right\}$$
(AII1d)

$$\bar{C}_{1212} = C_{1111}(1-a_1) \left\{ \frac{1}{2} + \frac{2f_0b_5}{2(1-a_1) + (1-f_0)(3-a_1)b_5} \right\}$$
(AII1e)

$$\bar{C}_{1313} = C_{1111}(1-a_1) \left\{ \frac{1}{2} + \frac{f_0[2b_4 + (1-f_0)(h_1 + 2b_4 + c_1b_4)] + 8f^+ f^- \lambda_1}{\hat{C}(f_0) - 8f^+ f^- \lambda_1} \right\}$$
(AII1f)

$$\bar{C}_{2222} = \bar{C}_{1111}; \quad \bar{C}_{2323} = \bar{C}_{1313}$$
 (AII1g)

$$\bar{C}_{ijkl} = \bar{C}_{ijlk} = \bar{C}_{klij}$$
(AII1h)

where

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$$\hat{C}(f_0) = 2(1-a_1) + (1-f_0)(c_1 + 2b_4 - c_1a_1) + (1-f_0)^2(c_1b_4 + h_1)$$
(AII1i)

$$\lambda_1 = \frac{(1-a_1)h_1}{h_1 + (2+c_1)(1-a_1+b_4)}$$
(AII1j)

The non-zero components of \bar{k} are

$$\bar{k}_{11} = \bar{k}_{22} = k_{11} \left\{ 1 + \frac{2f_0[c_1(1-a_1) + (1-f_0)(c_1b_4 + h_1)] + 16f^+f^-\lambda_1}{\hat{C}(f_0) - 8f^+f^-\lambda_1} \right\}$$
(AII2a)

$$\bar{k}_{33} = k_{11} \left\{ 1 + f_0 \left[c_2 + \frac{2(1 - f_0)h_2 + 16f^+ f^- \lambda_2}{2 + (1 - f_0)(b_1 + b_2)} \right] \right\}$$
(AII2b)

where

$$\lambda_2 = \frac{h_2}{2+b_1+b_2} \tag{AII2c}$$

The non-components of \bar{e} are

$$\bar{e}_{113} = \bar{e}_{223} = \frac{2(f^+ - f^-)(1 - a_1)e_{113}^*}{\hat{C}(f_0) - 8f^+ f^- \lambda_1}$$
(AII3a)

$$\bar{e}_{311} = \bar{e}_{322} = \frac{2(f^+ - f^-)e_{311}^*}{2 + (1 - f_0)(b_1 + b_2)}$$
(AII3b)

$$\bar{e}_{333} = (f^+ - f^-) \left\{ e_{333}^* - \frac{2(1 - f_0)b_3 e_{311}^*}{2 + (1 - f_0)(b_1 + b_2)} \right\}$$

$$\bar{e}_{pij} = \bar{e}_{pji}$$
(AII3c)

Appendix B: The closed-form solutions of the effective moduli of the piezocomposite with transversely isotropic piezoelectric spherical inclusion

As another important example, the 0–3 piezocomposite with transversely isotropic piezoelectric spherical inclusion shown in Fig. 1(b) is considered in detail. The material constants are the same as those in Appendix A, which can be expressed by eqns (A1)–(A6). The closed-form relations of the effective electroelastic moduli of this kind of piezocomposite are given.

The non-zero components of \bar{C} are:

$$\bar{C}_{1111} + \bar{C}_{1122} = C_{1111} \left\{ 1 + a_1 + f \frac{1}{\Omega_0} \left[15(1 - a_1)(b_1 + b_2) + (7 - 3a_1)b_0 + \frac{2k_{11}\varphi_2^2}{C_{1111}\Omega} \right] \right\}$$
(B1a)

$$\bar{C}_{1111} - \bar{C}_{1122} = (1 - a_1)C_{1111} \left\{ 1 + f \frac{15(b_1 - b_2)}{15(1 - a_1) + 2(4 - a_1)(b_1 - b_2)} \right\}$$
(B1b)

$$\bar{C}_{1133} = \bar{C}_{2233} = C_{1111} \left\{ a_1 + f \frac{1}{\Omega_0} \left[15(1-a_1)b_3 + (1+a_1)b_0 + \frac{k_{11}\varphi_2\varphi_3}{C_{1111}\Omega} \right] \right\}$$
(B1c)

$$\bar{C}_{3333} = C_{1111} \left\{ 1 + f \frac{1}{\Omega_0} \left[15(1-a_1)b_6 + (6-4a_1)b_0 + \frac{k_{11}\varphi_3^2}{C_{1111}\Omega} \right] \right\}$$
(B1d)

$$\bar{C}_{1313} = \bar{C}_{2323} = \frac{1}{2}(1-a_1)C_{1111}\left\{1 + f\frac{30}{\omega}[h_1 + (3+c_1)b_4]\right\}$$
(B1e)

$$\bar{C}_{1212} = \frac{1}{2}(1-a_1)C_{1111} \left\{ 1 + f \frac{30b_5}{15(1-a_1) + 2(4-a_1)b_5} \right\}$$

$$\bar{C}_{ijkl} = \bar{C}_{jikl} = \bar{C}_{klij}$$
(B1f)

The non-zero components of \mathbf{k} are:

$$\bar{k}_{11} = \bar{k}_{22} = k_{11} \left[1 + 3f \left(1 - \frac{3\omega_0}{\omega} \right) \right], \quad \bar{k}_{33} = k_{11} \left[1 + 3f \left(1 - \frac{3\Omega_0}{\Omega} \right) \right]$$
(B2)

The non-zero components of \bar{e} are:

$$\bar{e}_{113} = \bar{e}_{131} = \bar{e}_{223} = \bar{e}_{232} = f \frac{3k_{11}\varphi_1}{\omega}$$
 (B3a)

$$\bar{e}_{311} = \bar{e}_{322} = f \frac{3k_{11}\varphi_2}{\Omega}, \quad \bar{e}_{333} = f \frac{3k_{11}\varphi_3}{\Omega}$$
 (B3b)

where

$$\varphi_1 = 15(1-a_1) \times \frac{e_{113}^*}{k_{11}} \tag{B4a}$$

$$\varphi_2 = \frac{1}{k_{11}} [5(1-a_1)(e_0 - \bar{e}_0) - (7-3a_1)e_1 + (1+a_1)e_2]$$
(B4b)

$$\varphi_3 = \frac{1}{k_{11}} [5(1-a_1)(e_0 + 2\bar{e}_0) - 2(1+a_1)e_1 + (6-4a_1)e_2]$$
(B4c)

$$\omega_0 = 15(1-a_1) + 4(4-a_1)b_4 \tag{B5a}$$

$$\omega = 4(4 - a_1)h_1 + (3 + c_1)\omega_0 \tag{B5b}$$

$$\Omega_0 = 15(1-a_1) + (6-4a_1)(b_1+b_2) + (7-3a_1)b_6 - 4(1+a_1)b_3 + \frac{2}{3}(4-a_1)b_0$$
(B6a)

$$\Omega_1 = \frac{1}{9k_{11}C_{1111}} \left\{ 15(1-a_1)e_0^2 + 2(4-a_1)[6\bar{e}_0^2 + (e_2 - 2e_1)e_0 + 2(e_1 + e_2)\bar{e}_0 \right\}$$
(B6b)

$$\Omega = \Omega_1 + (3 + c_2)\Omega_0 \tag{B6c}$$

and

$$b_0 = (b_1 + b_2)b_6 - 2b_3^2 \tag{B7a}$$

$$e_0 = e_{333}^* + 2e_{311}^*, \quad \bar{e}_0 = e_{333}^* - e_{311}^*$$
(B7b)

$$e_1 = b_3 e_{333}^* - b_b e_{311}^*, \quad e_2 = (b_1 + b_2) e_{333}^* - 2b_3 e_{311}^*$$
(B7c)

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