Effective properties of spherically anisotropic piezoelectric composites

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Piezoelectric composites consisting of spherically anisotropic piezoelectric inclusions (i.e., piezoceramic material) in an infinite nonpiezoelectric matrix under a uniform electric field are theoretically investigated. Analytical solutions for the elastic displacements and the electric potentials are derived exactly. Taking account of the coupling effects of elasticity, permittivity, and piezoelectricity, formulas are derived for the effective dielectric and piezoelectric responses in the dilute limit. A piezoelectric response mechanism is revealed, in which the effective piezoelectric response vanishes irrespective of how much spherically anisotropic piezoelectric inclusions are inside. Moreover, the effective coupled responses of the piezoelectric composites show that the effective dielectric responses decrease (increase) as the inclusion elastic (piezoelectric) constants increase.

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I. INTRODUCTION

Piezoelectric composites have been extensively investigated theoretically due to their technological importance.[1–6] In particular, the effective responses of piezoelectric composites can be used to design smart materials, which have wide applications in ultrasonic transducers, underwater acoustics, biomedical imaging, etc.[2] Due to these applications, the theoretical and experimental investigations of the effective properties of piezoelectric composites become very important. Recently, many authors focused their attention on the analysis and estimation of the effective response of piezoelectric composites based on various methods. For example, Furukawa *et al.*[7] gave approximate expressions for the effective piezoelectric response of 0–3 composites based on the theoretical and experimental investigations. Under the assumption of very large dielectric constant of the inclusions, Jayasundere *et al.*[8] derived an effective piezoelectric formula for binary composites. Olson *et al.*[9] and Wong *et al.*[5,10,11] obtained explicit formulas for the effective piezoelectric coefficients for ferroelectric 0–3 composites and discussed the effects of electric conductivity on the effective dielectric and piezoelectric responses by effective-medium approximations. For piezoelectric fibrous composites, Benveniste and co-worker[12–14] derived a set of results for discussing universal relations between the effective responses and the local fields by means of uniform field and virtual work theorems. In addition, using the Green's function method and Eshelby's tensors, Chen,[15] Wang,[16] and Dunn and Taya[17] discussed the effective responses of the piezoelectric composites. While the above investigations had made a significant contribution to the analysis of the effective responses of piezoelectric composites, there are, however, few analytical results for piezoelectric composites due to the complexity of the piezoelectric problems. In this paper, we attempt to derive analytical solutions in the case of spherically anisotropic piezoelectric inclusions embedded in an infinite nonpiezoelectric matrix and to investigate the effective response mechanism for this kind of composites.

This research has a practical background because there are already many piezoelectric composites formed by piezoelectric inclusions suspended in nonpiezoelectric matrix. This kind of piezoelectric composites has been used in electroelastic sensors. Theoretically, for transversely isotropic piezoelectric composites, Furukawa *et al.*[18] and Furukawa and Fukuda[19] investigated the piezoelectric properties in the dilute limit, and Jiang *et al.*[20] obtained closed-form solutions for effective electroelastic moduli.

Despite spherically anisotropic piezoelectric materials have not been prepared experimentally, they were investigated theoretically by Kirichok[21] and the vibration property in rotating spherically anisotropic piezoceramic material was studied by Chen and Ding.[22] However, for spherically anisotropic piezoelectric composites, there are no investigations of their effective responses except for the elastoelectric field and thermoelastic problem.[23,24] Our aims are to study the dielectric and piezoelectric responses of the spherically anisotropic piezoelectric composites and to disclose their response mechanisms. In this paper, the piezoelectric composites with spherically anisotropic piezoelectric inclusions suspended in nonpiezoelectric matrix are treated theoretically, and analytical solutions for the elastic displacement and electric potential are derived. Based on these solutions, the formulas for effective dielectric and piezoelectric constants are formulated in the dilute limit of inclusion concentration.

In Sec. II, analytical solutions for the elastic displacement and the electric potentials are derived exactly for the spherically anisotropic piezoelectric composites having an isotropic nonpiezoelectric matrix under a uniform electric field. In Sec. III, effective dielectric and piezoelectric response formulas are given in the dilute limit and numerical results are performed to discuss the effects of elastic and piezoelectric properties on the effective dielectric responses. In Sec. IV, a brief conclusion is given.

II. ANALYTICAL SOLUTIONS OF A SPHERICALLY ANISOTROPIC PIEZOELECTRIC COMPOSITE

Consider an infinite isotropic nonpiezoelectric matrix containing a spherically anisotropic piezoelectric particle, where the origin of a spherical coordinate system is located at the center of the spherical inclusion. The matrix is assumed to be elastically and dielectrically isotropic. The constitutive equations in the inclusion and host regions are, respectively,

$$\sigma^i_{ij} = c^i_{ijkl}\gamma^j_{kl} - e^i_{kij}E^i_k, \quad D^i_i = e^i_{ikl}\gamma^j_{kl} + \varepsilon^i_{ik}E^i_k \quad \text{in } \Omega_i, \quad (1)$$

$$\sigma^h_{ij} = c^h_{ijkl}\gamma^h_{kl}, \quad D^h_i = \varepsilon^h_{ik}E^h_k \quad \text{in } \Omega_h, \quad (2)$$

where Ω_h (Ω_i) denotes the region occupied by matrix (inclusion), the subscripts $i,j,k,l=1,2,3$ denote the θ, φ, r directions, respectively, and the superscripts i and h denote the quantities in the inclusion and the host regions, respectively. σ, γ, D, and E are the stress, strain, electric displacement, and electric field, respectively, and c, e, and ε are the elastic stiffness, piezoelectric coefficient, and dielectric constant, respectively. In the absence of both the body forces and free electric charges, the governing equations in the inclusion and host regions are $\sigma^p_{ij,j}=0$ and $D^p_{i,i}=0$ ($p=i,h$), and the boundary conditions at the interface between inclusion and matrix are the continuity of the elastic displacement, electric potential, normal traction, and electric displacement. In spherical coordinates, the governing equations are

$$\frac{\partial\sigma_{rr}}{\partial r} + \frac{1}{r}\frac{\partial\sigma_{r\theta}}{\partial\theta} + \frac{1}{r\sin\theta}\frac{\partial\sigma_{\varphi r}}{\partial\varphi} + \frac{2\sigma_{rr} - \sigma_{\theta\theta} - \sigma_{\varphi\varphi} + \sigma_{r\theta}\cot\theta}{r} = 0,$$

$$\frac{\partial\sigma_{r\theta}}{\partial r} + \frac{1}{r}\frac{\partial\sigma_{\theta\theta}}{\partial\theta} + \frac{1}{r\sin\theta}\frac{\partial\sigma_{\theta\varphi}}{\partial\varphi} + \frac{(\sigma_{\theta\theta} - \sigma_{\varphi\varphi})\cot\theta + 3\sigma_{r\theta}}{r} = 0,$$

$$\frac{\partial\sigma_{\varphi r}}{\partial r} + \frac{1}{r}\frac{\partial\sigma_{\theta\varphi}}{\partial\theta} + \frac{1}{r\sin\theta}\frac{\partial\sigma_{\varphi\varphi}}{\partial\varphi} + \frac{2\sigma_{\theta\varphi}\cot\theta + 3\sigma_{\varphi r}}{r} = 0,$$

$$\frac{1}{r^2}\frac{\partial}{\partial r}(r^2 D_r) + \frac{1}{r\sin\theta}\frac{\partial(D_\theta\sin\theta)}{\partial\theta} + \frac{1}{r\sin\theta}\frac{\partial D_\varphi}{\partial\varphi} = 0.$$

In general, for a spherical particle with a radial polarization (i.e., a spherically anisotropic piezoelectric particle), the constitutive relations in the inclusion region in spherical coordinates are given as follows:[1]

$$\sigma_{\theta\theta} = c_{11}\gamma_{\theta\theta} + c_{12}\gamma_{\varphi\varphi} + c_{13}\gamma_{rr} - e_{31}E_r,$$

$$\sigma_{\varphi\varphi} = c_{12}\gamma_{\theta\theta} + c_{11}\gamma_{\varphi\varphi} + c_{13}\gamma_{rr} - e_{31}E_r,$$

$$\sigma_{rr} = c_{13}\gamma_{\theta\theta} + c_{13}\gamma_{\varphi\varphi} + c_{33}\gamma_{rr} - e_{33}E_r,$$

$$\sigma_{\varphi r} = 2c_{44}\gamma_{\varphi r} - e_{15}E_\varphi,$$

$$\sigma_{r\theta} = 2c_{44}\gamma_{r\theta} - e_{15}E_\theta,$$

$$\sigma_{\theta\varphi} = (c_{11} - c_{12})\gamma_{\varphi\theta},$$

$$D_\theta = 2e_{15}\gamma_{r\theta} + \varepsilon_{11}E_\theta,$$

$$D_\varphi = 2e_{15}\gamma_{\varphi r} + \varepsilon_{11}E_\varphi,$$

$$D_r = e_{31}\gamma_{\theta\theta} + e_{31}\gamma_{\varphi\varphi} + e_{33}\gamma_{rr} + \varepsilon_{33}E_r, \quad (3)$$

where we have omitted the superscript i (inclusion) for convenience. The subscripts i, j for the coefficients c_{ij} and e_{ij} in Eqs. (3) are used according to Nye's rule.[1] Because the matrix is a nonpiezoelectric and isotropic material, the constitutive relations in the host region are obtained from Eqs. (3) by simply omitting the piezoelectric coefficients e_{ij} and letting $c_{ijkl}=\lambda\delta_{ij}\delta_{kl}+\mu(\delta_{ik}\delta_{jl}+\delta_{il}\delta_{jk})$ and $\varepsilon_{11}=\varepsilon_{22}=\varepsilon_{33}$, where λ and μ are the Lame constants and δ_{ij} is the Kronecker delta. In spherical coordinates, the strain tensor γ can be expressed in terms of the elastic displacements:

$$\gamma_{rr} = \frac{\partial u_r}{\partial r},$$

$$\gamma_{\theta\theta} = \frac{1}{r}\left[\frac{\partial u_\theta}{\partial\theta} + u_r\right],$$

$$\gamma_{\varphi\varphi} = \frac{u_r}{r} + \frac{u_\theta\cot\theta}{r} + \frac{1}{r\sin\theta}\frac{\partial u_\varphi}{\partial\varphi},$$

$$\gamma_{r\theta} = \frac{1}{2}\left[\frac{1}{r}\frac{\partial u_r}{\partial\theta} + \frac{\partial u_\theta}{\partial r} - \frac{u_\theta}{r}\right],$$

$$\gamma_{\theta\varphi} = \frac{1}{2}\left[\frac{1}{r\sin\theta}\frac{\partial u_\theta}{\partial\varphi} + \frac{1}{r}\frac{\partial u_\varphi}{\partial\theta} - \frac{u_\varphi\cot\theta}{r}\right],$$

$$\gamma_{\varphi r} = \frac{1}{2}\left[\frac{\partial u_\varphi}{\partial r} - \frac{u_\varphi}{r} + \frac{1}{r\sin\theta}\frac{\partial u_r}{\partial\varphi}\right].$$

In the following, we will derive analytical solutions for the elastic displacement and the electric potential in the piezoelectric composite under a uniform electric field E_0 along the $\hat{z}$ direction.

In this case, owing to the symmetry about the $\hat{z}$ axis, the electric potential and the elastic displacement in the inclusion region can be written as

$$u^i_r = f_1(r)\cos\theta, \quad u^i_\theta = f_2(r)\sin\theta, \quad u^i_\varphi = 0, \quad \Phi^i = f_3(r)\cos\theta, \quad (4)$$

where $f_i(r)$ are the unknown functions. Using the elastic strain-displacement relations and substituting Eq. (4) into Eq. (3), we obtained three equations to determine these unknown functions:

$$c_{33}f_1'' + 2c_{33}(f_1'/r) + 2(c_{13} - c_{12} - c_{11} - c_{44})(f_1/r^2) + (2c_{13} + 2c_{44})(f_2'/r) + 2(c_{13} - c_{12} - c_{11} - c_{44})(f_2/r^2) + e_{33}f_3'' + 2(e_{33} - e_{31})(f_3'/r) - 2e_{15}(f_3/r^2) = 0, \quad (5)$$

$$-(c_{44} + c_{13})(f_1'/r) + (-2c_{44} - c_{11} - c_{12})(f_1/r^2) + c_{44}f_2'' + 2c_{44}(f_2'/r) + (-2c_{44} - c_{11} - c_{12})(f_2/r^2) - (e_{15} + e_{31}) \times (f_3'/r) - 2e_{15}(f_3/r^2) = 0, \quad (6)$$

$$e_{33}f_1'' + 2(e_{31}+e_{33})(f_1'/r) + 2(e_{31}-e_{15})(f_1/r^2) + 2(e_{31}+e_{15}) \times (f_2'/r) + 2(e_{31}-e_{15})(f_2/r^2) - \varepsilon_{33}f_3'' - 2\varepsilon_{33}(f_3'/r) + 2\varepsilon_{11}(f_3/r^2) = 0, \quad (7)$$

where we have used symbols f_i' and f_i'' to denote the first-order and the second-order derivatives with respect to the radial variable r, respectively. These equations are rewritten in a compact form,

$$\beta_{ji}^2 F_i^2 + \beta_{ji}^1 F_i^1 + \beta_{ji}^0 F_i^0 = 0, \quad j = 1,2,3, \quad (8)$$

where $F_i^2 = f_i''(r)$, $F_i^1 = (f_i'(r)/r)$, and $F_i^0 = (f_i(r)/r^2)$. The coefficients β_{ij}^2, β_{ij}^1, and β_{ij}^0 are listed as follows:

$$\beta_{11}^2 = c_{33}, \quad \beta_{11}^1 = 2c_{33}, \quad \beta_{11}^0 = 2(c_{13}-c_{12}-c_{11}-c_{44}),$$

$$\beta_{12}^2 = 0, \quad \beta_{12}^1 = 2(c_{13}+c_{44}), \quad \beta_{12}^0 = 2(c_{13}-c_{12}-c_{11}-c_{44}),$$

$$\beta_{13}^2 = e_{33}, \quad \beta_{13}^1 = 2(e_{33}-e_{31}), \quad \beta_{13}^0 = -2e_{15},$$

$$\beta_{21}^2 = 0, \quad \beta_{21}^1 = -(c_{44}+c_{13}), \quad \beta_{21}^0 = (-2c_{44}-c_{11}-c_{12}),$$

$$\beta_{22}^2 = c_{44}, \quad \beta_{22}^1 = 2c_{44}, \quad \beta_{22}^0 = (-2c_{44}-c_{11}-c_{12}),$$

$$\beta_{23}^2 = 0, \quad \beta_{23}^1 = -(e_{15}+e_{31}), \quad \beta_{23}^0 = -2e_{15},$$

$$\beta_{31}^2 = e_{33}, \quad \beta_{31}^1 = 2(e_{31}+e_{33}), \quad \beta_{31}^0 = 2(e_{31}-e_{15}),$$

$$\beta_{32}^2 = 0, \quad \beta_{32}^1 = 2(e_{31}+e_{15}), \quad \beta_{32}^0 = 2(e_{31}-e_{15}),$$

$$\beta_{33}^2 = -\varepsilon_{33}, \quad \beta_{33}^1 = -2\varepsilon_{33}, \quad \beta_{33}^0 = 2\varepsilon_{11}.$$

The unknown functions can be solved simply by setting $f_i(r) = A_i r^t$ and substituting them into Eq. (8), and the procedure leads to

$$\alpha_{ji} A_i = 0, \quad j = 1,2,3, \quad (9)$$

where the matrix $\alpha_{ji} \equiv \beta_{ji}^2 t(t-1) + \beta_{ji}^1 t + \beta_{ji}^0$. For nontrivial coefficients A_i, the condition $|\alpha_{ij}| = 0$ must be satisfied, and t can be solved from this condition. Let t_1^+, t_2^+, and t_3^+ denote the three real roots of Eq. (9). Thus, we have

$$f_i(r) = A_i^{+,1} r^{t_1^+} + A_i^{+,2} r^{t_2^+} + A_i^{+,3} r^{t_3^+}. \quad (10)$$

In fact, the coefficients $A_1^{+,j}$ and $A_2^{+,j}$ can be expressed in terms of $A_3^{+,j}$:

$$A_1^{+,j} = \alpha_{1,3}^j (t_j^+) A_3^{+,j},$$

$$A_2^{+,j} = \alpha_{2,3}^j (t_j^+) A_3^{+,j},$$

where $\alpha_{1,3}^j(t_j^+)$ and $\alpha_{2,3}^j(t_j^+)$ $(j=1,2,3)$ are solved from Eq. (9). The unknown coefficients $A_3^{+,j}$ are determined by applying the boundary conditions, i.e., matching with the solutions for the host regions. The elastic displacement, electric potential, stress, and electric displacement in inclusion region can be expressed in terms of the unknown coefficients $A_3^{+,j}$:

$$u_r^i = (\alpha_{1,3}^1 A_3^{+,1} r^{t_1^+} + \alpha_{1,3}^2 A_3^{+,2} r^{t_2^+} + \alpha_{1,3}^3 A_3^{+,3} r^{t_3^+}) \cos\theta,$$

$$u_\theta^i = (\alpha_{2,3}^1 A_3^{+,1} r^{t_1^+} + \alpha_{2,3}^2 A_3^{+,2} r^{t_2^+} + \alpha_{2,3}^3 A_3^{+,3} r^{t_3^+}) \sin\theta,$$

$$\Phi^i = (A_3^{+,1} r^{t_1^+} + A_3^{+,2} r^{t_2^+} + A_3^{+,3} r^{t_3^+}) \cos\theta,$$

$$\sigma_{rr}^i = \frac{1}{r}[\lambda_{rr}^1 A_3^{+,1} r^{t_1^+} + \lambda_{rr}^2 A_3^{+,2} r^{t_2^+} + \lambda_{rr}^3 A_3^{+,3} r^{t_3^+}] \cos\theta,$$

$$\sigma_{r\theta}^i = \frac{1}{r}[\lambda_{r\theta}^1 A_3^{+,1} r^{t_1^+} + \lambda_{r\theta}^2 A_3^{+,2} r^{t_2^+} + \lambda_{r\theta}^3 A_3^{+,3} r^{t_3^+}] \sin\theta,$$

$$D_r^i = \frac{1}{r}[\lambda_D^1 A_3^{+,1} r^{t_1^+} + \lambda_D^2 A_3^{+,2} r^{t_2^+} + \lambda_D^3 A_3^{+,3} r^{t_3^+}] \cos\theta, \quad (11)$$

where the new notations are defined by

$$\lambda_{rr}^k = (2c_{13}\alpha_{1,3}^k + 2c_{13}\alpha_{2,3}^k + c_{33}\alpha_{1,3}^k t_k^+ + e_{33} t_k^+),$$

$$\lambda_{r\theta}^k = (-c_{44}\alpha_{1,3}^k + c_{44}\alpha_{2,3}^k t_k^+ - c_{44}\alpha_{2,3}^k - e_{15}),$$

$$\lambda_D^k = (2e_{31}\alpha_{1,3}^k + 2e_{31}\alpha_{2,3}^k + e_{33}\alpha_{1,3}^k t_k^+ - \varepsilon_{33} t_k^+).$$

As the host region is an isotropic material, the elastic displacement and the electric potential under an external electric field along the $\hat{z}$ direction can be obtained by using Goodier's method,[25]

$$u_r^h = \left[-2\frac{B_1}{r^2} - 2B_2 + \alpha_{-1}B_2\right]\frac{\cos\theta}{r},$$

$$u_\theta^h = -\left[\frac{B_1}{r^2} + B_2\right]\frac{\sin\theta}{r},$$

$$\Phi^h = [-E_0 r + B_3/r^2]\cos\theta, \quad (12)$$

where $\alpha_{-1} = (10-12\nu^h)/(3-4\nu^h)$, $2\nu^h = \lambda^h/(\mu^h+\lambda^h)$. Here λ^h and μ^h are the Lame constants in the host region. The unknown coefficients B_i can be determined by applying the boundary conditions. The elastic stress tensor and electric displacement in the host region are given by

$$\sigma_{rr}^h = 2\mu^h\left[\left(\frac{2-8\nu^h}{1-2\nu^h} + \frac{-1+3\nu^h}{1-2\nu^h}\alpha_{-1}\right)B_2 + 6\frac{B_1}{r^2}\right]\frac{\cos\theta}{r^2},$$

$$\sigma_{r\theta}^h = \mu^h\left[\frac{6B_1}{r^2} + 4B_2 - \alpha_{-1}B_2\right]\frac{\sin\theta}{r^2},$$

$$D_r^h = \varepsilon^h[E_0 + 2B_3/r^3]\cos\theta. \quad (13)$$

Next, we apply the boundary conditions at the surface of inclusion to solve the unknown coefficients $A_3^{+,j}$ and B_i. The boundary conditions at the surface of a spherical inclusion with radius a are as follows:

$$u_r^i(r) = u_r^h(r)|_{r=a}, \quad u_\theta^i(r) = u_\theta^h(r)|_{r=a},$$

$$u_\varphi^i(r) = u_\varphi^h(r)|_{r=a}, \quad \sigma_{rr}^i(r) = \sigma_{rr}^h(r)|_{r=a},$$

$$\sigma_{r\theta}^i(r) = \sigma_{r\theta}^h(r)|_{r=a}, \quad \sigma_{r\varphi}^i(r) = \sigma_{r\varphi}^h(r)|_{r=a},$$

$$\Phi^i(r) = \Phi^h(r)|_{r=a}, \quad D_r^i(r) = D_r^h(r)|_{r=a}.$$

Thus, we obtained a set of algebraic equations for the six unknown coefficients $A_3^{+,j}$ and B_i:

$$\bar{\lambda}_{rr}^1 A_3^{+,1} a^{t_1^+ +1} + \bar{\lambda}_{rr}^2 A_3^{+,2} a^{t_2^+ +1} + \bar{\lambda}_{rr}^3 A_3^{+,3} a^{t_3^+ +1} = 0,$$

$$\bar{\lambda}_{r\theta}^1 A_3^{+,1} a^{t_1^+ +1} + \bar{\lambda}_{r\theta}^2 A_3^{+,2} a^{t_2^+ +1} + \bar{\lambda}_{r\theta}^3 A_3^{+,3} a^{t_3^+ +1} = 0,$$

$$\bar{\lambda}_D^1 A_3^{+,1} a^{t_1^+ -1} + \bar{\lambda}_D^2 A_3^{+,2} a^{t_2^+ -1} + \bar{\lambda}_D^3 A_3^{+,3} a^{t_3^+ -1} = 3E_0,$$

$$\beta_1^1 A_3^{+,1} a^{t_1^+ +3} + \beta_1^2 A_3^{+,2} a^{t_2^+ +3} + \beta_1^3 A_3^{+,3} a^{t_3^+ +3} = B_1,$$

$$\beta_2^1 A_3^{+,1} a^{t_1^+ +1} + \beta_2^2 A_3^{+,2} a^{t_2^+ +1} + \beta_2^3 A_3^{+,3} a^{t_3^+ +1} = B_2,$$

$$(3+\bar{\lambda}_D^1) A_3^{+,1} a^{t_1^+ +2} + (3+\bar{\lambda}_D^2) A_3^{+,2} a^{t_2^+ +2} + (3+\bar{\lambda}_D^3) A_3^{+,3} a^{t_3^+ +2} = 3B_3, \quad (14)$$

where the new notations are defined by

$$\bar{\lambda}_{rr}^k = \lambda_{rr}^k/(2\mu^h) - 6\beta_1^k - \beta_2^k[2 - 8\nu^h + \alpha_{-1}(3\nu^h - 1)]/(1 - 2\nu^h),$$

$$\beta_1^k = (-\alpha_{1,3}^k + 2\alpha_{2,3}^k - \alpha_{-1}\alpha_{2,3}^k)/\alpha_{-1},$$

$$\beta_2^k = (\alpha_{1,3}^k - 2\alpha_{2,3}^k)/\alpha_{-1},$$

$$\bar{\lambda}_{r\theta}^k = \lambda_{r\theta}^k/\mu^h - 6\beta_1^k - (4 - \alpha_{-1})\beta_2^k,$$

$$\bar{\lambda}_D^k = -2 + \lambda_D^k/\varepsilon^h.$$

Therefore, analytical solutions of the piezoelectric composite under an electric field along the $\hat{z}$ direction are derived. Similarly, the analytical solutions for an electric field in the $\hat{x}$ direction can be derived.

III. EFFECTIVE DIELECTRIC AND PIEZOELECTRIC RESPONSES

Based on the analytical solutions for the elastic displacements and the electric potentials for the piezoelectric composites, we will formulate a set of formulas to evaluate the effective dielectric constant ε^e, the effective elastic moduli c^e, and the effective piezoelectric constant e^e in these systems. As usual, these effective constants are connected by the effective constitutive relations: $\bar{\sigma}_{ij} = c_{ijkl}^e \bar{\gamma}_{kl} - e_{kij}^e \bar{E}_k$ and $\bar{D}_i = e_{ikl}^e \bar{\gamma}_{kl} + \varepsilon_{ik}^e \bar{E}_k$, where $\bar{A} = 1/V \int_{\Omega_i+\Omega_h} A dV$, and V is the total volume occupied by the composite. We build a set of equations to calculate the averages of these quantities,

$$\frac{1}{V}\int_{\Omega_i} [(c_{ijkl}^i - c_{ijkl}^h)\gamma_{kl} - (e_{kij}^i - e_{kij}^h)E_k] dV = \bar{\sigma}_{ij} - c_{ijkl}^h \bar{\gamma}_{kl} + e_{kij}^h \bar{E}, \quad (15)$$

$$\frac{1}{V}\int_{\Omega_i} [(e_{ikl}^i - e_{ikl}^h)\gamma_{kl} + (\varepsilon_{ik}^i - \varepsilon_{ik}^h)E_k] dV = \bar{D}_i - e_{ikl}^h \bar{\gamma}_{kl} - \varepsilon_{ik}^h \bar{E}_k. \quad (16)$$

Combining effective constitutive equations with Eqs. (15) and (16), we obtain

$$e_{ikl}^e \bar{\gamma}_{kl} + \varepsilon_{ik}^e \bar{E}_k = \varepsilon_{ik}^h \bar{E}_k + \frac{1}{V}\int_{\Omega_i} [e_{ikl}^i \gamma_{kl} + (\varepsilon_{ik}^i - \varepsilon_{ik}^h)E_k] dV, \quad (17)$$

$$c_{ijkl}^e \bar{\gamma}_{kl} - e_{kij}^e \bar{E}_k = c_{ijkl}^h \bar{\gamma}_{kl} + \frac{1}{V}\int_{\Omega_i} [(c_{ijkl}^i - c_{ijkl}^h)\gamma_{kl} - e_{kij}^i E_k] dV. \quad (18)$$

In order to estimate the effective dielectric constant ε_{zz}^e, we apply an electric field E_0 along the $\hat{z}$ direction. At dilute limit of the inclusion concentration, the formula for this effective dielectric constant is

$$\varepsilon_{zz}^e = \varepsilon_{zz}^h + \frac{1}{VE_0}\int_{\Omega_i} [e_{zkl}^i \gamma_{kl} + (\varepsilon_{zk}^i - \varepsilon_{zk}^h)E_k] dV, \quad k,l = x,y,z. \quad (19)$$

Meanwhile, the piezoelectric constant e_{zij}^e can be estimated as well,

$$e_{zij}^e = -\frac{1}{VE_0}\int_{\Omega_i} [(c_{ijkl}^i - c_{ijkl}^h)\gamma_{kl} - e_{kij}^i E_k] dV, \quad i,j,k,l = x,y,z. \quad (20)$$

Here, the quantities in Eqs. (19) and (20) are expressed in the Cartesian coordinates. Using transformations between the components in the spherical coordinates and the Cartesian coordinates, we have

$$D_z^i = (e_{31}\gamma_{\theta\theta} + e_{31}\gamma_{\varphi\varphi} + e_{33}\gamma_{rr} + \varepsilon_{33}E_r)\cos\theta - (2e_{15}\gamma_{r\theta} + \varepsilon_{11}E_\theta)\sin\theta. \quad (21)$$

Furthermore, we get the following formulas:

$$\varepsilon_{zk}^h E_k = \varepsilon_{rk}^h E_k \cos\theta - \varepsilon_{\theta k}^h E_k \sin\theta, \quad (22)$$

$$e_{zkl}^i \gamma_{kl} = (e_{31}\gamma_{\theta\theta} + e_{31}\gamma_{\varphi\varphi} + e_{33}\gamma_{rr})\cos\theta - (2e_{15}\gamma_{r\theta})\sin\theta. \quad (23)$$

Substituting Eqs. (21)–(23) into Eq. (19), we derive the analytical formula for the effective dielectric constant,

$$\varepsilon_{zz}^e = \varepsilon^h - p(\varepsilon_{rr} - \varepsilon^h)(A_3^{+,1}\alpha_1 t_1^+ + A_3^{+,2}\alpha_2 t_2^+ + A_3^{+,3}\alpha_3 t_3^+) - 2p(\varepsilon_{\theta\theta} - \varepsilon^h)(A_3^{+,1}\alpha_1 + A_3^{+,2}\alpha_2 + A_3^{+,3}\alpha_3) + p(\varsigma_1 A_3^{+,1} + \varsigma_2 A_3^{+,2} + \varsigma_3 A_3^{+,3}) - 2pe_{15}(\eta_1 A_3^{+,1} + \eta_2 A_3^{+,2} + \eta_3 A_3^{+,3}), \quad (24)$$

where p is the volume fraction of the inclusions, and the definitions of the new notations are

$$\alpha_k = a^{t_k^+ -1}/(t_k^+ + 2),$$

$$\varsigma_k = a^{t_k^+-1}[2e_{31}(\alpha_{1,3}^k + \alpha_{2,3}^k) + e_{33}t_k^+\alpha_{1,3}^k]/(t_k^+ + 2),$$

$$\eta_k = a^{t_k^+-1}(-\alpha_{1,3}^k - \alpha_{2,3}^k + t_k^+\alpha_{2,3}^k)/(t_k^+ + 2).$$

Equation (20) can be applied to estimate the effective piezoelectric response e_{zij}^e $(i,j=x,y,z)$. For example, e_{zzz}^e in the dilute limit is

$$e_{zzz}^e = -\frac{1}{VE_0}\int_{\Omega_i}[(c_{zzkl}^i - c_{zzkl}^h)\gamma_{kl} - e_{kzz}^i E_k]dV. \quad (25)$$

With the tensor transformations between the spherical coordinates and the Cartesian coordinates, we have

$$\sigma_{zz}^i = (c_{13}\gamma_{\theta\theta} + c_{13}\gamma_{\varphi\varphi} + c_{33}\gamma_{rr} - e_{33}E_r)\cos^2\theta - 2(2c_{44}\gamma_{r\theta} - e_{15}E_\theta)\cos\theta\sin\theta + (c_{11}\gamma_{\theta\theta} + c_{12}\gamma_{\varphi\varphi} + c_{13}\gamma_{rr} - e_{31}E_r)\sin^2\theta.$$

Substituting this formula into Eqs. (23) and (25) and considering the orthogonality, we can show

$$e_{zzz}^e = 0. \quad (26)$$

Similarly, we have derived $e_{zij}^e=0$ $(i,j=x,y,z)$. By applying electric field along the $\hat{x}$ direction, we can show that the effective piezoelectric constants e_{xij}^e and e_{yij}^e $(i,j=x,y,z)$ vanish as well. The effective piezoelectric responses that vanish in this case are due to the symmetry of spherically anisotropic piezoelectric composite system. When the spherically symmetry of the system is destroyed, the bulk effective piezoelectric properties will appear. For example, if a transversely isotropic piezoelectric spherical inclusion is immersed in a nonpiezoelectric matrix, there will be effective piezoelectric responses.[20]

In order to discuss the coupling effects of the elastic and piezoelectric properties on the effective dielectric constant, the effective dielectric constant ε_{zz}^e is calculated and the results are shown in Fig. 1. The volume fraction of the inclusion is $p=0.1$, and the elastic moduli and dielectric constant of the host material are $\nu^h=0.25$, $\mu^h=32$ GPa, and $\varepsilon^h=6\times10^{-9}$ C^2 N^{-1} m^{-2}. For the spherical inclusions, we introduce n to denote the variations in the elastic, piezoelectric, and dielectric constants: $c_{11}=16.6\times n\times$GPa, $c_{33}=16.2\times n\times$GPa, $c_{12}=7.7\times n\times$GPa, $c_{44}=4.3\times n\times$GPa, $c_{13}=7.8\times n\times$GPa, $e_{31}=-0.44\times n\times$C m^{-2}, $e_{33}=1.86\times n\times$C m^{-2}, $e_{15}=1.16\times n\times$C m^{-2}, $\varepsilon_{\theta\theta}=\varepsilon_{\varphi\varphi}=\varepsilon_{rr}=1.12\times n\times10^{-9}$ C^2 N^{-1} m^{-2}. In Fig. 1, the value of n is varied from 1 to 20 to show the effects on effective constants of the piezoelectric composite. For the "Elastic effect" curve in Fig. 1, n in elastic moduli varies from 1 to 20, while its value in dielectric and piezoelectric constants is set at 10. For the "Piezoelectric effect" curve in Fig. 1, n in piezoelectric coefficient varies from 1 to 20, while its value in dielectric constants and elastic moduli is taken as 10. For the "Dielectric effect" curve in Fig. 1, the value of n in dielectric constant changes from 1 to 20, and its value in piezoelectric constants is chosen as 10.

In order to test these formulas, we calculate the pure dielectric effect by using Eq. (24) and setting the piezoelectric constants zero. Thus, we can compare the result of Eq. (24) with that of classical Maxwell's effective dielectric response formula of spherical composites in the dilute limit. Excellent agreement is, indeed, shown in Fig. 1, where the dashed line and the solid line represent the results of Eq. (24) and Maxwell's dilute formula, respectively. The square-solid line (elastic effect) denotes the effect of the particle elastic moduli on the effective dielectric constant. It is clear that the effective dielectric constant decreases as the elastic moduli increase. This means that the elastic property of piezoelectric composites will reduce the bulk effective dielectric behavior. The effect of the piezoelectric property of inclusions on the effective dielectric property is denoted by the triangular-solid line (piezoelectric effect) in Fig. 1. The result shows that increasing the piezoelectric constant of the inclusion material enhances the effective dielectric constant. Of course, the effective dielectric constant also increases with the dielectric constant of inclusions. This is denoted by the circular-solid line in Fig. 1. In addition, we find that the effective dielectric response of the piezoelectric composite with fixed elastic and piezoelectric constants is larger than that of a pure dielectric composite by comparing the circular-solid line (piezoelectric composites having fixed elastic and piezoelectric constants) with the dashed line or the solid line (pure dielectric composites without piezoelectric properties). Moreover, for a fixed piezoelectric constant of the particles (n is set at 10), decreasing in elastic stiffness may induce a higher effective dielectric constant, as indicated by the square-solid line and the solid line in Fig. 1. These results show that the dielectric response of a piezoelectric composite is much more complex than that of a nonpiezoelectric composite. The results also indicate that there are, indeed, complex correlation and interactions among the elastic, the piezoelectric, and the dielectric properties.

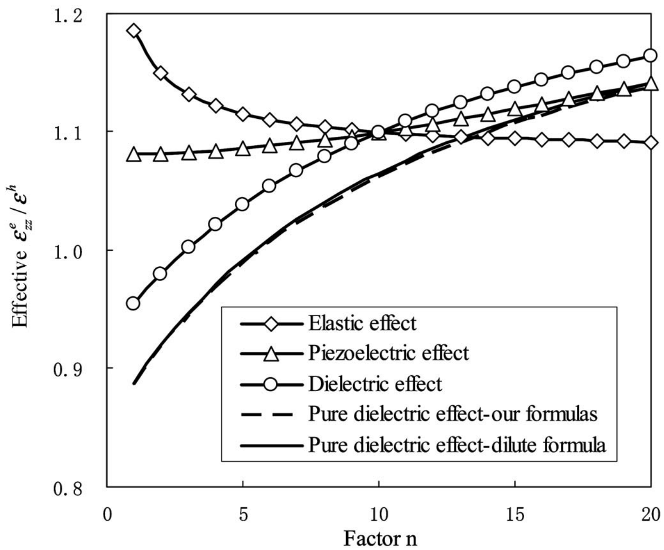

FIG. 1. The effective dielectric constants $\varepsilon_{zz}^e/\varepsilon^h$ versus the parameter factor n.

IV. CONCLUSIONS

Spherically anisotropic piezoelectric composites having a nonpiezoelectric matrix are investigated theoretically, and analytical solutions of the elastic displacement fields and electric potentials under an external electric field are derived. In the dilute limit, we have derived the effective dielectric and piezoelectric response formulas. The present work shows that spherically anisotropic piezoelectric composites do not have the bulk piezoelectric behavior due to the symmetry of the composite system and the radial polarization of the spherical particles. This means that piezoceramic composites can be designed for technological applications and the dielectric response of the composite can be enhanced by increasing the particle piezoelectric properties. However, we also found that if the elastic stiffness of the particles is too large, the effective dielectric response will be reduced. Also, we noted that if the elastic stiffness of the piezoelectric particles is reduced, it may increase the effective dielectric response because of the higher piezoelectric properties. This implies that the piezoelectric property of the particle material plays an important role in the coupled effective response of a piezoelectric composite. Moreover, based on the analytical solutions obtained, the effective response at higher concentration of the inclusions can be developed by means of an effective-medium approximation.

In this paper, we have derived the exact solution of the spherically anisotropic composites with nonpiezoelectric matrix. Furthermore, the spherically anisotropic properties of piezoelectric inclusions imply gradient profiles of properties in Cartesian coordinates. Thus, based on our results, one can also study the graded piezoelectric composites having graded inclusion materials,[26–30] so that the effective elastic, dielectric, and piezoelectric properties are controllable by changing the external electric field or the gradient profile of the materials. Furthermore, the nonlinear piezoelectric composites can be investigated because the electroelastic interactions are related to the nonlinear dielectric responses.[31–35]

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