

Thermoelectroelastic moduli of textured piezoelectric polycrystals: Exact solutions and bounds for film textures

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(Received 1 February 1999; accepted for publication 25 May 1999)

Conditions are given for the existence of exact solutions for the effective thermal and electroelastic moduli of polycrystals exhibiting fiber texture. The exact solutions are then developed through the consideration of a class of uniform fields in a polycrystal acted upon by various external thermal and electroelastic loadings. These exact results are verified by direct orientational averaging that shows the coincidence of the Voigt and Reuss bounds on the overall moduli. © 1999 American Institute of Physics. [S0021-8979(99)03317-4]

I. INTRODUCTION

This work is motivated by the increasing interest in piezoelectric and ferroelectric thin and thick films for a wide range of technological applications, and the realization of the importance of microstructural details on the overall properties of the films. Ferroelectric films are applied in electronic and electro-optic devices. Because of their high dielectric constants and breakdown voltages, BaTiO₃ films are the best storage dielectrics for ultralarge-scale integrated memory devices.¹ Preferentially oriented bulk bismuth titanate (BIT) films have received substantial attention for their applications in high temperature piezoelectric and integrated semi-conductive devices.² Ferroelectric piezoelectric transducer (PZT) films, because of their attractive piezoelectric and pyroelectric properties, are used in nonvolatile memories, thermal and ultrasonic image sensors, and surface acoustic wave filters.³

A prominent microstructural characteristic of many film systems is a columnar grain structure (highly oriented long needle-like grains). Sakashita *et al.*³ reported the fabrication of PZT films with the tetragonal perovskite structure and [001] texture. Stemmer *et al.*⁴ studied domain configurations in epitaxial ferroelectric PbTiO₃ films, where domains with the x_3 axis parallel and normal to the substrate surface were observed. A highly c -oriented BIT film was obtained on a silver foil, and was characterized by x -ray diffraction and transmission electron microscopy; no evidence of grain orientations other than [001] was found for one- through ten-layer films, however, when the number of layers was greater than ten, the [200] orientation was dominant.² The preferential orientation in the [200] direction is very attractive because the largest value of spontaneous polarization lies in x_1-x_3 plane and in the x_1 direction.

It is clear that piezoelectric and ferroelectric films often exist as polycrystals with a fiber texture where all of the

grains have a common crystallographic axis aligned normal to the film plane. This study is concerned with the connection between the characteristics of this microstructure and the overall linear thermal and electroelastic moduli of the film. A sound theoretical understanding of such microstructure-property relations is especially important because measurement of properties is often difficult for films. Such studies of the effective moduli of piezoelectric polycrystals are relatively limited.⁵⁻⁷ Because of the columnar grains and preferential orientation, the microgeometry of piezoelectric thin films is relatively simple and permits exact solutions for certain components of the overall moduli under some conditions. By exact solutions we mean instances where certain polycrystal moduli coincide with the corresponding single-crystal moduli. The development of such exact results is the objective of this study. As such, it shares the spirit of the studies of Benveniste⁶ (who obtained exact connections between polycrystal and crystal properties in two-dimensional piezoelectric polycrystals composed of single crystal of class 2 mm) and Kroner and Wawra⁸ (who obtained conditions for the coincidence of bounds in two-dimensional elastic polycrystals). Indeed, some of the results obtained here can be obtained from Benveniste's exact connections. In Sec. II we present the basic equations and notation used throughout. The exact solutions are derived in Sec. III, and then validated by direct orientational averaging to obtain Voigt and Reuss bounds that coincide in Sec. IV. Closed-form expressions for the Voigt and Reuss averages, are also presented in Sec. IV for cases when exact solutions do not exist. Finally some implications of the results along with some numerical results are presented in Sec. V. We note that although we only consider linear response, the results obtained here serve as a rigorous starting point for the study of more complicated nonlinear electroelastic constitutive response of ferroelectric films.

II. BASIC EQUATIONS AND NOTATION

We consider the piezoelectric, and thus inherently anisotropic, analog of the uncoupled theory of thermoelasticity,

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TABLE I. Restrictions on the electroelastic moduli of a piezoelectric crystal that result from rotational symmetry about the x_3 axis.

Crystal symmetry	Example crystals	Elastic constants	Piezoelectric constants	Dielectric constants
Twofold	Rochelle Salt, TGS	$S_{14}=S_{15}=S_{24}=S_{25}=S_{34}$ $=S_{35}=S_{46}=S_{56}=0$	$d_{11}=d_{12}=d_{13}=d_{16}=d_{21}$ $=d_{22}=d_{23}=d_{26}=d_{34}=d_{35}$ $=0$	$\tau_{13}=\tau_{23}=0$
Threefold	LiNbO ₃ ,	$S_{16}=S_{26}=S_{34}=S_{35}=S_{36}$ $=S_{45}=0,$ $S_{11}=S_{22}, S_{13}=S_{23},$ $S_{44}=S_{55}, 2S_{14}=S_{56},$ $2S_{25}=S_{46}, 2S_{24}=-S_{56},$ $2S_{15}=-S_{46},$ $S_{66}=2(S_{11}-S_{12})$	$d_{13}=d_{23}=d_{34}=d_{35}=d_{36}$ $=0,$ $d_{31}=d_{32}, d_{14}=-d_{25},$ $d_{15}=d_{24}, 2d_{11}=-d_{26},$ $2d_{12}=d_{26}, 2d_{22}=-d_{16},$ $d_{21}=-d_{22}$	$\tau_{12}=\tau_{13}$ $=\tau_{23}=0,$ $\tau_{11}=\tau_{22}$
Fourfold	BaTiO ₃ , PbZrO ₃ , PbTiO ₃	$S_{14}=S_{15}=S_{24}=S_{25}=S_{34}$ $=S_{35}=S_{36}=S_{45}=S_{46}$ $=S_{56}=0,$ $S_{11}=S_{22}, S_{13}=S_{23},$ $S_{44}=S_{55}, S_{16}=-S_{26}$	$d_{11}=d_{12}=d_{13}=d_{16}=d_{21}$ $=d_{22}=d_{23}=d_{26}=d_{34}=d_{35}$ $=d_{36}=0,$ $d_{14}=-d_{25}, d_{15}=d_{24},$ $d_{31}=d_{32}$	$\tau_{12}=\tau_{13}$ $=\tau_{23}=0,$ $\tau_{11}=\tau_{22}$
Sixfold		$S_{14}=S_{15}=S_{16}=S_{24}=S_{25}$ $=S_{26}=S_{34}=S_{35}=S_{36}$ $=S_{45}=S_{46}=S_{56}=0,$ $S_{11}=S_{22}, S_{13}=S_{23},$ $S_{44}=S_{55},$ $S_{66}=2(S_{11}-S_{12})$	$d_{11}=d_{12}=d_{13}=d_{16}=d_{21}$ $=d_{22}=d_{23}=d_{26}=d_{34}=d_{35}$ $=d_{36}=0,$ $d_{14}=-d_{25}, d_{15}=d_{24},$ $d_{31}=d_{32}$	$\tau_{12}=\tau_{13}$ $=\tau_{23}=0,$ $\tau_{11}=\tau_{22}$

where the electric and elastic fields are fully coupled, but temperature enters the problem only as a parameter through the constitutive equations. The field variables and material moduli are represented either by conventional indicial notation or by bold characters. The constitutive equations for the stationary linear response of a piezoelectric solid with general anisotropy can be expressed as:

$$\begin{aligned} \epsilon_p &= S_{pq} \sigma_q + d_{pk} E_k + \Delta_p \Theta, \\ D_i &= d_{iq} \sigma_q + \tau_{ik} E_k + \gamma_i \Theta, \end{aligned} \tag{1a}$$

or in an inverse form:

$$\begin{aligned} \sigma_p &= C_{pq} \epsilon_q - h_{pk} D_k + \lambda_p \Theta, \\ E_i &= -h_{iq} \epsilon_q + \beta_{ik} D_k + p_i \Theta. \end{aligned} \tag{1b}$$

In Eqs. (1a) and (1b) σ_p and ϵ_p are the elastic stress and strain, respectively; D_i and E_i are the electric displacement and field, respectively. S_{pq} , d_{iq} , and τ_{ik} are the elastic compliance tensor (measured in a constant electric field), the piezoelectric tensor, and the dielectric tensor (measured at a constant stress), respectively. Δ_p and γ_i are thermal expansion and pyroelectric coefficients, respectively, and Θ is the temperature change with respect to a reference temperature. The well known contracted notation for tensors is adopted.⁹ We introduce the matrix representation for these quantities:

$$\begin{aligned} \mathbf{X} &= \begin{bmatrix} \boldsymbol{\epsilon} \\ \mathbf{D} \end{bmatrix} \quad \mathbf{Y} = \begin{bmatrix} \boldsymbol{\sigma} \\ \mathbf{E} \end{bmatrix} \quad \mathbf{P} = \begin{bmatrix} \mathbf{S} & \mathbf{d}^t \\ \mathbf{d} & \boldsymbol{\tau} \end{bmatrix} \quad \mathbf{Q} = \begin{bmatrix} \mathbf{C} & -\mathbf{h}^t \\ -\mathbf{h} & \boldsymbol{\beta} \end{bmatrix} \\ \boldsymbol{\Gamma} &= \begin{bmatrix} \boldsymbol{\Delta} \\ \boldsymbol{\gamma} \end{bmatrix} \quad \boldsymbol{\Pi} = \begin{bmatrix} \boldsymbol{\lambda} \\ \mathbf{p} \end{bmatrix}, \end{aligned} \tag{2}$$

where the superscript t is used to denote the transpose of matrix, \mathbf{X} and \mathbf{Y} and 9×1 column vectors representing the

electroelastic field variables, \mathbf{P} and \mathbf{Q} are 9×9 matrices representing the electroelastic moduli, and $\boldsymbol{\Gamma}$ and $\boldsymbol{\Pi}$ are 9×1 column vectors representing the thermal moduli. The constitutive Eqs. (1a) and (1b) can then be rewritten as:

$$\begin{aligned} \mathbf{X} &= \mathbf{P}\mathbf{Y} + \boldsymbol{\Gamma}\Theta, \\ \mathbf{Y} &= \mathbf{Q}\mathbf{X} + \boldsymbol{\Pi}\Theta. \end{aligned} \tag{3}$$

By choosing different independent variables, other representations can be realized. We adopt the current representation because it leads to a positive definite energy function and moduli matrices \mathbf{P} and \mathbf{Q} , which are advantageous for the development of variational bounds.¹⁰

Assuming statistical homogeneity of a heterogeneous solid subjected to external loading consistent with the uniform fields \mathbf{Y}^0 with $\Theta=0$, the effective electroelastic moduli \mathbf{P}^* can be defined as:

$$\langle \mathbf{X} \rangle = \mathbf{P}^* \langle \mathbf{Y} \rangle, \tag{4}$$

where $\langle \cdot \rangle = \int (\cdot) d\Omega(\theta, \varphi, \phi)$ denotes an orientational volume average. Due to linearity:

$$\mathbf{Y}(\theta, \varphi, \phi) = \mathbf{A}(\theta, \varphi, \phi) \mathbf{Y}^0, \tag{5}$$

where (θ, φ, ϕ) are Euler angles that describe the orientation of a grain in the polycrystal (see, for example, Roe),¹¹ and $\mathbf{A}(\theta, \varphi, \phi)$ are the concentration tensors for a grain at the orientation (θ, φ, ϕ) . Substituting Eq. (5) into Eq. (4), combined with the constitutive Eq. (3) for individual grains at different orientations, and the average field theorems for heterogeneous piezoelectric solids,¹² the effective moduli can be written as:

$$\mathbf{P}^* = \langle \mathbf{P}(\theta, \varphi, \phi) \mathbf{A}(\theta, \varphi, \phi) \rangle. \tag{6}$$

TABLE II. Restrictions on the thermal and electroelastic moduli for the existence of uniform thermoelectroelastic fields under various loadings.

Loads	Elastic	Piezoelectric	Dielectric	Thermal	Symmetry
σ_1 or σ_2	$S_{11}=S_{22}, S_{13}=S_{23},$ $S_{66}=2(S_{11}-S_{12}),$ $S_{14}=S_{15}=S_{16}=S_{24}$ $=S_{25}=S_{26}=S_{36}=S_{46}$ $=S_{56}=0$	$d_{31}=d_{32}=d_{11}=d_{12}$ $=d_{21}=d_{22}=d_{16}=d_{26}$ $=d_{36}=0$	None	None	sixfold
σ_3 or σ_4 or σ_5	$S_{13}=S_{23}, S_{36}=0$ $S_{44}=S_{55}, S_{14}=S_{15}=S_{24}$ $=S_{25}=S_{45}=S_{46}=S_{56}$ $=0$	None $d_{15}=d_{24}, d_{14}=-d_{25}$	None None	None None	six, four, threefold six, fourfold
σ_6	$S_{11}=S_{22}, S_{13}=S_{23},$ $S_{66}=2(S_{11}-S_{12}),$ $S_{14}=S_{24}, S_{15}=S_{25},$ $S_{16}=S_{26}=S_{36}=S_{46}$ $=S_{56}=0$	$d_{31}=d_{32}, d_{11}=d_{12},$ $d_{21}=d_{22}, d_{16}=d_{26}$ $=d_{36}=0$	None	None	sixfold
E_1 or E_2	None	$d_{15}=d_{24}, d_{14}=-d_{25},$ $d_{11}=d_{12}=d_{21}=d_{22}$ $=d_{16}=d_{26}=0$	$\tau_{11}=\tau_{22},$ $\tau_{12}=0$	None	six, fourfold
E_3 or Θ	None None	$d_{31}=d_{32}, d_{36}=0$ None	None None	None $\Delta_1=\Delta_2,$ $\Delta_6=0$	six, four, threefold six, four, threefold

$\mathbf{P}(\theta, \varphi, \phi)$ are the electroelastic moduli of a grain at orientation (θ, φ, ϕ) expressed in the global (specimen) coordinate system. It is clear from Eq. (6) that estimation of effective moduli depends on the estimation of the concentration tensors $\mathbf{A}(\theta, \varphi, \phi)$. The simplest assumption is $\mathbf{A}(\theta, \varphi, \phi) = \mathbf{I}$, where in matrix notation \mathbf{I} is the 9×9 identity matrix. This amounts to assuming that the stress and electric field are uniform in the polycrystal and leads to:

$$\mathbf{P}^* = \langle \mathbf{P}(\theta, \varphi, \phi) \rangle. \tag{7}$$

Analogously, by assuming $\mathbf{X}(\theta, \varphi, \phi) = \mathbf{B}(\theta, \varphi, \phi)\mathbf{X}^0$, and letting $\mathbf{B}(\theta, \varphi, \phi) = \mathbf{I}$, we find:

$$\mathbf{P}^* = \langle \mathbf{P}^{-1}(\theta, \varphi, \phi) \rangle^{-1}, \tag{8}$$

where the superscript-1 denotes matrix inversion.

On the other hand, when the heterogeneous solid is subjected to the temperature change Θ with $\mathbf{Y}^0 = 0$, the effective thermal moduli can be defined by:

$$\langle \mathbf{X} \rangle = \mathbf{I}^* \Theta. \tag{9}$$

Introducing thermal concentration factors $\mathbf{a}(\theta, \varphi, \phi)$ defined by $\mathbf{Y}(\theta, \varphi, \phi) = \mathbf{a}(\theta, \varphi, \phi)\Theta$, the effective thermal moduli can be expressed as:

$$\mathbf{I}^* = \langle \mathbf{P}(\theta, \varphi, \phi)\mathbf{a}(\theta, \varphi, \phi) + \mathbf{I}(\theta, \varphi, \phi) \rangle. \tag{10}$$

This is achieved by substituting the constitutive Eq. (3) into Eq. (9), combined with the average field theorems. The simplest assumption $\mathbf{a}(\theta, \varphi, \phi) = 0$ yields:

$$\mathbf{I}^* = \langle \mathbf{I}(\theta, \varphi, \phi) \rangle. \tag{11}$$

The physical implication of this assumption is that no disturbances of the stress and electric fields develop in the polycrystal due to the temperature change.

Equations (7), (8), and (11) are exact when the electroelastic fields in the polycrystal caused by the external loading are uniform. Equations (7) and (8) are the piezoelectric analog of the Voigt and Reuss averages for heterogeneous elastic solids.^{13,14} Using a minimum potential theorem and the positive definiteness of \mathbf{P} , Li and Dunn¹⁰ have shown that the Voigt and Reuss estimates provide upper and lower bounds for the effective electroelastic moduli of heterogeneous piezoelectric solids. When there is an exact solution for the electroelastic moduli, the upper and lower bounds will then coincide. In Sec. III we pursue the conditions that lead to uniform electroelastic fields, and then exact solutions for the effective moduli, under various thermoelectroelastic loading.

III. CONDITIONS FOR UNIFORM FIELDS AND EXACT SOLUTIONS

We consider piezoelectric polycrystals with a fiber texture where all grains have the same symmetry axis aligned along the x_3 direction (normal to the plane for a planar film). For this class of materials, the individual grains respond in exactly the same way to the applied loading under certain conditions, regardless of their orientations, so that the elec-

TABLE III. Exact solutions for the thermoelectroelastic moduli for polycrystals with rotational symmetry around the x_3 axis.

Symmetry	Elastic	Piezoelectric	Dielectric	Thermal
Sixfold	All	All	All	All
Fourfold	$S_{13}=S_{23}, S_{55}=S_{44}, S_{33}$	$d_{14}=-d_{25}, d_{15}=d_{24}, d_{31}=d_{32}, d_{33}$	$\tau_{11}=\tau_{22}, \tau_{33}$	$\Delta_1=\Delta_2, \Delta_3$
Threefold	$S_{13}=S_{23}, S_{33}$	$d_{31}=d_{32}, d_{33}$	τ_{33}	$\Delta_1=\Delta_2, \Delta_3$
Twofold	None	None	None	None

troelastic fields in the polycrystalline are uniform, and the effective thermoelectroelastic moduli can be found exactly. To pursue the conditions for such uniform fields, consider a single crystal, possessing a six, four, three, or twofold symmetry axis parallel to the x_3 axis. The rotational symmetries of such a single crystal place restrictions on the electroelastic moduli, which are summarized in Table I. The restrictions on the thermal expansion coefficients are the same as those on the dielectric constants and the only nonzero pyroelectric constant is γ_3 . As a result, for crystals possessing six, four, three, or twofold symmetry, the constitutive Eq. (3) can be reduced to:

$$\begin{bmatrix} \epsilon_1 \\ \epsilon_2 \\ \epsilon_3 \\ \epsilon_4 \\ \epsilon_5 \\ \epsilon_6 \\ D_1 \\ D_2 \\ D_3 \end{bmatrix} = \begin{bmatrix} S_{11} & S_{12} & S_{13} & S_{14} & S_{15} & S_{16} & d_{11} & d_{21} & d_{31} \\ S_{12} & S_{22} & S_{23} & S_{24} & S_{25} & S_{26} & d_{12} & d_{22} & d_{32} \\ S_{13} & S_{23} & S_{33} & 0 & 0 & S_{36} & 0 & 0 & d_{33} \\ S_{14} & S_{24} & 0 & S_{44} & S_{45} & S_{46} & d_{14} & d_{24} & 0 \\ S_{15} & S_{25} & 0 & S_{45} & S_{55} & S_{56} & d_{15} & d_{25} & 0 \\ S_{16} & S_{26} & S_{36} & S_{46} & S_{56} & S_{66} & d_{16} & d_{26} & d_{36} \\ d_{11} & d_{12} & 0 & d_{14} & d_{15} & d_{16} & \tau_{11} & \tau_{12} & 0 \\ d_{21} & d_{22} & 0 & d_{24} & d_{25} & d_{26} & \tau_{12} & \tau_{22} & 0 \\ d_{31} & d_{32} & d_{33} & 0 & 0 & d_{36} & 0 & 0 & \tau_{33} \end{bmatrix} \times \begin{bmatrix} \sigma_1 \\ \sigma_2 \\ \sigma_3 \\ \sigma_4 \\ \sigma_5 \\ \sigma_6 \\ E_1 \\ E_2 \\ E_3 \end{bmatrix} + \begin{bmatrix} \Delta_1 \\ \Delta_2 \\ \Delta_3 \\ 0 \\ 0 \\ \Delta_6 \\ 0 \\ 0 \\ \gamma_3 \end{bmatrix} \Theta. \tag{12}$$

We now consider simple thermoelectroelastic loadings of the polycrystal, where only one of the ten σ_p , E_i , or Θ is nonzero, and determine under what conditions the corresponding strain and electrical displacement states in individual grains are invariant with respect to a rotation α about the x_3 direction of the grain. Since all grains have the same symmetry axis aligned along the x_3 direction, this invariance means that all grains respond in an identical way to the external loading. In such a situation, the grains do not disturb each other, so no elastic and electric field fluctuations develop. The corresponding strain and electrical displacement developed in the grains under a particular external loading

can be obtained from Eq. (12). The resulting electroelastic fields are invariant under the rotation α around the x_3 direction if, and only if, the thermoelectroelastic moduli involved in a particular loading do not change under such a rotation, so that they are identical before and after the tensor transformation. Here we demonstrate the derivation of these conditions for external loads σ_1 , E_1 , and Θ . For all other cases, only the final conditions are given and these are summarized in Table II. The exact solutions for the effective electroelastic moduli under a particular rotational symmetry are then summarized in Table III.

A. Case 1: Applied σ_1

When only σ_1 is applied, Eq. (12) shows that the corresponding strains and electric displacements are connected to σ_1 by S_{11} , S_{12} , S_{13} , S_{14} , S_{15} , S_{16} , d_{11} , d_{21} , and d_{31} . These constants are invariant under a rotation about the x_3 axis if the following equations, derived from the transformation laws for 4th and 3rd rank tensors, are satisfied:

$$\begin{aligned}
 & S_{11}(\cos^4 \alpha - 1) + S_{22} \sin^4 \alpha + (2S_{12} + S_{66})\cos^2 \alpha \sin^2 \alpha \\
 & \quad + 2 \cos \alpha \sin \alpha (S_{16} \cos^2 \alpha + S_{26} \sin^2 \alpha) = 0, \\
 & 2(S_{26} - S_{16})\cos 2\alpha + (S_{11} + S_{22} - 2S_{12} - S_{66})\sin 2\alpha = 0, \\
 & S_{36} \cos \alpha + (S_{23} - S_{13})\sin \alpha = 0, \\
 & S_{14}(\cos^3 \alpha - 1) + (S_{46} - S_{15})\cos^2 \alpha \sin \alpha + (S_{24} - S_{56})\sin^2 \alpha \\
 & \quad \times \cos \alpha - S_{25} \sin^3 \alpha = 0, \\
 & S_{15}(\cos^3 \alpha - 1) + (S_{14} + S_{56})\cos^2 \alpha \sin \alpha + (S_{25} + S_{46})\sin^2 \alpha \\
 & \quad \times \cos \alpha + S_{24} \sin^3 \alpha = 0, \\
 & S_{16}(\cos^4 \alpha - 1) - S_{26} \sin^4 \alpha + 3(S_{26} - S_{16})\cos^2 \alpha \sin^2 \alpha \\
 & \quad + \cos \alpha \sin \alpha [(S_{66} + 2S_{12} - 2S_{11})\cos^2 \alpha \\
 & \quad - (S_{66} + 2S_{12} - 2S_{22})\sin^2 \alpha] = 0, \\
 & d_{11}(\cos^3 \alpha - 1) + d_{22} \sin^3 \alpha + \cos \alpha \sin \alpha \\
 & \quad \times [(d_{16} + d_{21})\cos \alpha + (d_{12} + d_{26})\sin \alpha] = 0, \\
 & d_{21}(\cos^3 \alpha - 1) - d_{12} \sin^3 \alpha + \cos \alpha \sin \alpha \\
 & \quad \times [(d_{26} - d_{11})\cos \alpha + (d_{22} - d_{16})\sin \alpha] = 0, \\
 & (d_{32} - d_{31})\sin \alpha + d_{36} \cos \alpha = 0.
 \end{aligned}$$

In order to satisfy all of these conditions for any angle α , one can easily verify that it requires $S_{11} = S_{22}$, $S_{13} = S_{23}$, $S_{66} = 2(S_{11} - S_{12})$, $S_{14} = S_{15} = S_{16} = S_{24} = S_{25} = S_{26} = S_{36} = S_{46} = S_{56} = 0$, $d_{31} = d_{32}$, and $d_{11} = d_{12} = d_{21} = d_{22} = d_{16} = d_{26} = d_{36} = 0$. This requires that the crystal possess sixfold symmetry around the x_3 axis. Four, three, and twofold symmetry will not satisfy all the conditions. As a result, S_{11} , S_{12} , S_{13} , and d_{31} are exact in a polycrystal composed of crystals with a sixfold symmetry axis aligned in the x_3 direction.

B. Case 2: Applied E_1

When only E_1 is applied, the corresponding strains and electric displacements are connected to E_1 by d_{11} , d_{12} , d_{14} ,

d_{15} , d_{16} , τ_{11} , and τ_{12} . These constants are invariant under a rotation about the x_3 if the following equations, derived from the transformation laws for second and third rank tensors, are satisfied:

$$\begin{aligned}
 & d_{11}(\cos^3 \alpha - 1) + d_{22} \sin^3 \alpha \\
 & + \cos \alpha \sin \alpha [(d_{16} + d_{21}) \cos \alpha + (d_{12} + d_{26}) \sin \alpha] = 0, \\
 & d_{12}(\cos^3 \alpha - 1) + d_{21} \sin^3 \alpha \\
 & + \cos \alpha \sin \alpha [(d_{22} - d_{16}) \cos \alpha + (d_{11} - d_{26}) \sin \alpha] = 0, \\
 & (d_{24} - d_{15}) \cos \alpha - (d_{14} + d_{25}) \sin \alpha = 0, \\
 & (d_{14} + d_{25}) \cos \alpha + (d_{24} - d_{15}) \sin \alpha = 0, \\
 & d_{16}(\cos^3 \alpha - 1) - d_{26} \sin^3 \alpha + \cos \alpha \sin \alpha [(d_{26} + 2d_{12} - 2d_{11}) \\
 & \times \cos \alpha - (d_{16} + 2d_{21} - 2d_{22}) \sin \alpha] = 0, \\
 & 2\tau_{12} \cos \alpha + (\tau_{22} - \tau_{11}) \sin \alpha = 0, \\
 & 2\tau_{12} \sin \alpha + (\tau_{11} - \tau_{22}) \cos \alpha = 0.
 \end{aligned}$$

These conditions are satisfied if $d_{15} = d_{24}$, $d_{14} = -d_{25}$, $d_{11} = d_{12} = d_{21} = d_{22} = d_{16} = d_{26} = 0$, $\tau_{11} = \tau_{22}$, and $\tau_{12} = 0$, which requires that the crystal possess six or fourfold symmetry around the x_3 axis. Three and twofold symmetry will not satisfy all the conditions. As a result, d_{15} , d_{14} , and τ_{11} are exact in polycrystal composed of crystals with a six or fourfold symmetry axis aligned in the x_3 direction.

C. Case 3: Applied Θ

When there is only a temperature change applied, the corresponding strains and electric displacements are related to Θ by Δ_1 , Δ_2 , Δ_3 , Δ_6 , and γ_3 . These constants are invariant under a rotation about the x_3 axis if the following equations, derived from the transformation laws for first and second rank tensors, are satisfied:

$$\begin{aligned}
 & \Delta_6 \cos \alpha + (\Delta_2 - \Delta_1) \sin \alpha = 0, \\
 & \Delta_6 \sin \alpha + (\Delta_1 - \Delta_2) \cos \alpha = 0.
 \end{aligned}$$

These are satisfied if $\Delta_6 = 0$ and $\Delta_2 = \Delta_1$, which requires that the crystal possess six, four, or threefold symmetry. As a result, Δ_1 , Δ_3 , and γ_3 are exact in polycrystal composed of crystals with a six, four, or threefold symmetry axis aligned in x_3 direction.

For polycrystals composed of single crystals with a sixfold symmetry axis aligned along the sample x_3 axis, all of the effective thermoelectroelastic moduli are exact, and are equal to the single crystal values. As a result, rigorous upper and lower bounds on the electroelastic moduli should coincide. For polycrystals composed of single crystals with a twofold symmetry axis aligned along the x_3 axis, none of the effective moduli are exact. For four and threefold symmetry, a subset of the material moduli are exact. This is the case regardless of the overall symmetry of the polycrystal. It is worthwhile to mention that for threefold symmetry, $\tau_{11} = \tau_{22}$ is exact if the piezoelectric effect is absent. When piezoelectric effects exist, constraint on the piezoelectric constants when E_1 or E_2 is applied are not satisfied by crystals with threefold symmetry, even though constraints on the di-

electric constants are satisfied. The thermal expansion coefficients $\Delta_1 = \Delta_2$, however, are still exact, since the coupling between the thermal and electric field is one-way, i.e., the thermal field induces an electric field, but the electric field does not influence the thermal field.

IV. ORIENTATIONAL AVERAGES AND ELEMENTARY BOUNDS

In the previous section we showed that exact solutions exist for some of the thermoelectroelastic moduli of polycrystals with fiber texture. In this section, we develop expressions for bounds on these moduli under more general conditions when exact solutions do not exist. We show that the bounds coincide (with each other at the exact solution) when the criteria of the previous section are satisfied. To proceed, we describe the orientation distribution of grains in the polycrystal by an orientation distribution function (ODF) $w(\xi, \varphi, \phi)$, where $\xi = \cos \theta$, and θ , φ , and ϕ are Euler angles describing the orientation of a grain. The orientational average of a single crystal tensorial property \mathbf{H} weighted by the ODF is given by:

$$\langle \mathbf{H} \rangle = \int_0^{2\pi} \int_0^{2\pi} \int_{-1}^1 \mathbf{H}(\xi, \varphi, \phi) w(\xi, \varphi, \phi) d\xi d\varphi d\phi, \tag{13}$$

where $\mathbf{H}(\xi, \varphi, \phi)$ is the single crystal value of \mathbf{H} expressed in the sample coordinate system. To evaluate $\langle \mathbf{H} \rangle$, we follow Roe¹¹ and expand $w(\xi, \varphi, \phi)$ and $\mathbf{H}(\xi, \varphi, \phi)$ in a series of generalized associated Legendre functions:

$$w(\xi, \varphi, \phi) = \sum_{l=0}^{\infty} \sum_{m=-l}^l \sum_{n=-l}^l W_{lmn} Z_{lmn}(\xi) e^{-im\varphi} e^{-in\phi}, \tag{14}$$

$$\mathbf{H}(\xi, \varphi, \phi) = \sum_{l=0}^{\infty} \sum_{m=-l}^l \sum_{n=-l}^l \mathbf{H}_{lmn} Z_{lmn}(\xi) e^{-im\varphi} e^{-in\phi}. \tag{15}$$

Here $Z_{lmn}(\xi)$ are the generalized associated Legendre functions, and

$$\begin{aligned}
 W_{lmn} &= \frac{1}{4\pi^2} \int_0^{2\pi} \int_0^{2\pi} \int_{-1}^1 W(\xi, \varphi, \phi) \\
 &\times Z_{lmn}(\xi) e^{im\varphi} e^{in\phi} d\xi d\varphi d\phi, \tag{16}
 \end{aligned}$$

$$\begin{aligned}
 \mathbf{H}_{lmn} &= \frac{1}{4\pi^2} \int_0^{2\pi} \int_0^{2\pi} \int_{-1}^1 \mathbf{H}(\xi, \varphi, \phi) \\
 &\times Z_{lmn}(\xi) e^{im\varphi} e^{in\phi} d\xi d\varphi d\phi \tag{17}
 \end{aligned}$$

are the coefficients of the series. The texture coefficients W_{lmn} are thus sufficient to describe the orientation distribution function of the grains in the polycrystalline aggregate. Normalization of the ODF results in $W_{000} = 1/4\sqrt{2}\pi^2$. Using Eqs. (14) and (15), and taking into account the orthogonal property of $Z_{lmn}(\xi)$, Eq. (13) can be written as:

$$\langle \mathbf{H} \rangle = 4\pi^2 \sum_{l=0}^R \sum_{m=-l}^l \sum_{n=-l}^l \mathbf{H}_{lmn} W_{lmn}. \tag{18}$$

Here R is the rank of tensorial property \mathbf{H} . The range on the first summation results because only the first R terms in the series expansions of Eqs. (16) and (17) need to be considered in averaging a tensorial property of rank R .¹⁵

Let us now focus on polycrystals with a fiber texture, but otherwise arbitrarily oriented in the x_1 - x_2 plane, composed of 2 mm single crystals with electroelastic and thermal moduli of the following form:

$$\begin{bmatrix} S_{11} & S_{12} & S_{13} & 0 & 0 & 0 & 0 & 0 & d_{31} \\ S_{12} & S_{22} & S_{23} & 0 & 0 & 0 & 0 & 0 & d_{32} \\ S_{13} & S_{23} & S_{33} & 0 & 0 & 0 & 0 & 0 & d_{33} \\ 0 & 0 & 0 & S_{44} & 0 & 0 & 0 & d_{24} & 0 \\ 0 & 0 & 0 & 0 & S_{55} & 0 & d_{15} & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & S_{66} & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & d_{15} & 0 & \tau_{11} & 0 & 0 \\ 0 & 0 & 0 & d_{24} & 0 & 0 & 0 & \tau_{22} & 0 \\ d_{31} & d_{32} & d_{33} & 0 & 0 & 0 & 0 & 0 & \tau_{33} \end{bmatrix}$$

and

$$\begin{bmatrix} \Delta_1 \\ \Delta_2 \\ \Delta_3 \\ 0 \\ 0 \\ 0 \\ 0 \\ 0 \\ \gamma_3 \end{bmatrix} \quad (19)$$

This structure represents a broad class of piezoelectric crystals, where point groups 4 and 6 mm are special cases. Now, we assume that the polycrystal exhibits overall orthorhombic symmetry. For this combination of crystal and polycrystal symmetry, the nonzero independent texture coefficients are only W_{100} , W_{200} , W_{222} , W_{300} , W_{322} , W_{400} , W_{422} , and W_{444} . With this crystal and polycrystal symmetry, we computed the Voigt and Reuss averages for the effective moduli \mathbf{P} and \mathbf{Q} of Eq. (2) which are upper and lower bounds on \mathbf{P} and \mathbf{Q} . When the solution for a component of the effective moduli is exact, the Voigt and Reuss bounds coincide. The Voigt and Reuss averages for the effective electroelastic moduli are tabulated in Table IV. The averaging scheme for the thermal expansion coefficients is the same as that for dielectric constants. The average value for the pyroelectric coefficient is $\gamma_3^* = 4\sqrt{2/3}\pi^2\gamma_3W_{100}$. When $W_{lmn} = 0$, the material is isotropic, and there are no piezoelectric or pyroelectric effects. These effects are also absent when the sample possesses a center of symmetry which results in $W_{100} = W_{300} = W_{322} = 0$. The orientational averages in Table IV can be used to estimate the upper and lower bounds of electroelastic moduli of polycrystals with fiber texture.

The derivations of the nonzero texture coefficients for a polycrystal with a fiber texture deserve more discussion. For polycrystals with a fiber texture along x_3 , the texture coefficients depend on the texture in the x_1 - x_2 plane. Given the

orientation distribution in the x_1 - x_2 plane, we compute the texture coefficients according to Eq. (16). For this purpose let us assume an arbitrary distribution function $f(\phi)$ in x_1 - x_2 plane, where ϕ is the angle between X_1 of the local crystal system and x_1 of global sample system. The ODF can then be written as:

$$w(\xi, \varphi, \phi) = \delta(\xi - 1)f(\phi), \quad (20)$$

where $\delta(\xi - 1)$ is the Dirac delta function used to denote the perfect alignment of the grains. The only restrictions on $f(\phi)$ is that it is normalized and must satisfy the sample and crystal symmetry elements.¹⁴ Substituting Eq. (20) into Eq. (16), we obtain the following nonzero texture coefficients:

$$W_{100} = \frac{\sqrt{3/2}}{4\pi^2}, \quad W_{200} = \frac{\sqrt{5/2}}{4\pi^2},$$

$$W_{300} = \frac{\sqrt{7/2}}{4\pi^2}, \quad W_{400} = \frac{\sqrt{9/2}}{4\pi^2},$$

$$W_{222} = W_{2\bar{2}\bar{2}} = \frac{\sqrt{5/2}}{4\pi^2} \int_0^{2\pi} f(\phi)e^{i2\phi}d\phi,$$

$$W_{322} = W_{3\bar{2}\bar{2}} = \frac{\sqrt{7/2}}{4\pi^2} \int_0^{2\pi} f(\phi)e^{i2\phi}d\phi,$$

$$W_{422} = W_{4\bar{2}\bar{2}} = \frac{\sqrt{9/2}}{4\pi^2} \int_0^{2\pi} f(\phi)e^{i2\phi}d\phi,$$

$$W_{444} = W_{4\bar{4}\bar{4}} = \frac{\sqrt{9/2}}{4\pi^2} \int_0^{2\pi} f(\phi)e^{i4\phi}d\phi. \quad (21)$$

Interestingly, W_{100} , W_{200} , W_{300} , and W_{400} are independent of the texture in the x_1 - x_2 plane. Equations (21) can be used with the results in Table IV to compute the electroelastic moduli. For 2 mm crystals where there is only a twofold symmetry axis, there are no exact solutions available according to Table III. This can also be verified by Eq. (21) and Table IV, since the Voigt–Reuss upper and lower bounds do not coincide. For a polycrystal composed of 4 mm tetragonal crystals so that there is a fourfold symmetry axis, all of the thermoelectroelastic moduli are exact, except S_{11} and S_{66} , according to Table III. This is also evident from the results of the Voigt–Reuss averages summarized in Table IV and the texture coefficients in Eq. (21). The upper bound on \mathbf{P} was calculated directly using the Voigt averages. The lower bound on \mathbf{P} was calculated by inverting \mathbf{P} to obtain \mathbf{Q} as a function of \mathbf{P} , computing the Reuss average of \mathbf{Q} , and finally inverting these results to obtain the lower bounds. We verified that all of the bounds do indeed coincide and recover the single crystal values, except for S_{11} and S_{66} , regardless of the texture in x_1 - x_2 plane, i.e., the value of W_{222} , W_{322} , W_{422} , and W_{444} . When the constituent crystals exhibit 6 mm symmetry, the upper and lower bounds for S_{11} and S_{66} also coincide, regardless of the texture in the x_1 - x_2 plane.

TABLE IV. (a) Elastic moduli of an orthorhombic polycrystal with a fiber texture in terms of single crystal (2 mm) moduli and the texture coefficients.

Elastic moduli
$\langle S_{11} \rangle = S_{11}^0 + hW_{200} + kW_{222} + 3iW_{400} + mW_{422} + nW_{444},$ $\langle S_{12} \rangle = S_{12}^0 + 2jW_{200} + iW_{400} - nW_{444},$ $\langle S_{13} \rangle = S_{12}^0 - jW_{200} + lW_{222} - 4iW_{400} - mW_{422},$ $\langle S_{22} \rangle = S_{11}^0 + hW_{200} - kW_{222} + 3iW_{400} - mW_{422} + nW_{444},$ $\langle S_{23} \rangle = S_{12}^0 - jW_{200} - lW_{222} - 4iW_{400} + mW_{422},$ $\langle S_{33} \rangle = S_{11}^0 - 2hW_{200} + 8iW_{400},$ $\langle S_{44} \rangle = 2(S_{11}^0 - S_{12}^0) - (a - 2c)W_{200} + (2e - d)W_{222} - 16bW_{400} + 4fW_{422},$ $\langle S_{55} \rangle = 2(S_{11}^0 - S_{12}^0) - (a - 2c)W_{200} - (2e - d)W_{222} - 16bW_{400} - 4fW_{422},$ $\langle S_{66} \rangle = 2(S_{11}^0 - S_{12}^0) - (a - 2c)W_{200} + 4bW_{400} - 4gW_{444},$ $\langle C_{11} \rangle = C_{11}^0 + aW_{200} + dW_{222} + 3bW_{400} + fW_{422} + gW_{444},$ $\langle C_{12} \rangle = C_{12}^0 + 2cW_{200} + bW_{400} - gW_{444},$ $\langle C_{13} \rangle = C_{12}^0 - cW_{200} + eW_{222} - 4bW_{400} - fW_{422},$ $\langle C_{22} \rangle = C_{11}^0 + aW_{200} - dW_{222} + 3bW_{400} - fW_{422} + gW_{444},$ $\langle C_{23} \rangle = C_{12}^0 - cW_{200} - eW_{222} - 4bW_{400} + fW_{422},$ $\langle C_{33} \rangle = C_{11}^0 - 2aW_{200} + 8bW_{400},$ $\langle C_{44} \rangle = \frac{C_{11}^0 - C_{12}^0}{2} - \frac{a - 2c}{4}W_{200} + \frac{2e - d}{4}W_{222} - 4bW_{400} + fW_{422},$ $\langle C_{35} \rangle = \frac{C_{11}^0 - C_{12}^0}{2} - \frac{a - 2c}{4}W_{200} - \frac{2e - d}{4}W_{222} - 4bW_{400} - fW_{422},$ $\langle C_{66} \rangle = \frac{C_{11}^0 - C_{12}^0}{2} - \frac{a - 2c}{4}W_{200} + bW_{400} - gW_{444},$

where

$$S_{11}^0 = (3S_{11} + 2S_{12} + 2S_{13} + 3S_{22} + 2S_{23} + 3S_{33} + S_{44} + S_{55} + S_{66})/15,$$

$$S_{12}^0 = (2S_{11} + 8S_{12} + 8S_{13} + 2S_{22} + 8S_{23} + 2S_{33} - S_{44} - S_{55} - S_{66})/30,$$

$$h = 2\sqrt{10}\pi^2(6S_{11} + 4S_{12} - 2S_{13} + 6S_{22} - 2S_{23} - 12S_{33} - S_{44} - S_{55} + 8S_{66})/105,$$

$$i = \sqrt{2}\pi^2(3S_{11} + 2S_{12} - 8S_{13} + 3S_{22} - 8S_{23} + 8S_{33} - 4S_{44} - 4S_{55} + S_{66})/210,$$

$$j = 2\sqrt{10}\pi^2(S_{11} + 10S_{12} - 5S_{13} + S_{22} - 2S_{33} + S_{44} + S_{55} - 2S_{66})/105,$$

$$k = 2\sqrt{10}\pi^2(6C_{11} + 2C_{13} - 6C_{22} - 2C_{23} - S_{44} + S_{55})/35,$$

$$l = 2\sqrt{10}\pi^2(S_{11} + S_{13} - S_{22} - S_{23} + S_{44} - S_{55})/35,$$

$$m = 2\sqrt{2}\pi^2(S_{11} - 2S_{13} - S_{22} + 2S_{33} + S_{44} - S_{55})/21,$$

$$n = \sqrt{2}\pi^2(S_{11} - 2S_{12} + S_{22} - S_{66})/6,$$

$$C_{11}^0 = (3C_{11} + 2C_{12} + 2C_{13} + 3C_{22} + 2C_{23} + 3C_{33} + 4C_{44} + 4C_{55} + 4C_{66})/15,$$

$$C_{12}^0 = (C_{11} + 4C_{12} + 4C_{13} + C_{22} + 4C_{23} + C_{33} - 2C_{44} - 2C_{55} - 2C_{66})/15,$$

$$a = 4\sqrt{10}\pi^2(3C_{11} + 2C_{12} - C_{13} + 3C_{22} - C_{23} - 6C_{33} - 2C_{44} - 2C_{55} + 4C_{66})/105,$$

$$b = \sqrt{2}\pi^2(3C_{11} + 2C_{12} - 8C_{13} + 3C_{22} - 8C_{23} + 8C_{33} - 16C_{44} - 16C_{55} + 4C_{66})/210,$$

$$c = 2\sqrt{10}\pi^2(C_{11} + 10C_{12} - 5C_{13} + C_{22} - 2C_{33} + 4C_{44} + 4C_{55} - 8C_{66})/105,$$

$$d = 4\sqrt{10}\pi^2(3C_{11} + C_{13} - 3C_{22} - C_{23} - 2C_{44} + 2C_{55})/35,$$

$$e = 2\sqrt{10}\pi^2(C_{11} + C_{13} - C_{22} - C_{23} + 4C_{44} - 4C_{55})/35,$$

$$f = 2\sqrt{2}\pi^2(C_{11} - 2C_{13} - C_{22} + 2C_{23} + 4C_{44} - 4C_{55})/21,$$

$$g = \sqrt{2}\pi^2(C_{11} - 2C_{12} + C_{22} - 4C_{66})/6.$$

(b) Piezoelectric moduli for an orthorhombic polycrystal with a fiber texture in terms of single crystal (2 mm) moduli and the texture coefficients.

Piezoelectric moduli
$\langle d_{15} \rangle = \frac{4\sqrt{6}\pi^2}{15}uW_{100} + \frac{4\sqrt{14}\pi^2}{35}vW_{300} + \frac{4\sqrt{14}\pi^2}{21}wW_{322},$ $\langle d_{24} \rangle = \frac{4\sqrt{6}\pi^2}{15}uW_{200} + \frac{4\sqrt{14}\pi^2}{35}vW_{300} - \frac{4\sqrt{14}\pi^2}{21}wW_{322},$ $\langle d_{31} \rangle = \frac{4\sqrt{6}\pi^2}{15}xW_{100} + \frac{2\sqrt{14}\pi^2}{35}vW_{300} + \frac{2\sqrt{14}\pi^2}{21}wW_{322},$ $\langle d_{33} \rangle = \frac{4\sqrt{6}\pi^2}{15}xW_{100} + \frac{2\sqrt{14}\pi^2}{35}vW_{300} - \frac{2\sqrt{14}\pi^2}{21}wW_{322},$ $\langle d_{33} \rangle = \frac{4\sqrt{6}\pi^2}{15}yW_{100} - \frac{4\sqrt{14}\pi^2}{35}vW_{300},$ $\langle h_{15} \rangle = \frac{2\sqrt{6}\pi^2}{15}pW_{100} + \frac{2\sqrt{14}\pi^2}{35}qW_{300} + \frac{2\sqrt{14}\pi^2}{21}rW_{322},$ $\langle h_{24} \rangle = \frac{2\sqrt{6}\pi^2}{15}pW_{100} + \frac{2\sqrt{14}\pi^2}{35}qW_{300} - \frac{2\sqrt{14}\pi^2}{21}rW_{322},$

TABLE IV. (Continued)

$$\begin{aligned} \langle h_{31} \rangle &= \frac{4\sqrt{6}\pi^2}{15} sW_{100} + \frac{2\sqrt{14}\pi^2}{35} qW_{300} + \frac{2\sqrt{14}\pi^2}{21} rW_{322}, \\ \langle h_{33} \rangle &= \frac{4\sqrt{6}\pi^2}{15} sW_{100} + \frac{2\sqrt{14}\pi^2}{35} qW_{300} - \frac{2\sqrt{14}\pi^2}{21} rW_{322}, \\ (h_{33}) &= \frac{4\sqrt{6}\pi^2}{15} tW_{100} - \frac{4\sqrt{14}\pi^2}{35} qW_{300}, \end{aligned}$$

where

$$\begin{aligned} u &= (3d_{15} + 3d_{24} - 2d_{31} - 2d_{32} + 4d_{33})/2, \\ v &= d_{15} + d_{24} + d_{31} + d_{32} - 2d_{33}, \\ w &= d_{15} - d_{24} + d_{31} - d_{32}, \\ x &= (-d_{15} - d_{24} + 4d_{31} + 4d_{32} + 2d_{33})/2, \\ y &= d_{15} + d_{24} + d_{31} + d_{32} + 3d_{33}, \\ p &= 3h_{15} + 3h_{24} - h_{31} - h_{32} + 2h_{33}, \\ q &= 2h_{15} + 2h_{24} + h_{31} + h_{32} - 2h_{33}, \\ r &= 2h_{15} - 2h_{24} + h_{31} - h_{32}, \\ s &= -h_{15} - h_{24} + 2h_{31} + 2h_{32} + h_{33}, \\ t &= 2h_{15} + 2h_{24} + h_{31} + h_{32} + 3h_{33}. \end{aligned}$$

(c) Dielectric moduli for an orthorhombic polycrystal with a fiber texture in terms of single crystal moduli (2 mm) and the texture coefficients.

Dielectric moduli

$$\begin{aligned} \langle \tau_{11} \rangle &= \tau^0 + \frac{2\sqrt{10}\pi^2}{15} \chi W_{200} + \frac{2\sqrt{10}\pi^2}{5} (\tau_{11} - \tau_{22}) W_{222}, \\ \langle \tau_{22} \rangle &= \tau^0 + \frac{2\sqrt{10}\pi^2}{15} \chi W_{200} - \frac{2\sqrt{10}\pi^2}{5} (\tau_{11} - \tau_{22}) W_{222}, \\ \langle \tau_{33} \rangle &= \tau^0 - \frac{4\sqrt{10}\pi^2}{15} \chi W_{200} \\ \langle \beta_{11} \rangle &= \beta^0 + \frac{2\sqrt{10}\pi^2}{15} \omega W_{200} + \frac{2\sqrt{10}\pi^2}{5} (\beta_{11} - \beta_{22}) W_{222}, \\ \langle \beta_{22} \rangle &= \beta^0 + \frac{2\sqrt{10}\pi^2}{15} \omega W_{200} - \frac{2\sqrt{10}\pi^2}{5} (\beta_{11} - \beta_{22}) W_{222}, \\ \langle \beta_{33} \rangle &= \beta^0 - \frac{4\sqrt{10}\pi^2}{15} \omega W_{200}, \end{aligned}$$

where

$$\begin{aligned} \tau^0 &= \frac{\tau_{11} + \tau_{22} + \tau_{33}}{3}, \\ \chi &= \frac{\tau_{11} + \tau_{22} - 2\tau_{33}}{3}, \\ \beta^0 &= \frac{\beta_{11} + \beta_{22} + \beta_{33}}{3}, \\ \omega &= \frac{\beta_{11} + \beta_{22} - 2\beta_{33}}{3}. \end{aligned}$$

To further demonstrate the results, consider a transversely isotropic polycrystal composed of 4 mm single crystals. In this case, $W_{222} = W_{322} = W_{422} = W_{444} = 0$. The bounds on S_{11} and S_{66} can be expressed as:

$$\begin{aligned} \frac{2(S_{11}^2 - S_{12}^2) + (3S_{11} - S_{12})S_{66}}{2(2S_{11} - 2S_{12} + S_{66})} \\ \leq \langle S_{11} \rangle \leq \frac{6S_{11} + 2S_{12} + S_{66}}{8}, \end{aligned} \tag{22}$$

$$\frac{4(S_{11} - S_{12})S_{66}}{2S_{11} - 2S_{12} + S_{66}} \leq \langle S_{66} \rangle \leq S_{11} - S_{12} + \frac{S_{66}}{2}. \tag{23}$$

One can readily verify that the upper and lower bounds coincide when $2(S_{11} - S_{12}) = S_{66}$, which reduces the fourfold symmetry to sixfold symmetry, again consistent with the results of Table III.

Finally, we show some numerical results for piezoelectric BaTiO₃ polycrystals having film textures. The electroelastic moduli of both materials and the corresponding Voigt–Reuss averages are listed in Table V, where the single crystal values of BaTiO₃ are obtained from Berlincourt and Jaffe.¹⁶ Only those effective properties without exact solutions are listed, i.e., S_{11} and S_{66} . For those not tabulated, the exact solutions recover the single crystal values. For the

TABLE V. Electroelastic moduli of BaTiO₃ single crystals and polycrystals with film textures. Units of S_{ij} are Pa⁻¹², of d_{ij} are 10⁻¹² m/V, and of τ_{ij} are 10⁻⁹ C²Nm².

S_{11}	S_{12}	S_{13}	S_{33}	S_{44}	S_{66}
8.05	-2.34	-5.25	15.72	18.42	8.84
d_{31}	d_{33}	d_{15}	τ_{11}	τ_{33}	
-34.50	85.60	392.27	25.79	1.46	
S_{11}^U	S_{11}^L	S_{66}^U	S_{66}^L		
6.56	5.95	14.84	12.41		

components tabulated in Table V the bounds are quite tight and both the upper and lower bounds would seem to be reasonable estimates of the polycrystal property.

V. CONCLUSIONS

We have determined the conditions that permit exact solutions for some thermoelectroelastic moduli of piezoelectric polycrystals with fiber texture. This was accomplished by the establishment of uniform electroelastic fields in polycrystals

with fiber texture. More general expressions for bounds and estimates on polycrystal properties in terms of single crystal moduli and the appropriate texture coefficients were also developed. These were used to verify the exact solutions.

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