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Stress formulation of acoustoelasticity

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ABSTRACT

Acoustoelasticity describes the relationship between elastic wave velocities and the initial stress present in a material. Traditional theories consider successive deformations involving small amplitude wave motion superimposed on an initially deformed material. Then, a constitutive relationship must be applied to relate the initial static deformation to the desired relationships involving initial stress, resulting in expressions of wave velocities involving a mix of elastic stiffness and compliance constants. In this article, a pure stress formulation is developed for acoustoelasticity. In this setting, the problem involves the superposition of a dynamic stress wave on an initially stressed material configuration, rather than the superposition of kinematic variables. The phase velocity of the stress wave is naturally related to the initial stress through only the compliance constants. Thus, compliances are the fundamental constants of acoustoelasticity.

1. Introduction

Description of elastic wave motion in solid continua is usually considered using the displacement equations of motion, i.e., Navier's equations [1]. While displacement forms are commonplace, it is possible to formulate so-called stress equations of motion with stress being the dependent variable [2]. A pure stress formalism was pioneered by Ignaczak beginning in the 1950s [2] followed by a proof of the necessary and sufficient conditions on the stress tensor appropriate for elastodynamics [3]. However, modeling of elastic wave applications, in ultrasonics for example, are still largely based on displacement equations [4–8]. More recently, Ostoja-Starzewski reinvigorated exploring elastic waves from the stress equations of motion and gave several examples and applications [9]. One example included the scarce use of stress equations of motion for nonlinear elastodynamics for which the work of Bustamante and Sfyris [10] was highlighted [9].

In our previous work, the pure stress equations of motion were derived and solved for a homogeneous anisotropic solid [11]. Solutions arose from the consideration of an eigenvalue problem, which governs the phase velocity of the stress wave and the tensorial stress components (eigenvectors). Both the phase velocities and stress components were found to depend on elastic compliance constants rather than elastic stiffnesses [11]. It was demonstrated that these solutions were consistent with the displacement formalism and the stress eigenvalue problem is the dual formalism to the well-known Christoffel equations [11]. While the previous work demonstrated that displacement and stress formalisms were consistent, possible benefits of proceeding with the stress formalism were not explicitly given.

In this paper, the stress formalism is applied to acoustoelasticity [12], which describes how the velocities of an elastic material are influenced by internal stresses. Acoustoelasticity is an inherent nonlinear effect in which the dynamic

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deformation of the wave couples with the initial deformation in the solid. Thus, the present paper provides an additional development [10] of the stress formalism to nonlinear elastodynamics. The topic of acoustoelasticity has garnered significant interest over the years in its application to the nondestructive measurement of stress in addition to measuring third-order elastic constants when the stress is intentionally applied and known [13–19]. In both of these measurement applications the stress is the primary independent variable. Here, we show that the stress formulation provides a natural treatment of acoustoelasticity because it focuses on the applied stress as the input to the velocity change. We also show that the optimal third order constants are those of compliance, the inverse of stiffness. Readers interested in the background and historical accounts of acoustoelasticity are encouraged to consult the references of Pao, Sachse, and Fukuoka [13], Kim and Sachse [14], Thurston [15], Norris [16], Man and Lu [17], Ogden [18], and Shams, Destrade, and Ogden [19].

To provide some additional context and introduction to the present work, we recall the theory as given by Pao and Gamer [20], and also borrow solutions from Thurston and Brugger [21]. The equation of motion or wave equation for an elastic continuum containing a static stress follows from Cauchy's first law of motion (Eq. (12) of Pao and Gamer [20])

$$\frac{\partial}{\partial a_l}\left(\sigma^i_{kl}\frac{\partial u_i}{\partial a_k}+\sigma_{kl}\frac{\partial u^i_i}{\partial a_k}+\sigma_{il}\right)=\rho_0\frac{\partial^2 u_i}{\partial t^2}. \tag{1}$$

The superscript i follows the notation of Pao and Gamer [20] and refers to quantities present at the initial state of the material, e.g., the initial stress is denoted as $\boldsymbol{\sigma}^i$ and the initial displacement as $\mathbf{u}^i$. Here, the superscript i should not be confused with the index notation seen in the subscripts describing tensor components. The quantities $\boldsymbol{\sigma}$ and $\mathbf{u}$ are the (incremental) dynamic stress and displacement of the elastic wave as a function of the natural coordinates $\mathbf{a}$, respectively, with notation fully defined in Section 2. A natural first step in solving any partial differential equation with mixed variables like Eq. (1) is to cast it in terms of a single dependent variable, usually the displacement $\mathbf{u}$. In doing so, material dependent constitutive behavior must be applied to relate $\boldsymbol{\sigma}$ to $\mathbf{u}$. For Eq. (1) specifically, it is also desirable to have a single independent variable, usually the stress $\boldsymbol{\sigma}^i$. Thus, constitutive relationships are also needed to relate $\mathbf{u}^i$ to $\boldsymbol{\sigma}^i$. Applying constitutive relations in the traditional displacement formulation leads to phase velocities that are complicated expressions containing a mix of fourth- and sixth-rank tensors of elastic stiffnesses (c_{ijkl}, c_{ijklmn}) and compliances (s_{ijkl}). For example, the bulk wave phase velocity solution to Eq. (1) for an anisotropic material with an externally generated stress $\boldsymbol{\sigma}^i$ is [21]

$$\rho_0 c^2=\rho_0 c_0^2+\left(\hat{n}_p\hat{n}_q+2\rho_0 c_0^2\hat{u}_i\hat{u}_j s_{ijpq}+\hat{u}_i\hat{n}_j\hat{u}_k\hat{n}_l c_{ijklmn}s_{mnpq}\right)\sigma^i_{pq}, \tag{2}$$

where ρ_0, c_0, $\hat{\mathbf{n}}$, and $\hat{\mathbf{u}}$ are the density, phase velocity at zero stress, propagation direction, and displacement direction, respectively. As an example, for a longitudinal wave propagating in an isotropic material containing a uniaxial stress σ^i_{33}, Eq. (2) reduces to [21]

$$\rho_0 c^2=\rho_0 c_0^2+\left[s_{12}(2\rho_0 c_0^2+3c_{123}+10c_{144}+8c_{456})+2s_{44}(c_{123}+2c_{144})\right]\sigma^i_{33}. \tag{3}$$

While Eq. (3) is relatively simple, evaluation requires the knowledge of both compliance and stiffness constants or at least the ability to convert between them.

In the present work, we demonstrate that casting Eq. (1) to include only $\boldsymbol{\sigma}$ as the dependent variable results in solutions that depend only on compliance constants. This proves that compliance constants are the fundamental material parameters in acoustoelasticity, at least when acoustoelasticity is defined as the relationship between the phase velocity c of an elastic wave and stress $\boldsymbol{\sigma}^i$. Alternatively, the dual problem of considering the relationship between the phase velocity c of an elastic wave and strain $\boldsymbol{\epsilon}^i$ would depend on only stiffnesses. Consistency between the formulations is easily observed through application of stiffness–compliance relationships, which are well-known for fourth-rank tensors $c_{ijmn}s_{mnkl}=I_{ijkl}$, but relations between c_{ijklmn} and s_{ijklmn} are more obscure. This obscurity likely prevented researchers from noticing that expressions, like Eq. (3), could be written in terms of compliance constants only, which is a primary result of the current article.

This article is organized as follows. The general formalism of acoustoelasticity using stress as the dependent variable is given in Section 2. Plane wave propagation is considered in Section 3 using a perturbation expansion to find the dependence of phase velocity of the elastic wave on the initial stress. Consistency between the present stress formulation and the traditional displacement formalism is established in Section 4 using a newly proposed set of constants that simplify conversion between third order stiffness and compliance.

2. Stress formulation of acoustoelasticity

The notation follows closely to that of Pao and Gamer [13,20]. Consider three material configurations: a natural one having coordinates $\mathbf{a}$, an initial one having coordinates $\mathbf{X}$, and a current one having coordinates $\mathbf{x}$. The displacement $\mathbf{u}^i=\mathbf{X}-\mathbf{a}$ is the displacement associated with a static and homogeneous deformation. The displacement from the initial to the current configuration is that of the wave $\mathbf{u}=\mathbf{x}-\mathbf{X}$, which is dynamic and spatially-dependent, and the full, or final displacement is $\mathbf{u}^f=\mathbf{u}^i+\mathbf{u}=\mathbf{x}-\mathbf{a}$ [20]. The second Piola–Kirchhoff stress associated with the initial deformation is $\boldsymbol{\sigma}^i$, the dynamic deformation is $\boldsymbol{\sigma}(\mathbf{a},t)$, and, thus, $\boldsymbol{\sigma}^f=\boldsymbol{\sigma}^i+\boldsymbol{\sigma}$. Cauchy's law governs the wave motion and is given in

Eq. (1) [see also Eq. (12) of Ref. [20]]. To begin the process of casting Eq. (1) to be in terms of $\boldsymbol{\sigma}$ and $\boldsymbol{\sigma}^i$, we differentiate Eq. (1) with respect to the coordinate $\mathbf{a}$ to give

$$\frac{\partial^2}{\partial a_j \partial a_l}\left(\sigma^i_{kl}\frac{\partial u_i}{\partial a_k} + \sigma_{kl}\frac{\partial u^i_i}{\partial a_k} + \sigma_{il}\right) = \rho_0 \frac{\partial \ddot{u}_i}{\partial a_j}, \tag{4}$$

where the standard dot notation is employed for time differentiation. The Lagrangian strain associated with the superposition of the deformations is

$$E^f_{ij} = \frac{1}{2}\left(\frac{\partial u^f_i}{\partial a_j} + \frac{\partial u^f_j}{\partial a_i} + \frac{\partial u^f_k}{\partial a_i}\frac{\partial u^f_k}{\partial a_j}\right). \tag{5}$$

Differentiating Eq. (5) twice with respect to time gives

$$\ddot{E}^f_{ij} = \frac{1}{2}\left(\frac{\partial \ddot{u}_i}{\partial a_j} + \frac{\partial \ddot{u}_j}{\partial a_i} + \frac{\partial u^i_k}{\partial a_i}\frac{\partial \ddot{u}_k}{\partial a_j} + \frac{\partial \ddot{u}_k}{\partial a_i}\frac{\partial u^i_k}{\partial a_j}\right), \tag{6}$$

where the relationship $\dot{\mathbf{u}}^f = \dot{\mathbf{u}}^i + \dot{\mathbf{u}} = \dot{\mathbf{u}}$ is applied as the initial displacement is assumed to be static and time-independent. Additionally, dynamic displacements of order two and higher are neglected. Clearly, the right-hand side of Eq. (4) is related to and contained in Eq. (6). Now, the strain and its time-derivatives in terms of stress can be obtained through the constitutive relation,

$$\begin{aligned} E^f_{ij} &= s_{ijkl}\sigma^f_{kl} + \frac{1}{2}s_{ijklmn}\sigma^f_{kl}\sigma^f_{mn} \\ &\approx s_{ijkl}\left(\sigma^i_{kl} + \sigma_{kl}\right) + s_{ijklmn}\sigma^i_{kl}\sigma_{mn}, \end{aligned} \tag{7}$$

where s_{ijkl} and s_{ijklmn} are compliance tensors and we used $\boldsymbol{\sigma}^f = \boldsymbol{\sigma}^i + \boldsymbol{\sigma}$ to arrive at the second expression. Eq. (7) is a constitutive relationship between the Green–Lagrange strain $\mathbf{E}^f$ and the second Piola–Kirchhoff stress $\boldsymbol{\sigma}^f$ including the effects of the initial stress $\boldsymbol{\sigma}^i$. A dual constitutive relationship between stress and strain was derived by Pao and Gamer [20] in their Eq. (21). Theories of large acoustoelasticity have been obtained through constitutive relationships expanded to third-order [22,23]. Eq. (7) could also be expanded to third-order in stress by including the fourth-order compliances $s_{ijklmnpq}$, but is beyond the scope of the current article. Two time-derivatives of Eq. (7) lead to

$$\ddot{E}^f_{ij} = s_{ijkl}\ddot{\sigma}_{kl} + s_{ijklmn}\sigma^i_{kl}\ddot{\sigma}_{mn}, \tag{8}$$

which combined with Eq. (6) gives

$$s_{ijkl}\ddot{\sigma}_{kl} + s_{ijklmn}\sigma^i_{kl}\ddot{\sigma}_{mn} = \frac{1}{2}\left(\frac{\partial \ddot{u}_i}{\partial a_j} + \frac{\partial \ddot{u}_j}{\partial a_i} + \frac{\partial u^i_k}{\partial a_i}\frac{\partial \ddot{u}_k}{\partial a_j} + \frac{\partial \ddot{u}_k}{\partial a_i}\frac{\partial u^i_k}{\partial a_j}\right). \tag{9}$$

Now, the displacements in the right-hand side of Eq. (9) can be eliminated using Eq. (4). After simplification, the stress equation of motion is obtained,

$$\begin{aligned} \rho_0\left(s_{ijkl} + s_{ijklmn}\sigma^i_{mn}\right)\ddot{\sigma}_{kl} =& \frac{1}{2}\left(\frac{\partial^2\sigma_{ik}}{\partial a_j\partial a_k} + \frac{\partial^2\sigma_{jk}}{\partial a_i\partial a_k}\right) \\ &+ \left(s_{ijkl}\frac{\partial^2\sigma_{kl}}{\partial a_m\partial a_n} + s_{ikmn}\frac{\partial^2\sigma_{kl}}{\partial a_j\partial a_l} + s_{jkmn}\frac{\partial^2\sigma_{kl}}{\partial a_i\partial a_l}\right)\sigma^i_{mn}, \end{aligned} \tag{10}$$

where terms involving two or more spatial derivatives on the initial displacement are zero because of the homogeneity of the initial stress and the linearized strain-stress relationship was used. In the absence of an initial stress $\boldsymbol{\sigma}^i$, the traditional stress equation is readily observed [11]. Eq. (10) can be cast into the equivalent form

$$\rho_0\left(s_{ijkl} + s_{ijklmn}\sigma^i_{mn}\right)\ddot{\sigma}_{kl} = \left[\delta_{lq}I_{ijkp} + \left(\delta_{jp}\delta_{lq}s_{ikmn} + \delta_{ip}\delta_{lq}s_{jkmn} + \delta_{mp}\delta_{nq}s_{ijkl}\right)\sigma^i_{mn}\right]\frac{\partial^2\sigma_{kl}}{\partial a_p\partial a_q}, \tag{11}$$

of which the factoring is made possible through the use of the Kronecker delta function. It is noted that while strain-stress constitutive relationships were used to arrive at Eqs. (10) and (11), elastic stiffness constants never entered the picture. Thus, acoustoelasticity is fundamentally connected to second- and third-order compliance constants (s_{ijkl} and s_{ijklmn}, respectively).

Now, homogeneity of the stress field and material properties permits the assumption that wave motion is a function of a single spatial coordinate $x = \hat{\mathbf{n}} \cdot \mathbf{a}$ so that $\boldsymbol{\sigma} = \boldsymbol{\sigma}(x, t)$. Then, Eq. (11) reduces to a uni-dimensional stress wave equation

$$\rho_0\left(s_{ijkl} + s_{ijklmn}\sigma^i_{mn}\right)\ddot{\sigma}_{kl} = \left(N_{ijkl} + N_{ijklmn}\sigma^i_{mn}\right)\frac{\partial^2\sigma_{kl}}{\partial x^2}, \tag{12}$$

and

$$N_{ijkl} = \frac{1}{4}\left(\delta_{ik}\hat{n}_j\hat{n}_l + \delta_{jk}\hat{n}_i\hat{n}_l + \delta_{il}\hat{n}_j\hat{n}_k + \delta_{jl}\hat{n}_i\hat{n}_k\right), \tag{13a}$$

$$N_{ijklmn} = \frac{1}{2}\left(\delta_{ip}\hat{n}_j + \delta_{jp}\hat{n}_i\right)\left(\delta_{kq}\hat{n}_l + \delta_{lq}\hat{n}_k\right)s_{pqmn} + s_{ijkl}\hat{n}_m\hat{n}_n. \tag{13b}$$

The resulting expression in Eq. (12) is a wave equation with the dynamic stress wave $\boldsymbol{\sigma}(x,t)$ as the dependent variable, which couples with the initial stress $\boldsymbol{\sigma}^i$. Thus, Eq. (12) is a generalization of the stress equations of motion derived previously for stress-free anisotropic solids [11].

In closing this section, we offer a couple of remarks. Firstly, Eq. (12) governs the propagation of the stress $\boldsymbol{\sigma}(x,t)$ where x describes spatial coordinates along $\hat{\mathbf{n}}$ relative to the undeformed material configuration. Thus, both x and $\hat{\mathbf{n}}$ are not influenced by the deformation. Alternatively, a formulation relative to initial or deformed coordinates causes the propagation direction and associated coordinates to be influenced by loading. Thurston and Brugger [21] discuss the advantages of using the undeformed configuration when utilizing applied loads for measuring third-order elastic constants. Several references derive and report resulting formulas when utilizing the different coordinates [16,20,21,23–25]. In Refs. [24] and [25], the focus is on electroelastic materials containing a bias (via external static loading or electric field) for which the natural and initial coordinates describe points in the described reference and intermediate material configurations as a result of the bias. Tiersten [25] motivates the employment of natural coordinates by stating, "Since in the typical situation it is undesirable to measure the geometry each time the bias is varied, it is advantageous to use the X_L (natural coordinates) as independent variables". Further discussion was provided by Sinha [24].

Lastly, the present stress formalism incorporates the homogeneous initial stress $\boldsymbol{\sigma}^i$ more naturally than the traditional displacement formulations. In displacement based acoustoelastic models several steps are generally required to incorporate $\boldsymbol{\sigma}^i$ into the model. For example, a homogeneous irrotational deformation is assumed to allow displacement or deformation gradients to be transformed to strain (sometimes using polar decomposition). Then, the strain is written in terms of the homogeneous stress $\boldsymbol{\sigma}^i$ via a constitutive relationship [see Eqs. (82)–(86) of Huang et al. [26] as an example].

3. Plane wave propagation

Assume the stress wave $\boldsymbol{\sigma}(x,t)$ to be of harmonic form with wave number k, angular frequency ω, and amplitude described by the second rank tensor $\mathring{\boldsymbol{\sigma}}$. Then, Eq. (12) can be written as an algebraic system of equations

$$\left\{N_{ijkl} + N_{ijklmn}\sigma^i_{mn} - \lambda\left(s_{ijkl} + s_{ijklmn}\sigma^i_{mn}\right)\right\}\mathring{\sigma}_{kl} = 0, \tag{14}$$

where $\lambda = \rho_0 c^2$. Note that N_{ijkl}, $N_{ijklmn}\sigma^i_{mn}$ and $s_{ijklmn}\sigma^i_{mn}$ carry the same symmetry as the compliance s_{ijkl} in that components are unaltered under the interchange $i \leftrightarrow j$ or $k \leftrightarrow l$ or $ij \leftrightarrow kl$. Eq. (14) is the dual form to the traditional Christoffel equations derived from the displacement formulation,

$$\left\{\Gamma_{ijkl}\hat{n}_j\hat{n}_l - \lambda\delta_{ik}\right\}\hat{u}_k = 0, \tag{15}$$

which is reported in Eq. (36) in Pao and Gamer [20] where $\boldsymbol{\Gamma}$ is a tensor containing second- and third-order elastic stiffnesses. Note that we utilize the notation $\mathring{\boldsymbol{\sigma}}$ rather than $\hat{\boldsymbol{\sigma}}$ as the tensorial part of $\mathring{\boldsymbol{\sigma}}$ is not normalized in general. Further discussion of stress and displacement eigenvectors is provided in previous work [11].

3.1. Perturbation expansion

To help determine the acoustoelastic relations, a perturbative solution is assumed,

$$\lambda = \lambda^0 + \Delta\lambda, \tag{16a}$$

$$\mathring{\boldsymbol{\sigma}} = \mathring{\boldsymbol{\sigma}}^0 + \Delta\mathring{\boldsymbol{\sigma}}, \tag{16b}$$

where $\lambda^0 = \rho_0 c_0^2$ and $\mathring{\boldsymbol{\sigma}}^0$ represent the solutions in the absence of initial stress, and the additional terms are linear in the initial stress $\boldsymbol{\sigma}^i$. A standard perturbation expansion of Eq. (14) yields

$$\left\{N_{ijkl} - \lambda^0 s_{ijkl}\right\}\mathring{\sigma}^0_{kl} = 0, \tag{17a}$$

$$\left\{N_{ijkl} - \lambda^0 s_{ijkl}\right\}\Delta\mathring{\sigma}_{kl} + \left\{\left(N_{ijklmn} - \lambda^0 s_{ijklmn}\right)\sigma^i_{mn} - \Delta\lambda s_{ijkl}\right\}\mathring{\sigma}^0_{kl} = 0. \tag{17b}$$

Multiplying Eq. (17b) by $\mathring{\sigma}^0_{ij}$ and using Eq. (14) gives

$$\begin{aligned}\Delta\lambda &= \frac{\left(N_{ijklmn} - \lambda^0 s_{ijklmn}\right)\mathring{\sigma}^0_{ij}\mathring{\sigma}^0_{kl}\sigma^i_{mn}}{s_{pqrs}\mathring{\sigma}^0_{pq}\mathring{\sigma}^0_{rs}} \\ &= \hat{n}_i\sigma^i_{ij}\hat{n}_j + \frac{\left(2\hat{n}_i\hat{n}_k s_{jlmn} - \lambda^0 s_{ijklmn}\right)\mathring{\sigma}^0_{ij}\mathring{\sigma}^0_{kl}\sigma^i_{mn}}{s_{pqrs}\mathring{\sigma}^0_{pq}\mathring{\sigma}^0_{rs}} \\ &= \hat{n}_i\sigma^i_{ij}\hat{n}_j + \frac{2}{\lambda^0}\mathring{\sigma}^0_{ik}\hat{n}_k\mathring{\sigma}^0_{jl}\hat{n}_l s_{ijmn}\sigma^i_{mn} - s_{ijklmn}\mathring{\sigma}^0_{ij}\mathring{\sigma}^0_{kl}\sigma^i_{mn}.\end{aligned} \tag{18}$$

Note that $s_{pqrs}\mathring{\sigma}^0_{pq}\mathring{\sigma}^0_{rs}$ is positive on account of the positive definite nature of the compliance.

Table 1
Terms found in $\Delta\lambda$ of Eq. (20) for longitudinal (L) and shear wave modes (T) where the first subscript $\perp$ or $||$ indicates the propagation direction being perpendicular or parallel to the loading direction for uniaxial stress cases, respectively. For shear waves, a second subscript ($\perp$ or $||$) indicates the displacement direction relative to the loading direction. No subscripts are needed for equal triaxial loading case. Note that $s = 3s_{12} + 2s_{44}$ and $\beta = 1/(s_{44}s)$. The following relationships can be used for converting the entries to stiffnesses: $s = 1/(3\kappa)$, $s_{44} = 1/(4c_{44})$, $\beta s = 4c_{44}$, and $\beta s_{44} = 3\kappa$ where c_{44} and κ are the shear and bulk moduli, respectively.

Mode	Loading	$\hat{\mathbf{n}}\cdot\boldsymbol{\sigma}^i\hat{\mathbf{n}}$	$\hat{\mathbf{n}}\cdot\mathring{\boldsymbol{\sigma}}\boldsymbol{\sigma}^i\mathring{\boldsymbol{\sigma}}\hat{\mathbf{n}}$	$\operatorname{tr}\mathring{\boldsymbol{\sigma}}$	$\operatorname{tr}(\mathring{\boldsymbol{\sigma}}\boldsymbol{\sigma}^i)$	$\operatorname{tr}\mathring{\boldsymbol{\sigma}}^2$	$\operatorname{tr}(\mathring{\boldsymbol{\sigma}}\boldsymbol{\sigma}^i\mathring{\boldsymbol{\sigma}})$
$\mathrm{L}_\perp$	Uniaxial	0	0	βs_{44}	$(2s_{44}-s)\,\beta\sigma^i/6$	$\left(2s_{44}^2+s^2\right)\beta^2/3$	$(2s_{44}-s)^2\,\beta^2\sigma^i/36$
$\mathrm{L}_{\|\|}$	Uniaxial	σ^i	σ^i	βs_{44}	$(s_{44}+s)\,\beta\sigma^i/3$	$\left(2s_{44}^2+s^2\right)\beta^2/3$	$(s_{44}+s)^2\,\beta^2\sigma^i/9$
$\mathrm{T}_{\perp\|\|}$	Uniaxial	0	σ^i	0	0	$(\beta s)^2/8$	$(\beta s)^2\,\sigma^i/16$
$\mathrm{T}_{\perp\perp}$	Uniaxial	0	0	0	0	$(\beta s)^2/8$	0
$\mathrm{T}_{\|\|\perp}$	Uniaxial	σ^i	0	0	0	$(\beta s)^2/8$	$(\beta s)^2\,\sigma^i/16$
L	Equal triaxial	σ^i	σ^i	βs_{44}	$\beta s_{44}\sigma^i$	$\left(2s_{44}^2+s^2\right)\beta^2/3$	$\left(2s_{44}^2+s^2\right)\beta^2\sigma^i/6$
T	Equal triaxial	σ^i	σ^i	0	0	$(\beta s)^2/8$	$(\beta s)^2\,\sigma^i/8$

Analysis [11] of the leading order equation in Eq. (17a) shows that it has six independent solutions, three of which have λ^0 positive non-zero corresponding to propagating wave modes. The other three solutions with $\lambda^0 = 0$ are associated with *incompatible* stresses and strains that are not physically permissible solutions. Therefore, the following will only consider the propagating wave modes.

3.2. Propagating stress wave solutions: $\lambda^0 > 0$

Define the traction vector of the leading order unstressed solution $\mathring{t}_i^0 = \mathring{\sigma}_{ij}^0\hat{n}_j$. The traction vectors of the propagating solutions are non-zero, which can be deduced from the displacement formulation in Section 4. Then, Eq. (18) can be rewritten as

$$\Delta\lambda = \hat{n}_k\hat{n}_l\sigma_{kl}^i + \left(2\mathring{t}_i^0\mathring{t}_j^0 s_{ijmn} - \lambda^0 s_{ijklmn}\mathring{\sigma}_{ij}^0\mathring{\sigma}_{kl}^0\right)\frac{\lambda^0\sigma_{mn}^i}{\mathring{\mathbf{t}}^0\cdot\mathring{\mathbf{t}}^0}. \tag{19}$$

For an isotropic solid, Eq. (19) reduces to

$$\begin{aligned}\Delta\lambda = {} & \hat{\mathbf{n}}\cdot\boldsymbol{\sigma}^i\hat{\mathbf{n}} + 2\lambda^0\left(s_{12}\operatorname{tr}\boldsymbol{\sigma}^i + 2s_{44}\hat{\mathbf{n}}\cdot\mathring{\boldsymbol{\sigma}}\boldsymbol{\sigma}^i\mathring{\boldsymbol{\sigma}}\hat{\mathbf{n}}\right) - \frac{s_1}{27}\left(\operatorname{tr}\mathring{\boldsymbol{\sigma}}\right)^2\operatorname{tr}\boldsymbol{\sigma}^i \\ & - \frac{s_2}{3}\left[2\operatorname{tr}(\mathring{\boldsymbol{\sigma}}\boldsymbol{\sigma}^i)\operatorname{tr}\mathring{\boldsymbol{\sigma}} - (\operatorname{tr}\mathring{\boldsymbol{\sigma}})^2\operatorname{tr}\boldsymbol{\sigma}^i + \operatorname{tr}\mathring{\boldsymbol{\sigma}}^2\operatorname{tr}\boldsymbol{\sigma}^i\right] \\ & - \frac{s_3}{9}\left[9\operatorname{tr}(\mathring{\boldsymbol{\sigma}}\boldsymbol{\sigma}^i\mathring{\boldsymbol{\sigma}}) - 3\operatorname{tr}\mathring{\boldsymbol{\sigma}}^2\operatorname{tr}\boldsymbol{\sigma}^i - 6\operatorname{tr}(\mathring{\boldsymbol{\sigma}}\boldsymbol{\sigma}^i)\operatorname{tr}\mathring{\boldsymbol{\sigma}} + 2(\operatorname{tr}\mathring{\boldsymbol{\sigma}})^2\operatorname{tr}\boldsymbol{\sigma}^i\right]\end{aligned} \tag{20}$$

where s_1, s_2 and s_3 are new third-order compliance constants defined in Appendix. Several acoustoelastic relationships for specific propagation and displacement directions relative to uniaxial or equal triaxial stress can be established from Eq. (20) and use of Table 1 where the various operations involving $\mathring{\boldsymbol{\sigma}}$ have been determined. In Table 1 the term σ^i is the pertinent stress component for the particular loading case. For general $\boldsymbol{\sigma}^i$, consider a longitudinal wave propagating in the $\hat{\mathbf{n}} = [100]$ direction, then Eq. (20) can be shown to reduce to

$$\begin{aligned}\Delta\lambda_{[100]} = {} & \left(1 + 4s_{44}\lambda^0\right)\sigma_{11}^i + \left(4s_{12}\lambda^0 - \frac{s_1}{27s^2} - \frac{s_2}{18s_{44}^2}\right)\left(\sigma_{11}^i + \sigma_{22}^i + \sigma_{33}^i\right) \\ & - \left(\frac{s_2}{9s_{44}s} + \frac{s_3}{36s_{44}^2}\right)\left(2\sigma_{11}^i - \sigma_{22}^i - \sigma_{33}^i\right),\end{aligned} \tag{21}$$

where the subscript [100] is used to denote the displacement direction, $\lambda^0 = (1 + s_{12}/s_{44})/s$, and $s = 3s_{12} + 2s_{44}$. For a shear wave propagating in the $\hat{\mathbf{n}} = [100]$ direction and displacement in the $\hat{\mathbf{u}} = [010]$ direction,

$$\Delta\lambda_{[010]} = \sigma_{11}^i + \sigma_{22}^i + \left(\frac{s_{12}}{2s_{44}} - \frac{s_2}{24s_{44}^2}\right)\left(\sigma_{11}^i + \sigma_{22}^i + \sigma_{33}^i\right) - \frac{s_3}{48s_{44}^2}\left(\sigma_{11}^i + \sigma_{22}^i - 2\sigma_{33}^i\right). \tag{22}$$

where $\lambda^0 = (4s_{44})^{-1}$. The case of a shear wave propagating in the $\hat{\mathbf{n}} = [100]$ direction with displacement in the $\hat{\mathbf{u}} = [001]$ direction follows by interchanging the 22 and 33 components of the stress tensor $\boldsymbol{\sigma}^i$ in Eq. (22). Acoustoelastic birefringence is observed when constructing the difference in these two shear wave cases, $\Delta\lambda_{[010]} - \Delta\lambda_{[001]} = (1 - s_3/(4s_{44})^2)(\sigma_{22}^i - \sigma_{33}^i)$, which is equivalent to the traditional expression $\Delta\lambda_{[010]} - \Delta\lambda_{[001]} = (1 + c_{456}/c_{44})(\sigma_{22}^i - \sigma_{33}^i)$.

Often, the third-order elastic parameters are determined from observing how the parameter $\Delta\lambda$ changes as a function of stress [16,21,27]. Thus, the so-called stress derivatives $\mathrm{d}\Delta\lambda/\mathrm{d}\sigma^i$ are of importance. Table 2 gives expressions for the stress derivatives for the uniaxial and equal triaxial stress cases. The expressions in Eqs. (21), (22), and the entries in Table 2 are new forms of acoustoelastic expressions and are fully consistent with the "natural velocity" formulas seen in

Table 2
Stress derivatives. The first subscript ($\perp$ or $||$) is propagation direction relative to the loading direction in the uniaxial stress. For shear waves, a second subscript ($\perp$ or $||$) indicates the displacement direction relative to the loading direction. No subscripts are needed for equal triaxial loading case. Note that $s = 3s_{12} + 2s_{44}$.

Mode	Loading	$\mathrm{d}\Delta\lambda/\mathrm{d}\sigma^i$
$L_\perp$	Uniaxial	$-\frac{2}{9} + \frac{2s}{9s_{44}} - \frac{4s_{44}}{9s} - \frac{1}{108s^2s_{44}^2}\left[4s_{44}^2s_1 + 6s_2s\left(s-2s_{44}\right) - 3s_3s^2\right]$
$L_{\|\|}$	Uniaxial	$\frac{19}{9} + \frac{2s}{9s_{44}} + \frac{8s_{44}}{9s} - \frac{1}{54s^2s_{44}^2}\left[2s_1s_{44}^2 + 3s_2s\left(s+4s_{44}\right) + 3s_3s^2\right]$
$T_{\perp\|\|}$ or $T_{\|\|\perp}$	Uniaxial	$\frac{2}{3} + \frac{s}{6s_{44}} - \frac{1}{48s_{44}^2}\left(2s_2 + s_3\right)$
$T_{\perp\perp}$	Uniaxial	$-\frac{1}{3} + \frac{s}{6s_{44}} - \frac{1}{24s_{44}^2}\left(s_2 - s_3\right)$
L	Equal triaxial	$\frac{5}{3} + \frac{2s}{3s_{44}} - \frac{1}{18s^2s_{44}^2}\left(2s_1s_{44}^2 + 3s_2s^2\right)$
T	Equal triaxial	$1 + \frac{s}{2s_{44}} - \frac{s_2}{8s_{44}^2}$

Table 3
Stress derivatives for incompressible solids. In the incompressible limit, $s \to 0$, $s_1 \to 0$, $s_2 \to 8s_{44}^2$, $s_3 \to 8s_{456}$ with s_{44} and s_{456} remaining finite.

Mode	Loading	$\mathrm{d}\Delta\lambda/\mathrm{d}\sigma^i$
$T_{\perp\|\|}$ or $T_{\|\|\perp}$	Uniaxial	$\frac{1}{3}\left(1 - \frac{s_3}{16s_{44}^2}\right)$
$T_{\perp\perp}$	Uniaxial	$-\frac{2}{3}\left(1 - \frac{s_3}{16s_{44}^2}\right)$
T	Equal triaxial	0

Table IV of Thurston and Brugger [21] once the conversions between stiffnesses and compliances are made. The natural velocity as defined by Thurston and Brugger [21] refers to the wave speed relative to the natural dimensions of the solid. The natural velocity is advantageous in practical cases involving applied loads because it can be calculated using the travel time of the wave and the distance of propagation in the undeformed configuration of the material [21]. In other words, the model naturally accounts for changes in the distance of propagation during loading. In addition, as explained by Thurston and Brugger [21], the natural velocities (eigenvalues) are not influenced by the rotation of the displacement directions (eigenvectors) during the deformation. One does not need to consider how the displacements are being influenced by stress when measuring the stress-derivative of the natural velocity. Thus, a set of experiments to measure the several third-order elastic constants (or compliances) can be constructed by performing wave experiments along various pure mode directions [28,29]. Formulas for the natural velocities found in Thurston and Brugger [21] were used to measure the third-order elastic constants of quartz [27] and langasite [30] crystals.

The acoustoelastic relations can be formulated for incompressible materials by substituting the behavior of the compliances or stiffnesses subject to constraints of incompressibility. Destrade and Ogden [31] and Saccomandi and Vergori [32] derived the behavior of the third- and fourth-order stiffnesses and demonstrated that the shear stiffnesses remain finite whereas other constants are unbound. Destrade and Ogden applied these results to traditional acoustoelasticty to derive the velocities of shear waves in incompressible solids undergoing either uniaxial or uniform pressure loading [31]. Recently, Kube derived the behavior of the second-, third-, and fourth-order compliances for use in stress-based formalisms of elastodynamics including the present work [33]. Unlike the stiffnesses, all of the compliance constants were found to remain finite when constraints of incompressibility are applied [33]. The present model is easily extended to incompressibility by utilizing the results: $s \to 0$, $s_{12} \to -\frac{2}{3}s_{44}$, $s_1 \to 0$, $s_2 \to 8s_{44}^2$, $s_3 \to 8s_{456}$ with s_{44} and s_{456} remaining finite [33]. Applying these results to the case of the shear wave in Eq. (22) gives

$$\Delta\lambda_{[010]} = \left(\frac{s_3}{(4s_{44})^2} - 1\right)\left(p^i + \sigma_{33}^i\right) \tag{23}$$

where $p^i = -\left(\sigma_{11}^i + \sigma_{22}^i + \sigma_{33}^i\right)/3$. The case for a shear wave propagating in $\hat{\mathbf{n}} = [100]$ while having displacement in $\hat{\mathbf{u}} = [001]$ direction is obtained, as before, by swapping the terms σ_{22}^i and σ_{33}^i. The birefringence relationships are the same as the compressible case. Table 3 provides the three finite cases when incompressibility is applied to the compliance terms seen in the entries of Table 2.

The formula in Eq. (23) and results in Table 2 with the stiffness/compliance relationships $s_3 = -c_{456}/c_{44}^3$ and $s_{44} = 1/(4c_{44})$ are new and different than those reported previously [31,34]. This disparity stems simply from the present derivation being formulated relative to the natural coordinates rather than the initial coordinates. The expressions relative to initial and natural coordinates are found in Tables II and V of Kube et al. respectively [23]. The results in the present work are consistent with Table V whereas the results seen in Table 2 are consistent with Refs. [31] and [34] as expected.

The consideration of acoustoelasticity of incompressible materials demonstrates a possible advantage of using the stress formulation. Namely, the stress formulation relies on second- and third-order compliance constants that must remain finite for incompressible solids whereas some stiffness components are unbound and can asymptotically approach infinity at different rates [31–33].

4. Connection with the displacement formulation

In this section, the results obtained in Section 3 are shown to be consistent with the traditional acoustoelastic formulas derived previously [20,21,23,27]. The time harmonic displacement solution for the unstressed medium satisfies

$$\left(c_{ijkl}\hat{n}_j\hat{n}_l - \lambda^0\delta_{ik}\right)\hat{u}_k = 0 \tag{24}$$

where $c_{ijkl}s_{klmn} = s_{ijkl}c_{klmn} = I_{ijmn}$. Hence, $\mathring{\sigma}^0_{ij} \to c_{ijkl}\hat{n}_l\hat{u}_k$ and $\mathring{t}^0_i \to c_{ijkl}\hat{n}_j\hat{n}_l\hat{u}_k = \lambda^0\hat{u}_i$. At the same time, the analog of Eq. (7), which relates strain to stress is

$$\sigma^f_{ij} = c_{ijkl}E^f_{kl} + \frac{1}{2}c_{ijklmn}E^f_{kl}E^f_{mn} \tag{25}$$

where the third order stiffness is

$$c_{ijklmn} = -c_{ijpq}c_{klrs}c_{mntu}s_{pqrstu}. \tag{26}$$

Recall, that the small initial strain is

$$e^i_{ij} = s_{ijkl}\sigma^i_{kl} \tag{27}$$

then Eq. (19) becomes

$$\Delta\lambda = \hat{n}_k\hat{n}_l\sigma^i_{kl} + 2\lambda^0\hat{u}_i\hat{u}_j e^i_{ij} + c_{ijklmn}\hat{u}_i\hat{u}_k\hat{n}_j\hat{n}_l e^i_{mn}. \tag{28}$$

At this order of approximation, the small initial strain in Eqs. (27) and (28) allows for the second Piola–Kirchhoff stress to be approximated by the Cauchy stress if desired. For an isotropic solid, Eq. (28) expressed in terms of the initial strain $\mathbf{e}^i$ reduces to

$$\begin{aligned}\Delta\lambda = {} & 2\lambda^0\hat{\mathbf{u}}\cdot\mathbf{e}^i\hat{\mathbf{u}} + \frac{1}{3}(3\kappa-2\mu)\operatorname{tr}\mathbf{e}^i + 2\mu\,\hat{\mathbf{n}}\cdot\mathbf{e}^i\hat{\mathbf{n}} + \frac{c_1}{27}\left(\hat{\mathbf{n}}\cdot\hat{\mathbf{u}}\right)^2\operatorname{tr}\mathbf{e}^i \\ & + \frac{c_2}{6}\left[\left(1-(\hat{\mathbf{n}}\cdot\hat{\mathbf{u}})^2\right)\operatorname{tr}\mathbf{e}^i + 4\left(\hat{\mathbf{n}}\cdot\hat{\mathbf{u}}\right)\left(\hat{\mathbf{n}}\cdot\mathbf{e}^i\hat{\mathbf{u}}\right)\right] \\ & + \frac{c_3}{36}\left[9\left(\hat{\mathbf{n}}\cdot\mathbf{e}^i\hat{\mathbf{n}} + \hat{\mathbf{u}}\cdot\mathbf{e}^i\hat{\mathbf{u}}\right) - 2\left(3-(\hat{\mathbf{n}}\cdot\hat{\mathbf{u}})^2\right)\operatorname{tr}\mathbf{e}^i - 6\left(\hat{\mathbf{n}}\cdot\hat{\mathbf{u}}\right)\left(\hat{\mathbf{n}}\cdot\mathbf{e}^i\hat{\mathbf{u}}\right)\right],\end{aligned} \tag{29}$$

where $\kappa = (9s_{12}+6s_{44})^{-1}$ and $\mu = c_{44} = (4s_{44})^{-1}$ are the bulk and shear moduli, respectively. In arriving at Eq. (29), we made use of the isotropic tensors having components of the third-order elastic stiffnesses given in Appendix. Using the stiffness/compliance relations derived in the Appendix, Eq. (29) can be cast in terms of the initial stress $\boldsymbol{\sigma}^i$,

$$\begin{aligned}\Delta\lambda = {} & \frac{\lambda^0}{\mu}\hat{\mathbf{u}}\cdot\boldsymbol{\sigma}^i\hat{\mathbf{u}} + \frac{2}{3}\lambda^0\left(\frac{1}{3\kappa}-\frac{1}{2\mu}\right)\operatorname{tr}\boldsymbol{\sigma}^i + \hat{\mathbf{n}}\cdot\boldsymbol{\sigma}^i\hat{\mathbf{n}} + \frac{c_1}{81\kappa}\left(\hat{\mathbf{n}}\cdot\hat{\mathbf{u}}\right)^2\operatorname{tr}\boldsymbol{\sigma}^i \\ & + \frac{c_2}{18}\left[\left(\frac{1}{\kappa}+\left(\frac{1}{3\kappa}-\frac{2}{\mu}\right)\left(\hat{\mathbf{n}}\cdot\hat{\mathbf{u}}\right)^2\right)\operatorname{tr}\boldsymbol{\sigma}^i + \frac{6}{\mu}\left(\hat{\mathbf{n}}\cdot\hat{\mathbf{u}}\right)\left(\hat{\mathbf{n}}\cdot\boldsymbol{\sigma}^i\hat{\mathbf{u}}\right)\right] \\ & + \frac{c_3}{72\mu}\left[9\left(\hat{\mathbf{n}}\cdot\boldsymbol{\sigma}^i\hat{\mathbf{n}} + \hat{\mathbf{u}}\cdot\boldsymbol{\sigma}^i\hat{\mathbf{u}}\right) - 2\left(3-(\hat{\mathbf{n}}\cdot\hat{\mathbf{u}})^2\right)\operatorname{tr}\boldsymbol{\sigma}^i - 6\left(\hat{\mathbf{n}}\cdot\hat{\mathbf{u}}\right)\left(\hat{\mathbf{n}}\cdot\boldsymbol{\sigma}^i\hat{\mathbf{u}}\right)\right].\end{aligned} \tag{30}$$

The expression seen in Eq. (30) is equivalent to Eq. (41) in Kube et al. [23] once the third-order elastic constants c_1, c_2, and c_3 are converted to the Landau and Lifshitz constants $\mathcal{A}$, $\mathcal{B}$, and $\mathcal{C}$ through the relationships $c_1 = 6\mathcal{A} + 54\mathcal{B} + 54\mathcal{C}$, $c_2 = 2\mathcal{A}+6\mathcal{B}$, and $c_3 = 2\mathcal{A}$. We remind the reader that the expressions derived in this work and Eq. (41) in Kube et al. [23] are based on coordinates and density in the undeformed or natural configuration. The corresponding expressions based on coordinates and density relative to the deformed material configuration are found in Kube et al. [23] and Thurston and Brugger [21]. By writing Eq. (30) in terms of s_1, s_2, and s_3 using the relationships found in Eq. (A.11), we obtain the alternative expression in terms of compliances,

$$\begin{aligned}\Delta\lambda = {} & \frac{\lambda^0}{\mu}\hat{\mathbf{u}}\cdot\boldsymbol{\sigma}^i\hat{\mathbf{u}} + \frac{2}{3}\lambda^0\left(\frac{1}{3\kappa}-\frac{1}{2\mu}\right)\operatorname{tr}\boldsymbol{\sigma}^i + \hat{\mathbf{n}}\cdot\boldsymbol{\sigma}^i\hat{\mathbf{n}} - \frac{s_1\kappa^2}{3}\left(\hat{\mathbf{n}}\cdot\hat{\mathbf{u}}\right)^2\operatorname{tr}\boldsymbol{\sigma}^i \\ & - \frac{2s_2\mu}{9}\left[\left(3\mu+(\mu-6\kappa)\left(\hat{\mathbf{n}}\cdot\hat{\mathbf{u}}\right)^2\right)\operatorname{tr}\boldsymbol{\sigma}^i + 18\kappa\left(\hat{\mathbf{n}}\cdot\hat{\mathbf{u}}\right)\left(\hat{\mathbf{n}}\cdot\boldsymbol{\sigma}^i\hat{\mathbf{u}}\right)\right] \\ & - \frac{s_3\mu^2}{9}\left[9\left(\hat{\mathbf{n}}\cdot\boldsymbol{\sigma}^i\hat{\mathbf{n}} + \hat{\mathbf{u}}\cdot\boldsymbol{\sigma}^i\hat{\mathbf{u}}\right) - 2\left(3-(\hat{\mathbf{n}}\cdot\hat{\mathbf{u}})^2\right)\operatorname{tr}\boldsymbol{\sigma}^i - 6\left(\hat{\mathbf{n}}\cdot\hat{\mathbf{u}}\right)\left(\hat{\mathbf{n}}\cdot\boldsymbol{\sigma}^i\hat{\mathbf{u}}\right)\right].\end{aligned} \tag{31}$$

Finally, this form for $\Delta\lambda$ is equal to the expression in Eq. (20) by noting the stress terms in the latter can be expressed as $\operatorname{tr}\mathring{\boldsymbol{\sigma}} = 3\kappa(\hat{\mathbf{n}}\cdot\hat{\mathbf{u}})$, $\hat{\mathbf{n}}\cdot\mathring{\boldsymbol{\sigma}}\boldsymbol{\sigma}^i\mathring{\boldsymbol{\sigma}}\hat{\mathbf{n}} = \hat{\mathbf{u}}\cdot\boldsymbol{\sigma}^i\hat{\mathbf{u}}$, $\operatorname{tr}(\mathring{\boldsymbol{\sigma}}\boldsymbol{\sigma}^i) = (\kappa-2\mu/3)(\hat{\mathbf{n}}\cdot\hat{\mathbf{u}})\operatorname{tr}\boldsymbol{\sigma}^i + 2\mu\hat{\mathbf{n}}\cdot\boldsymbol{\sigma}^i\hat{\mathbf{u}}$, $\operatorname{tr}\mathring{\boldsymbol{\sigma}}^2 = 2\mu^2 + (3\kappa^2+2\mu^2/3)(\hat{\mathbf{n}}\cdot\hat{\mathbf{u}})^2$, $\operatorname{tr}(\mathring{\boldsymbol{\sigma}}\boldsymbol{\sigma}^i\mathring{\boldsymbol{\sigma}}) = (\kappa-2\mu/3)^2(\hat{\mathbf{n}}\cdot\hat{\mathbf{u}})^2\operatorname{tr}\boldsymbol{\sigma}^i + 2\mu\,(2\kappa-\mu/3)\,(\hat{\mathbf{n}}\cdot\hat{\mathbf{u}})\hat{\mathbf{n}}\cdot\boldsymbol{\sigma}^i\hat{\mathbf{u}} + \mu^2\left(\hat{\mathbf{n}}\cdot\boldsymbol{\sigma}^i\hat{\mathbf{n}} + \hat{\mathbf{u}}\cdot\boldsymbol{\sigma}^i\hat{\mathbf{u}}\right)$.

5. Conclusion

The pure stress formulation of elastodynamics [2,9] has been extended and applied to acoustoelasticity. The stress equations of motion provide a more natural setting for acoustoelasticity when the initial stress in the material is of primary interest (rather than initial strain). In the derivation, it is shown that the second- and third-order compliance constants fundamentally connect the wave velocities to initial stress. This differs from the traditional displacement formulation in which wave velocities relate to initial stress through combinations of stiffnesses and compliances. Consistency between the present model and displacement-based model is established through the stiffness–compliance relationships developed in the Appendix. The stiffness–compliance relationships are not new. Thus, the traditional acoustoelastic relations could have been cast in terms of compliances only without resort to the pure stress formulation. To our knowledge, this connection has not been established previously. The new acoustoelastic relations are established for the canonical loading cases of uniaxial and equal triaxial stress. General formulas for isotropic symmetry and arbitrary loading are given explicitly. The corresponding relationships for incompressible materials is then given using the behavior of the compliance constants [31] for situations when the material tends toward incompressibility.

CRediT authorship contribution statement

Christopher M. Kube: Conceptualization, Writing – original draft, Writing – review & editing. **Andrew N. Norris:** Conceptualization, Writing – original draft, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Appendix. Relations between isotropic third order stiffness and compliance moduli

A.1. Proposed new third order elasticity coefficients

In addition to the standard reduced index or Voigt notation c_{IJK} $(= c_{ijklmn})$ there are several alternative notations for third-order elastic constants, including those of [35–39] which can each be expressed in terms of c_{IJK}, see Table 1 of Ref. [16] and of Ref. [40]. Despite this multiplicity of notation, none are convenient for relating elements of stiffness c_{IJK} and compliance s_{IJK} to one another. Here we propose a notation for TOE coefficients designed for just this purpose.

The motivation is the fact that isotropic linear elasticity is simply cast using the hydrostatic and deviatoric parts of the stress and strain. These are defined respectively as $\boldsymbol{\sigma}'$, $\mathbf{e}'$ and $\boldsymbol{\sigma}''$, $\mathbf{e}''$, through the relations

$$\begin{aligned} \boldsymbol{\sigma} &= \boldsymbol{\sigma}' + \boldsymbol{\sigma}'', \quad \boldsymbol{\sigma}' = \hat{\boldsymbol{\delta}}\ \mathrm{tr}\,\boldsymbol{\sigma}, \\ \mathbf{e} &= \mathbf{e}' + \mathbf{e}'', \quad \mathbf{e}' = \hat{\boldsymbol{\delta}}\ \mathrm{tr}\,\mathbf{e}, \end{aligned} \tag{A.1}$$

where

$$\hat{\delta}_{ij} = \frac{1}{3}\delta_{ij}. \tag{A.2}$$

The linearly elastic constitutive relations are then

$$\boldsymbol{\sigma}' = 3\kappa\,\mathbf{e}' \ \text{ and } \ \boldsymbol{\sigma}'' = 2\mu\,\mathbf{e}'' \tag{A.3}$$

where κ and $\mu = c_{44}$ are the bulk and shear moduli.

The only second order, or quadratic, isotropic combinations of the hydrostatic and deviatoric parts of $\mathbf{e}$ are $(\mathrm{tr}\,\mathbf{e})^2$ and $\mathrm{tr}(\mathbf{e}''^2)$, implying that the strain energy density is a linear combination of the two. Similarly, the only third order isotropic combinations are $(\mathrm{tr}\,\mathbf{e})^3$, $\mathrm{tr}\,\mathbf{e}\ \mathrm{tr}(\mathbf{e}''^2)$, and $\mathrm{tr}(\mathbf{e}''^3)$. This indicates that the strain energy density U can be expressed as

$$U = \frac{1}{2!}\Big(3\kappa\,\frac{(\mathrm{tr}\,\mathbf{E})^2}{3} + 2\mu\ \mathrm{tr}(\mathbf{E}''^2)\Big) + \frac{1}{3!}\Big(c_1(\mathrm{tr}\,\mathbf{E})^3 + c_2\,\mathrm{tr}\,\mathbf{E}\ \mathrm{tr}(\mathbf{E}''^2) + c_3\,\mathrm{tr}(\mathbf{E}''^3)\Big) + \cdots . \tag{A.4}$$

where c_1, c_2 and c_3 are the proposed TOE constants. We next relate these elastic constants to the standard ones appearing in the general form of the strain energy

$$U = \frac{1}{2!}c_{ijkl}E_{ij}E_{kl} + \frac{1}{3!}c_{ijklmn}E_{ij}E_{kl}E_{mn} + \cdots . \tag{A.5}$$

A.2. Connection with c_{ijklmn} and s_{ijklmn}

Define the fourth order isotropic tensors **J** and **K**

$$J_{ijkl} = \frac{1}{3}\delta_{ij}\delta_{kl}, \quad K_{ijkl} = I_{ijkl} - J_{ijkl} \tag{A.6}$$

where $I_{ijkl} = \frac{1}{2}(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk})$ is the identity. Note that **J** and **K** have the symmetries of elastic moduli, and satisfy $\mathbf{J}^2 = \mathbf{J}$, $\mathbf{K}^2 = \mathbf{K}$, $\mathbf{JK} = \mathbf{KJ} = 0$ where $(AB)_{ijkl} = A_{ijmn}B_{mnkl}$. The partition of unity $\mathbf{J} + \mathbf{K} = \mathbf{I}$ lies behind the independence of the hydrostatic/deviatoric components of stress and strain: $\boldsymbol{\sigma}' = \mathbf{J}\boldsymbol{\sigma}$, $\boldsymbol{\sigma}'' = \mathbf{K}\boldsymbol{\sigma}$. Also, second order stiffness and compliance tensors are

$$c_{ijkl} = 3\kappa J_{ijkl} + 2\mu K_{ijkl}, \quad s_{ijkl} = (3\kappa)^{-1}J_{ijkl} + (2\mu)^{-1}K_{ijkl}. \tag{A.7}$$

Referring to Eqs. (A.4) and (A.5), the third order isotropic moduli can be expressed as linear combinations of three independent tensors,

$$c_{ijklmn} = c_1 P_{ijklmn} + c_2 Q_{ijklmn} + c_3 R_{ijklmn} \tag{A.8}$$

where the sixth order isotropic tensors **P**, **Q** and **R** have elements

$$P_{ijklmn} = \hat{\delta}_{ij}\hat{\delta}_{kl}\hat{\delta}_{mn}, \tag{A.9a}$$

$$Q_{ijklmn} = \hat{\delta}_{ij}K_{klmn} + \hat{\delta}_{kl}K_{mnij} + \hat{\delta}_{mn}K_{ijkl}, \tag{A.9b}$$

$$R_{ijklmn} = K_{ijpq}K_{klqr}K_{mnrp}. \tag{A.9c}$$

It then follows from Eq. (26) and the definitions in Eqs. (A.9), using the properties of **J** and **K**, along with identities like $J_{ijkl}\hat{\delta}_{kl} = \hat{\delta}_{ij}$, that

$$s_{ijklmn} = s_1 P_{ijklmn} + s_2 Q_{ijklmn} + s_3 R_{ijklmn}, \tag{A.10}$$

where

$$s_1 = -\frac{c_1}{(3\kappa)^3}, \tag{A.11a}$$

$$s_2 = -\frac{c_2}{3\kappa(2\mu)^2}, \tag{A.11b}$$

$$s_3 = -\frac{c_3}{(2\mu)^3}. \tag{A.11c}$$

These simple connections between the TOE coefficients are the primary reason for introducing a new set of TOE moduli. In addition, the new coefficients have physical meaning analogous to the bulk and shear moduli: the c_1 term involves only hydrostatic stress/strain, the c_3 term is related to deviatoric (shear) stress/strain, while c_2 is the only energy contribution at cubic approximation to involve coupling between hydrostatic and deviatoric deformations.

This suggests an algebra for relating s_{IJK} and c_{IJK}. Of the six moduli c_{111}, c_{112}, c_{123}, c_{166}, c_{144}, c_{456}, we choose c_{112}, c_{144} and c_{456} as primary, in terms of which the others are

$$\begin{pmatrix} c_{111} \\ c_{123} \\ c_{166} \end{pmatrix} = \begin{bmatrix} 1 & 4 & 8 \\ 1 & -2 & 0 \\ 0 & 1 & 2 \end{bmatrix} \begin{pmatrix} c_{112} \\ c_{144} \\ c_{456} \end{pmatrix}. \tag{A.12}$$

Evaluation of the elements of P, Q and R, as indicated in Table 4, imply the relations

$$c_1 = 27c_{112} + 24c_{456}, \tag{A.13a}$$

$$c_2 = 6c_{144} + 8c_{456}, \tag{A.13b}$$

$$c_3 = 8c_{456}. \tag{A.13c}$$

These, along with the pivot relations in Eq. (A.11), imply explicit relations between the TOE stiffness and compliance,

$$\begin{pmatrix} c_{112} \\ c_{144} \\ c_{456} \end{pmatrix} = \begin{bmatrix} \frac{1}{27} & 0 & -\frac{1}{9} \\ 0 & \frac{1}{6} & -\frac{1}{6} \\ 0 & 0 & -\frac{1}{8} \end{bmatrix} \begin{bmatrix} -(3\kappa)^3 & 0 & 0 \\ 0 & -3\kappa(2\mu)^2 & 0 \\ 0 & 0 & -(2\mu)^3 \end{bmatrix} \begin{bmatrix} 27 & 0 & 24 \\ 0 & 6 & 8 \\ 0 & 0 & 8 \end{bmatrix} \begin{pmatrix} s_{112} \\ s_{144} \\ s_{456} \end{pmatrix}. \tag{A.14}$$

In summary,

$$\begin{pmatrix} c_{112} \\ c_{144} \\ c_{456} \end{pmatrix} = -(3\kappa)^3 \begin{bmatrix} 1 & 0 & \frac{8}{9}(1-\alpha^3) \\ 0 & \alpha^2 & \frac{4}{3}(1-\alpha)\alpha^2 \\ 0 & 0 & \alpha^3 \end{bmatrix} \begin{pmatrix} s_{112} \\ s_{144} \\ s_{456} \end{pmatrix}, \tag{A.15a}$$

Table 4
Elements of the third order isotropic tensors P, Q and R.

IJK	P_{IJK}	Q_{IJK}	R_{IJK}
112	$\frac{1}{27}$	0	$-\frac{1}{9}$
144	0	$\frac{1}{6}$	$-\frac{1}{6}$
456	0	0	$\frac{1}{8}$

$$\begin{pmatrix} s_{112} \\ s_{144} \\ s_{456} \end{pmatrix} = -(3\kappa)^{-3} \begin{bmatrix} 1 & 0 & \frac{8}{9}(1-\alpha^{-3}) \\ 0 & \alpha^{-2} & \frac{4}{3}(1-\alpha^{-1})\alpha^{-2} \\ 0 & 0 & \alpha^{-3} \end{bmatrix} \begin{pmatrix} c_{112} \\ c_{144} \\ c_{456} \end{pmatrix} \tag{A.15b}$$

where $\alpha = \frac{2\mu}{3\kappa}$. More explicitly,

$$s_{123} = -\frac{1}{27\kappa^3}\left(c_{123} + 2c_{144} + \frac{8}{9}c_{456}\right) + \frac{1}{18\kappa\mu^2}(3c_{144} + 4c_{456}) - \frac{2}{9\mu^3}c_{456}, \tag{A.16a}$$

$$s_{144} = -\frac{1}{36\kappa\mu^2}(3c_{144} + 4c_{456}) + \frac{1}{6\mu^3}c_{456}, \tag{A.16b}$$

$$s_{456} = -\frac{1}{8\mu^3}c_{456} \tag{A.16c}$$

and $s_{112} = s_{123} + 2s_{144}$, $s_{155} = s_{144} + 2s_{456}$, $s_{111} = s_{123} + 6s_{144} + 8s_{456}$.

The relations Eq. (A.11) between third order stiffness and compliance coefficients are known. Thus, Cousins [41] presented six such relations valid for materials of cubic symmetry, which reduce to the three in Eq. (A.11) under isotropy, although the physical connection with the hydrostatic and deviatoric partition of stress and strain was not noted. In a separate note we show how the six Cousins identities derive from an analogous partition of stress and strain appropriate to cubic symmetry.

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