

Nonlinear Elastic Relations in Equations of States of Solids

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Abstract

This paper presents various nonlinear elastic relationships of solids subjected to homogeneous triaxial stresses. The broad spectra of the material symmetry of solids are covered, ranging from the triclinic to the isotropic symmetry. Special emphasis is given to three cases in which (i) the directions of three principal stresses coincide with those of orthorhombic/orthotropic or higher symmetry in materials, (ii) the direction of the uniaxial stresses is along one of the material symmetry directions, and (iii) hydrostatic pressures are applied. It is found that nonlinear relations between stress and strain, and between elastic moduli and stress/strain, are more conveniently described in terms of the second, third, and fourth order elastic compliance constants rather than using the corresponding stiffness constants. It is shown that not only pure strain but also rotation contributes to nonlinear elasticity in solids with monotropic or lower symmetry.

Keywords: nonlinear elastic relations in equations of state of solids; monotropic or lower symmetry; orthorhombic/orthotropic on higher symmetry; Cauchy-Green deformation gradients tensor G_{ij} ; thermodynamic elastic stiffness/compliance coefficients $C_{ijkl\dots}$ and $S_{ijkl\dots}$

1. Introduction

This paper extends nonlinear relations between strains and stresses and those between effective elastic moduli and stress/strain to include the contribution of the fourth order elastic constants, and express these relations in the most general case of anisotropy, which include monotropic/monoclinic and triclinic systems.

Murnaghan [1,2] developed the finite deformation theory using the second and third order elastic constants of isotropic and crystalline materials. One of his fundamental contributions is a derivation of a nonlinear relation between the Cauchy stress and thermodynamic (second Piola-Kirchhoff) stress. Birch [3,4] extended Murnaghan's work to cubic materials and derived the well-known Birch's equation of state in geology, which relates the density or the specific volume to a hydrostatic pressure acting on the material.

Brugger [5] gave an elegant definition of higher order elastic constants, and using his definition, Thurston and Brugger [6] derived expressions for the ultrasonic wavespeeds in stressed solids in terms of the second and third order elastic constants. Extensive data of the second and third order elastic constants of numerous solids of various symmetry groups were compiled by Hearmon [7], and Every and McCurdy [8]. Some useful relations at finite deformation between various thermodynamic and effective elastic coefficients are described by Thurston [9,10]. Thermodynamics of finite deformation in reference to the stress-free state is given in detail in Ref. [10] and in the book of Wallace [11]. The author of this article [12] generalized the thermodynamics of elastic solids to a state of arbitrary stress level and derived numerous relations between different thermodynamic variables at finite deformation, including the expressions of the Young's modulus and Poisson's ratio in terms of the second order thermodynamic elastic stiffness coefficients and three principal stresses acting on the material.

The amount of nonlinearity as compared with that of linearity for ordinary solids is quite small below the elastic limit of a material for strain less than 1 % but it can be detected by precision ultrasonic measurements [13,14]. For elastic strains larger than 1%, which are usually observed under large hydrostatic/quasi-hydrostatic pressures, the contribution of the nonlinearity term is significant and should be accounted for in the meaningful solution of elasticity problems.

In the case of homogeneous uniaxial loading of fine whiskers, very large strains may be present. A nonlinear stress-strain relation and a strain dependence of the Young's modulus were

demonstrated in a tension testing of various whiskers [15,16]. Ruoff [17,18] analyzed similar problems using finite elasticity and found the conditions for macroscopic elastic instabilities in tension and compression of perfect crystals of diamond, silicon and germanium. Recently, the author [19] derived the various nonlinear relations between stress and strain, and the stress/strain dependence of the Young's modulus and Poisson's ratio, in terms of the second and third order elastic constants for cubic and isotropic solids under homogeneous uniaxial loading.

The present paper extends the author's recent work [19] to the case of general anisotropic solids under triaxial stresses. There have been so far piecemeal approaches by the fourth order elastic constants are related to wave speeds and effective elastic moduli of solids under stress in cubic and isotropic media. The contribution of the fourth order elastic stiffness/compliance constants is extended to the most general case of anisotropy, which include monotropic/monoclinic and triclinic systems. It will be shown that in the most general case under triaxial stresses, not only the pure strain but the rotation of a material plays a role in these nonlinear relations.

1. Description of deformation states and symbols

Consider a solid specimen in the stress-free natural state. The specimen may possess anisotropy, which ranges from isotropic to triclinic symmetry system. The Cartesian coordinates of a particle in the natural state are denoted by vector \mathbf{a} . The specimen is stressed to an arbitrary finite deformation under homogeneously applied triaxial stresses. The stressed body is said to be in an initial state, which is a static state denoted by the Cartesian coordinates \mathbf{X} . The initial state can be any state, which may include the natural state \mathbf{a} as a special case, since deformation is arbitrary. Finally, a small deformation is superposed on the initial state. We call this state a current or the final state, which is denoted the Cartesian coordinates \mathbf{x} . We denote the density of the natural, initial and current states by ρ_a , ρ_X , and ρ_x , respectively.

Let a tensor $\mathbf{T}(\mathbf{A};\mathbf{B})$ denote a physical variable. The variable \mathbf{T} is evaluated in the state represented by a coordinate system \mathbf{A} and it takes \mathbf{B} as a reference coordinate system from which a deformation is measured. Both \mathbf{A} and \mathbf{B} can be \mathbf{x} , \mathbf{X} , or \mathbf{a} . When both \mathbf{A} and \mathbf{B} are represented by the same coordinate system, we denote the tensor \mathbf{T} by a single argument, as shown in the examples $\mathbf{T}(\mathbf{a};\mathbf{a}) = \mathbf{T}(\mathbf{a})$, $\mathbf{T}(\mathbf{X};\mathbf{X}) = \mathbf{T}(\mathbf{X})$, and $\mathbf{T}(\mathbf{x};\mathbf{x}) = \mathbf{T}(\mathbf{x})$. The Cauchy stresses

$\sigma_{ij}(\mathbf{a})$, $\sigma_{ij}(\mathbf{X})$ and $\sigma_{ij}(\mathbf{x})$ are the typical examples of this representation and are always evaluated at and referred for deformation to the same coordinate system.

The displacements between various deformation states are given by

$$\mathbf{u}(\mathbf{X}; \mathbf{a}) = \mathbf{X} - \mathbf{a} \quad \mathbf{u}(\mathbf{x}; \mathbf{a}) = \mathbf{x} - \mathbf{a} \quad \mathbf{u}(\mathbf{x}; \mathbf{X}) = \mathbf{x} - \mathbf{X} \quad (1)$$

The deformation gradient $\partial X_i / \partial a_j$ and the displacement gradient $\partial u_i(\mathbf{X}; \mathbf{a}) / \partial a_j$ are not uniquely specified unless some constraints are given to the conventionally adopted Cartesian coordinate system. Even the application of hydrostatic pressures on the general case of a triclinic crystal induces not only pure strain but also rotation between the crystallographic axes a , b , and c . The angle between the a and c axes under applied (hydrostatic) stresses will change in a monoclinic/monotropic material, in which the ac plane is chosen to be a mirror plane. A particular Cartesian system is chosen such that it has its 3 direction along the c axis, its 1 direction in the ac plane and its 2 direction normal to the ac plane in order to make up a right-handed system. This coordinate system is consistent with the IRE standards [20]. In the chosen Cartesian system the orientation of the (1,3) material planes and that of the material lines lying along the 3 direction are fixed. The specified constraints are the relations between the chosen Cartesian system and material axes of symmetry. The Cartesian system under applied stresses may rotate with respect to the fixed reference in a laboratory. It does not matter, as long as the chosen Cartesian system uniquely specifies the deformation gradient with respect to axes of material symmetry, which may coincide with the crystallographic axes in crystalline materials.

These constraints can be easily translated into the special conditions on the deformation gradients. X_2 does not depend on a_1 or a_3 , because the (1,3) material planes have a fixed orientation. The material lines lying along the 3 direction are chosen to be independent of rotation, and therefore both X_1 and X_2 do not depend on a_3 . Thus, the constraints are expressed as

$$\partial X_1 / \partial a_3 = \partial X_2 / \partial a_3 = \partial X_2 / \partial a_1 = 0. \quad (2)$$

The symmetric part ε_{ij} and antisymmetric part ω_{ij} of the displacement gradient can be written as

$$\varepsilon_{ij}(\mathbf{X}; \mathbf{a}) = \frac{1}{2} \left[\frac{\partial u_i(\mathbf{X}; \mathbf{a})}{\partial a_j} + \frac{\partial u_j(\mathbf{X}; \mathbf{a})}{\partial a_i} \right] = \frac{1}{2} \left(\frac{\partial X_i}{\partial a_j} + \frac{\partial X_j}{\partial a_i} - \delta_{ij} \right) = \varepsilon_{ji}(\mathbf{X}; \mathbf{a}), \quad (3)$$

$$\omega_{ij}(\mathbf{X}; \mathbf{a}) = \frac{1}{2} \left[\frac{\partial u_i(\mathbf{X}; \mathbf{a})}{\partial a_j} - \frac{\partial u_j(\mathbf{X}; \mathbf{a})}{\partial a_i} \right] = \frac{1}{2} \left(\frac{\partial x_i}{\partial a_j} - \frac{\partial x_j}{\partial a_i} \right) = -\omega_{ji}(\mathbf{X}; \mathbf{a}). \quad (4)$$

A great majority of cases involving deformations in this article are from the natural state \mathbf{a} to the initial state \mathbf{X} . Hence, it is understood that when the argument specifying the evaluation and reference states is absent in the relevant expressions of deformation and displacement gradients, they are implied with the argument $\mathbf{X}; \mathbf{a}$ unless otherwise specified. The deformation gradients $\alpha_{ij} \equiv \partial X_i / \partial a_j$ are given by

$$\alpha_{ij} \equiv \frac{\partial X_i}{\partial a_j} = \delta_{ij} + \frac{\partial u_i}{\partial a_j} = \delta_{ij} + \varepsilon_{ij} + \omega_{ij}. \quad (5)$$

For convenience of notation, we introduce

$$u_{ij} \equiv \frac{\partial u_i}{\partial a_j} \quad \beta_{ij} \equiv \frac{\partial a_i}{\partial x_j}. \quad (6)$$

A Lagrangian strain η_{ij} with respect to the stress-free state is expressed as

$$\eta_{ij}(\mathbf{X}; \mathbf{a}) = \frac{1}{2} \left(\frac{\partial X_p}{\partial a_i} \frac{\partial X_p}{\partial a_j} - \delta_{ij} \right) = \frac{1}{2} (G_{ij} - \delta_{ij}) = \varepsilon_{ij} + \frac{1}{2} u_{pi} u_{pj}, \quad (7)$$

where and henceforth summation over the repeated indices is implied unless otherwise specified, and the Cauchy-Green deformation gradients tensor G_{ij} is defines as

$$G_{ij} \equiv \frac{\partial X_p}{\partial a_i} \frac{\partial X_p}{\partial a_j} . \quad (8)$$

Here, the symbol G_{ij} , instead of conventional C_{ij} , is used to denote the Cauchy-Green tensor in order to distinguish it from the elastic stiffness matrix commonly represented by $C_{\alpha\beta}$.

It follows from Eq. 4 and the constraints Eq. 2 that

$$\begin{aligned} \omega_{11} = \omega_{22} = \omega_{33} &= 0, \\ \omega_{12} = \varepsilon_{12} &= -\omega_{21}, \\ \omega_{23} = -\varepsilon_{23} &= -\omega_{23}, \\ \omega_{31} = \varepsilon_{12} &= -\omega_{13}. \end{aligned} \quad (9)$$

Let's define W_n as

$$W_n \equiv \frac{1}{2} \varepsilon_{ijn} \omega_{ij}, \quad (10)$$

where ε_{ijn} represents a permutation tensor. Multiplying ε_{rsn} on both sides, one obtains

$$\omega_{ij} = \varepsilon_{ijn} W_n = -\varepsilon_{ij1} \varepsilon_{23} + \varepsilon_{ij2} \varepsilon_{13} + \varepsilon_{ij3} \varepsilon_{12} . \quad (11)$$

With reference to the initial state, we define according to Brugger's convention [5] the thermodynamic stress $\tau_{ij}(\mathbf{x}; \mathbf{X})$ and the adiabatic and isothermal thermodynamic elastic stiffness/compliance coefficients $C_{ijkl\dots}$ and $S_{ijkl\dots}$ of the n 'th ($n \geq 2$) order as

$$\tau_{ij}(\mathbf{x}; \mathbf{X}) \equiv \rho_X \left[\frac{\partial U}{\partial \eta_{ij}}(\mathbf{x}; \mathbf{X}) \right]_{S; \mathbf{X}} = \rho_X \left[\frac{\partial F}{\partial \eta_{ij}}(\mathbf{x}; \mathbf{X}) \right]_{T; \mathbf{X}} \quad (12)$$

$$\eta_{ij}(\mathbf{x}; \mathbf{X}) \equiv \rho_X \left[\frac{\partial H}{\partial \tau_{ij}}(\mathbf{x}; \mathbf{X}) \right]_{S; \mathbf{X}} = \rho_X \left[\frac{\partial G}{\partial \tau_{ij}}(\mathbf{x}; \mathbf{X}) \right]_{T; \mathbf{X}} \quad (13)$$

$$C_{ijkl\dots}^S(\mathbf{x}; \mathbf{X}) \equiv \rho_X \left[\frac{\partial^n U}{\partial \eta_{ij}(\mathbf{x}; \mathbf{X}) \partial \eta_{kl}(\mathbf{x}; \mathbf{X}) \dots} \right]_{S; \mathbf{X}} = \left[\frac{\partial^{n-1} \tau_{ij}(\mathbf{x}; \mathbf{X})}{\partial \eta_{kl}(\mathbf{x}; \mathbf{X}) \dots} \right]_{S; \mathbf{X}} \quad (n \geq 2) \quad (14)$$

$$C_{ijkl\dots}^T(\mathbf{x}; \mathbf{X}) \equiv \rho_X \left[\frac{\partial^n F}{\partial \eta_{ij}(\mathbf{x}; \mathbf{X}) \partial \eta_{kl}(\mathbf{x}; \mathbf{X}) \dots} \right]_{T; \mathbf{X}} = \left[\frac{\partial^{n-1} \tau_{ij}(\mathbf{x}; \mathbf{X})}{\partial \eta_{kl}(\mathbf{x}; \mathbf{X}) \dots} \right]_{T; \mathbf{X}} \quad (n \geq 2) \quad (15)$$

$$S_{ijkl\dots}^S(\mathbf{x}; \mathbf{X}) \equiv \rho_X \left[\frac{\partial^n H}{\partial \tau_{ij}(\mathbf{x}; \mathbf{X}) \partial \tau_{kl}(\mathbf{x}; \mathbf{X}) \dots} \right]_{S; \mathbf{X}} = \left[\frac{\partial^{n-1} \eta_{ij}(\mathbf{x}; \mathbf{X})}{\partial \tau_{kl}(\mathbf{x}; \mathbf{X}) \dots} \right]_{S; \mathbf{X}} \quad (n \geq 2) \quad (16)$$

$$S_{ijkl\dots}^T(\mathbf{x}; \mathbf{X}) \equiv \rho_X \left[\frac{\partial^n G}{\partial \tau_{ij}(\mathbf{x}; \mathbf{X}) \partial \tau_{kl}(\mathbf{x}; \mathbf{X}) \dots} \right]_{T; \mathbf{X}} = \left[\frac{\partial^{n-1} \eta_{ij}(\mathbf{x}; \mathbf{X})}{\partial \tau_{kl}(\mathbf{x}; \mathbf{X}) \dots} \right]_{T; \mathbf{X}} \quad (n \geq 2), \quad (17)$$

where U , F , H , and G are respectively the internal energy, Helmholtz free energy, enthalpy, and Gibbs free energy per unit mass. S denotes the entropy and T the temperature. Both $C_{ijkl\dots}^{S/T}(\mathbf{x}; \mathbf{X})$ and $S_{ijkl\dots}^{S/T}(\mathbf{x}; \mathbf{X})$ are generally a function of stress. In the special case of the initial state being identical to the stress-free natural state, the quantities given in Eqs. 12-17 are defined by replacing \mathbf{X} by \mathbf{a} .

The special types of thermodynamic elastic coefficients are the second, third and fourth order elastic stiffness constants (SOESC, TOESC, and FOESC) defined as

$$\begin{aligned}
C_{ijkl}^{S/T}(\mathbf{a}) &\equiv C_{ijkl}^{S/T}(\mathbf{a}; \mathbf{a}) & C_{ijklmn}^{S/T}(\mathbf{a}) &\equiv C_{ijklmn}^{S/T}(\mathbf{a}; \mathbf{a}) & C_{ijklmnpq}^{S/T}(\mathbf{a}) &\equiv C_{ijklmnpq}^{S/T}(\mathbf{a}; \mathbf{a}) & (18) \\
(\text{SOESC}) & & (\text{TOESC}) & & (\text{FOESC}) & &
\end{aligned}$$

and the second, third and fourth order elastic compliance constants (SOECC, TOECC, and FOECC) defined as

$$\begin{aligned}
S_{ijkl}^{S/T}(\mathbf{a}) &\equiv S_{ijkl}^{S/T}(\mathbf{a}; \mathbf{a}) & S_{ijklmn}^{S/T}(\mathbf{a}) &\equiv S_{ijklmn}^{S/T}(\mathbf{a}; \mathbf{a}) & S_{ijklmnpq}^{S/T}(\mathbf{a}) &\equiv S_{ijklmnpq}^{S/T}(\mathbf{a}; \mathbf{a}), & (19) \\
(\text{SOECC}) & & (\text{TOECC}) & & (\text{FOECC}) & &
\end{aligned}$$

which are all evaluated at and referred to the stress-free natural state \mathbf{a} . When the superscript S or T is omitted in the notation of the elastic stiffness and compliance coefficients, it is henceforth understood that they refer to both adiabatic and isothermal processes. The SOESC and SOECC satisfy the full symmetry relations in their subscript indices and can be abbreviated using the Voigt indices α, β , and λ ($\alpha, \beta, \lambda = 1, 2, \dots, 6$). Both SOESC and SOECC matrices, $C_{\alpha\beta}$ and $S_{\alpha\beta}$, are symmetric, i.e., $C_{\alpha\beta} = C_{\beta\alpha}$ and $S_{\alpha\beta} = S_{\beta\alpha}$ and satisfy the reciprocal relations $C_{\alpha\lambda} S_{\lambda\beta} = \delta_{\alpha\beta}$.

2. Nonlinear relations between strains and stresses

The Cauchy stress σ_{kl} and the thermodynamic stress τ_{ij} are related by the Murnaghan's equation

$$\tau_{ij}(\mathbf{X}; \mathbf{a}) = J \beta_{ik} \beta_{jl} \sigma_{kl}(\mathbf{X}) \quad \sigma_{ij}(\mathbf{X}) = J^{-1} \alpha_{ik} \alpha_{jl} \tau_{ij}(\mathbf{X}; \mathbf{a}) \quad (21)$$

where the α and β coefficients are defined in Eqs. 5 and 6, respectively, and J denotes the Jacobian of deformation gradient and is expressed as

$$J(\mathbf{X}; \mathbf{a}) \equiv \det \left[\frac{\partial X_i}{\partial a_j} \right] = \frac{V_{\mathbf{X}}}{V_n} = \frac{\rho_{\mathbf{a}}}{\rho_{\mathbf{X}}} \quad (22)$$

In the above Eq. 22, V and ρ denote the volume and the density of a specimen, respectively.

Now we evaluate $Q_{ijkl}(\mathbf{a}) \equiv (\partial \varepsilon_{ij} / \partial \sigma_{kl})_{\mathbf{a}}$ at the stress-free state. It can be easily shown from Eqs. 7, 21, and 22 that

$$\begin{aligned} (\partial \varepsilon_{ij} / \partial \sigma_{kl})_{\mathbf{a}} &= (\partial \eta_{ij} / \partial \sigma_{kl})_{\mathbf{a}} = (\partial \eta_{ij} / \partial \tau_{rs})_{\mathbf{a}} (\partial \tau_{rs} / \partial \sigma_{kl})_{\mathbf{a}} \\ &= (\partial \eta_{ij} / \partial \tau_{rs})_{\mathbf{a}} \frac{1}{2} (\delta_{rk} \delta_{sl} + \delta_{rl} \delta_{sk}) = S_{ijkl}(\mathbf{a}), \\ Q_{ijkl}(\mathbf{a}) &\equiv (\partial \varepsilon_{ij} / \partial \sigma_{kl})_{\mathbf{a}} = (\partial \eta_{ij} / \partial \tau_{rs})_{\mathbf{a}} = S_{ijkl}(\mathbf{a}), \end{aligned} \quad (23)$$

where $S_{ijkl}(\mathbf{a}) \equiv (\partial \eta_{ij} / \partial \tau_{kl})_{\mathbf{a}}$ denote the second order elastic compliance coefficients (SOECC) evaluated at the natural state. Then, $W_{ijkl}(\mathbf{a})$ defined as $(\partial \omega_{ij} / \partial \sigma_{kl})_{\mathbf{a}}$ is equal to

$$W_{ijkl}(\mathbf{a}) \equiv \left(\frac{\partial \omega_{ij}}{\partial \sigma_{kl}} \right)_{\mathbf{a}} = -\varepsilon_{ij1} S_{23kl}(\mathbf{a}) + \varepsilon_{ij2} S_{13kl}(\mathbf{a}) + \varepsilon_{ij3} S_{12kl}(\mathbf{a}). \quad (24)$$

$F_{ijkl}(\mathbf{a})$ defined as $\left[\frac{\partial}{\partial \sigma_{kl}} \left(\frac{\partial X_i}{\partial a_j} \right) \right]_{\mathbf{a}}$ is equal to

$$F_{ijkl}(\mathbf{a}) \equiv \left[\frac{\partial}{\partial \sigma_{kl}} \left(\frac{\partial x_i}{\partial a_j} \right) \right]_{\mathbf{a}} = \left[\frac{\partial}{\partial \sigma_{kl}} \left(\frac{\partial u_i}{\partial a_j} \right) \right]_{\mathbf{a}} = S_{ijkl}(\mathbf{a}) + W_{ijkl}(\mathbf{a}). \quad (25)$$

Differentiating $(\partial X_i / \partial a_k)(\partial a_k / \partial X_j) = \delta_{ij}$ with respect to σ_{mn} and evaluating the result at the natural state \mathbf{a} , one obtains

$$\left[\frac{\partial}{\partial \sigma_{mn}} \left(\frac{\partial a_i}{\partial X_j} \right) \right]_{\mathbf{a}} = -F_{ijmn}(\mathbf{a}) = - \left[\frac{\partial}{\partial \sigma_{mn}} \left(\frac{\partial X_i}{\partial a_j} \right) \right]_{\mathbf{a}} \quad (26)$$

The deformation gradients can be expanded in Taylor series to the first power in Cauchy stress as

$$\alpha_{ij} = \frac{\partial X_i}{\partial a_j} = \delta_{ij} + F_{ijkl} \sigma_{kl} + \dots = \delta_{ij} + (S_{ijkl} + W_{ijkl}) \sigma_{kl} + \dots \quad (27)$$

$$\beta_{ij} = \frac{\partial a_i}{\partial X_j} = \delta_{ij} - F_{ijkl} \sigma_{kl} + \dots = \delta_{ij} - (S_{ijkl} + W_{ijkl}) \sigma_{kl} + \dots .$$

In Eq. 27 and henceforth, the evaluation state \mathbf{a} is dropped in the notation of elastic modulus coefficients $S_{ijkl\dots}$, $W_{ijkl\dots}$, and $F_{ijkl\dots}$, unless otherwise specified. A Lagrangian strain η_{ij} can be expanded in Taylor series with powers of the Cauchy stress.

$$\eta_{ij} = S_{ijkl} \tau_{kl} + (1/2) S_{ijklmn} \tau_{kl} \tau_{mn} + \dots = S_{ijkl} J \beta_{kp} \beta_{lq} \sigma_{pq} + (1/2) S_{ijklmn} J^2 \beta_{kp} \beta_{lq} \beta_{mrp} \beta_{nsp} \sigma_{pq} \sigma_{rs} \dots ,$$

where S_{ijklmn} are the third order elastic compliance constants (TOESC). Substituting Eq. (27₂) into the above equation yields

$$\eta_{ij} = S_{ijkl} \sigma_{kl} + \left(S_{ijkl} S_{hhmn} - 2S_{ijkh} F_{hlmn} + \frac{1}{2} S_{ijklmn} \right) \sigma_{kl} \sigma_{mn} + \dots ,$$

which can be written as

$$\eta_{ij} = S_{ijkl} \sigma_{kl} + [ijklmn]_{\omega} \sigma_{kl} \sigma_{mn} \dots , \quad (28)$$

where

$$[ijklmn]_{\omega} \equiv S_{ijkl} S_{hhmn} - 2S_{ijkh} F_{hlmn} + (1/2) S_{ijklmn} . \quad (29)$$

Let's turn our attention to evaluating the second derivative $Q_{ijklmn} \equiv \frac{\partial^2 \varepsilon_{ij}}{\partial \sigma_{mn} \partial \sigma_{kl}}$ at the natural state

a. First, note the following identities at the natural state:

$$\begin{aligned} (\partial \tau_{rs} / \partial \sigma_{mn})_{\mathbf{a}} &= (1/2)(\delta_{rm} \delta_{sn} + \delta_{rn} \delta_{sm}) \\ (\partial \sigma_{ab} / \partial \tau_{rs})_{\mathbf{a}} &= (1/2)(\delta_{ar} \delta_{bs} + \delta_{as} \delta_{br}). \end{aligned} \quad (30)$$

Making use of Eqs. (30) and (23), a stress derivative of the Jacobian defined in Eq. 22 at the natural state is readily found to be

$$\left(\frac{\partial J}{\partial \sigma_{kl}} \right)_{\mathbf{a}} = \left(\frac{\partial J}{\partial \eta_{gh}} \frac{\partial \eta_{gh}}{\partial \tau_{rs}} \frac{\partial \tau_{rs}}{\partial \sigma_{kl}} \right)_{\mathbf{a}} = \left(J G^{-1}_{hg} S_{ghrs} \frac{\partial \tau_{rs}}{\partial \sigma_{kl}} \right)_{\mathbf{a}} = S_{hhkl}(\mathbf{a}), \quad (31)$$

where G^{-1}_{hg} is the inverse of $G_{hg} = G_{gh} = \frac{\partial x_k}{\partial a_g} \frac{\partial x_k}{\partial a_h}$, the Cauchy-Green tensor defined in Eq. 8.

Recalling Eq. 7, one finds

$$\begin{aligned} Q_{ijkl} &\equiv \frac{\partial \varepsilon_{ij}}{\partial \sigma_{kl}} = \frac{\partial \eta_{ij}}{\partial \tau_{pq}} \frac{\partial \tau_{pq}}{\partial \sigma_{kl}} - \frac{1}{2} \left(\frac{\partial u_{pi}}{\partial \sigma_{kl}} u_{pj} + u_{pi} \frac{\partial u_{pj}}{\partial \sigma_{kl}} \right) \\ &= S_{ijpq}(\mathbf{x}) \frac{\partial}{\partial \sigma_{kl}} (J \beta_{pa} \beta_{qb} \sigma_{ab}) - \frac{1}{2} \left(\frac{\partial u_{pi}}{\partial \sigma_{kl}} u_{pj} + u_{pi} \frac{\partial u_{pj}}{\partial \sigma_{kl}} \right). \end{aligned}$$

Our purpose lies in evaluating Q_{ijklmn} at the natural state, where $\mathbf{u} = 0$. Neglecting the terms containing u_{pi} and u_{pj} terms, Q_{ijklmn} can be written as

$$Q_{ijklmn} \equiv \frac{\partial^2 \varepsilon_{ij}}{\partial \sigma_{mn} \partial \sigma_{kl}} = \frac{\partial \tau_{rs}}{\partial \sigma_{mn}} \frac{\partial}{\partial \tau_{rs}} \left[S_{ijpq} \sigma_{ab} (\partial / \partial \sigma_{kl}) (J \beta_{pa} \beta_{qb}) + J \beta_{pa} \beta_{qb} (1/2) (\delta_{ka} \delta_{lb} + \delta_{kb} \delta_{la}) \right]$$

$$- \frac{1}{2} \left(\frac{\partial u_{pi}}{\partial \sigma_{kl}} \frac{\partial u_{pj}}{\partial \sigma_{mn}} + \frac{\partial u_{pi}}{\partial \sigma_{mn}} \frac{\partial u_{pj}}{\partial \sigma_{kl}} \right).$$

With the help of Eqs. (30) and (31), the above equation evaluated at the natural state \mathbf{a} is after some algebra expressed as

$$Q_{ijklmn}(\mathbf{a}) \equiv \left(\frac{\partial^2 \varepsilon_{ij}}{\partial \sigma_{kl} \partial \sigma_{mn}} \right)_{\mathbf{a}} = S_{ijkl} S_{hhmn} + S_{ijmn} S_{hhkl} - (S_{ijmh} F_{hnkl} + S_{ijnh} F_{hmkl})$$

$$- (S_{ijkh} F_{hlmn} + S_{ijlh} F_{hkmn}) - (1/2) (F_{pikl} F_{pjmn} + F_{pimn} F_{pjkl}) + S_{ijklmn},$$

which, when used in multiplication with the product $\sigma_{kl} \sigma_{mn}$, is simplified to

$$Q_{ijklmn}(\mathbf{a}) \equiv \left(\frac{\partial^2 \varepsilon_{ij}}{\partial \sigma_{kl} \partial \sigma_{mn}} \right)_{\mathbf{a}} = 2S_{ijkl} S_{hhmn} - 4S_{ijkh} F_{hlmn} - F_{hikl} F_{hjmn} + S_{ijklmn}. \quad (32)$$

ε_{ij} can be expanded as

$$\varepsilon_{ij} = S_{ijkl} \sigma_{kl} + (1/2) Q_{ijklmn} \sigma_{kl} \sigma_{mn} + \dots \quad (33)$$

It is readily shown that

$$W_{ijklmn} \equiv \left(\frac{\partial^2 \omega_{ij}}{\partial \sigma_{kl} \partial \sigma_{mn}} \right)_{\mathbf{a}} = -\varepsilon_{ij1} Q_{23klmn}(\mathbf{a}) + \varepsilon_{ij2} Q_{31klmn}(\mathbf{a}) + \varepsilon_{ij3} Q_{12klmn}(\mathbf{a}), \quad (34)$$

$$F_{ijklmn} \equiv \left[\frac{\partial^2}{\partial \sigma_{kl} \partial \sigma_{mn}} \left(\frac{\partial x_i}{\partial a_j} \right) \right]_{\mathbf{a}} = \left[\frac{\partial^2}{\partial \sigma_{kl} \partial \sigma_{mn}} \left(\frac{\partial u_i}{\partial a_j} \right) \right]_{\mathbf{a}} = Q_{ijklmn}(\mathbf{a}) + W_{ijklmn}(\mathbf{a}). \quad (35)$$

ω_{ij} , $\partial u_i / \partial a_j$, and $\partial X_i / \partial a_j$ are now expanded in Taylor series to the second power in stress.

$$\omega_{ij} = W_{ijkl} \sigma_{kl} + (1/2) W_{ijklmn} \sigma_{kl} \sigma_{mn} + \dots, \quad (36)$$

$$\partial u_i / \partial a_j = F_{ijkl} \sigma_{kl} + (1/2) F_{ijklmn} \sigma_{kl} \sigma_{mn} + \dots, \quad (37)$$

$$\partial X_i / \partial a_j = \delta_{ij} + F_{ijkl} \sigma_{kl} + (1/2) F_{ijklmn} \sigma_{kl} \sigma_{mn} + \dots. \quad (38)$$

Note that for materials of orthorhombic (orthotropic) or higher symmetry,

$$S_{12kk} = S_{kk12} = S_{23kk} = S_{kk23} = S_{13kk} = S_{kk13} = 0 \quad (k \text{ fixed; } k = 1, 2, \text{ or } 3). \quad (39)$$

If their directions of material symmetry coincide with those of three principal stresses, i.e., $\sigma_{kl} = \sigma_k \delta_{kl}$ (k fixed and $k = 1, 2, 3$), it follows from Eq. 24 that

$$W_{ijkl} = -\varepsilon_{ij1} S_{23kk}(\mathbf{a}) + \varepsilon_{ij2} S_{13kk}(\mathbf{a}) + \varepsilon_{ij3} S_{12kk}(\mathbf{a}) = 0 \quad (k \text{ fixed}),$$

$$W_{ijkl} = W_{ijklmn} = W_{ijklmn..} = 0, \quad (40)$$

which indicates that no rotation is involved, i.e.,

$$\omega_{ij} = 0 \quad (\text{no rotation}) \quad (41)$$

$$F_{ijkl} = S_{ijkl} \quad F_{ijklmn} = Q_{ijklmn} = 2S_{ijkl}S_{hhmn} - 4S_{ijkh}S_{hlmn} - S_{hikl}S_{hjmn} + S_{ijklmn} \quad (42)$$

However, when the directions of acting principal stresses deviate from those of material symmetry of a solid with orthorhombic/orthotropic or higher symmetry at the stress-free natural state, the solid medium possesses in a general case triclinic or monotropic symmetry, which has the non-vanishing compliances $S_{12kk} \neq 0$, $S_{23kk} \neq 0$, and $S_{13kk} \neq 0$ (k fixed; $k = 1, 2$, or 3). Therefore, the rotation tensors do not vanish in general and Eqs. 40-42 are not valid. The exceptional case is an initially isotropic solid that possesses under general triaxial stresses an orthotropic symmetry, the directions of which always coincide with those of the applied principal stresses. Under hydrostatic pressures a material preserves its initial symmetry at the stress-free state.

Twice differentiating $\alpha_{ih}\beta_{hj} = \delta_{ij}$ with respect to σ_{kl} and evaluating at the natural state, one obtains

$$\beta_{ij} \equiv \frac{\partial \alpha_i}{\partial x_j} = \delta_{ij} - F_{ijkl}\sigma_{kl} + (1/2)(F_{ihkl}F_{hjmn} + F_{ihmn}F_{hjk l} - F_{ijklmn})\sigma_{kl}\sigma_{mn} + \dots \quad (43)$$

We also expand the Jacobian J and its inverse J^{-1} to the power of two in Cauchy stress. For this purpose, use is made of J and J^2 written in terms of Lagrangian strains as

$$J = 1 + \eta_{aa} + (1/2)\eta_{aa}\eta_{bb} - \eta_{ab}\eta_{ab} + \dots \quad (44)$$

$$J^2 = 1 + 2\eta_{aa} + 2\eta_{aa}\eta_{bb} - 2\eta_{ab}\eta_{ab} + \dots,$$

which on substitution of Eqs. 28 and 29 yield J as

$$J = 1 + S_{hhkl}\sigma_{kl} + \left(\frac{3}{2}S_{ggkl}S_{hhmn} - 2S_{hhkg}F_{glmn} - S_{ghkl}S_{ghmn} + \frac{1}{2}S_{hhklmn} \right) \sigma_{kl}\sigma_{mn} + \dots \quad (45)$$

Differentiating $JJ^{-1} = 1$ once and twice with respect to Cauchy stress and evaluating each time at the natural state, one finds

$$J^{-1} = 1 - S_{hhkl} \sigma_{kl} + \left(2S_{hhkg} F_{glmn} + S_{ghkl} S_{ghmn} - \frac{1}{2} S_{ggkl} S_{hhmn} - \frac{1}{2} S_{hhklmn} \right) \sigma_{kl} \sigma_{mn} + \dots \quad (46)$$

Finally, one can find an expression for Lagrangian strain η_{ij} , which includes the contribution of the cubic power in Cauchy stress and term containing the fourth order elastic compliance constants (FOECC) $S_{ijklmnpq}$. Beginning with

$$\begin{aligned} \eta_{ij} &= S_{ijkl} \tau_{kl} + \frac{1}{2} S_{ijklmn} \tau_{kl} \tau_{mn} + \frac{1}{6} S_{ijklmnpq} \tau_{kl} \tau_{mn} \tau_{pq} + \dots \\ &= S_{ijkl} J \beta_{kr} \beta_{ls} \sigma_{rs} + \frac{1}{2} S_{ijklmn} \beta_{kr} \beta_{ls} \beta_{mt} \beta_{nu} \sigma_{rs} \sigma_{tu} + \frac{1}{6} S_{ijklmnpq} \beta_{kr} \beta_{ls} \beta_{mt} \beta_{nu} \beta_{pv} \beta_{qw} \sigma_{rs} \sigma_{tu} \sigma_{vw} + \dots, \end{aligned}$$

and making use of Eqs. 43-45 in the above equation, one obtains after a straightforward but involved algebra

$$\eta_{ij} = S_{ijkl} \sigma_{kl} + [ijklmn]_{\omega} \sigma_{kl} \sigma_{mn} + [ijklmnpq]_{\omega} \sigma_{kl} \sigma_{mn} \sigma_{pq} + \dots, \quad (47)$$

where $[ijklmn]_{\omega}$ is defined in Eq. 29 and

$$\begin{aligned} [ijklmnpq]_{\omega} &= S_{ijkl} \left(\frac{3}{2} S_{ggmn} S_{hhpq} - 2S_{ggmh} F_{hnpq} - S_{ghmn} S_{ghpq} + \frac{1}{2} S_{hhmnpq} \right) \\ &\quad + S_{ijkq} \left(2F_{ghmn} F_{hlpq} - 2S_{hhmn} F_{glpq} - F_{glmnpq} \right) + S_{ijgh} F_{gkmn} F_{hlpq} \cdot \\ &\quad + S_{hhpq} S_{ijklmn} - 2F_{hkpq} S_{ijhlmn} + \frac{1}{6} S_{ijklmnpq} \end{aligned} \quad (48)$$

From Eqs. 7, 37 and 48, a similar expression for ε_{ij} is readily shown to be

$$\begin{aligned} \varepsilon_{ij} = & S_{ijkl}\sigma_{kl} + \left([ijklmn]_{\omega} - \frac{1}{2}F_{hikl}F_{hjmn} \right) \sigma_{kl}\sigma_{mn} \\ & + \left([ijklmnpq]_{\omega} - \frac{1}{4}F_{hikl}F_{hjmn} - \frac{1}{4}F_{hikl}F_{himnpq} \right) \sigma_{kl}\sigma_{mn}\sigma_{pq} + \dots \end{aligned} \quad (49)$$

The expressions for $\partial u_i/\partial a_j$ and $\partial x_i/\partial a_j$ in terms of the Cauchy stress to the power of three are obtained by substituting Eq. 40 into the following relations.

$$\begin{aligned} \partial u_i/\partial a_j = & \varepsilon_{ij} + \omega_{ij} = \varepsilon_{ij} - \varepsilon_{ij1}\varepsilon_{23} + \varepsilon_{ij2}\varepsilon_{13} + \varepsilon_{ij3}\varepsilon_{12}, \\ \partial X_i/\partial a_j = & \delta_{ij} + u_{ij} = \delta_{ij} + \varepsilon_{ij} - \varepsilon_{ij1}\varepsilon_{23} + \varepsilon_{ij2}\varepsilon_{13} + \varepsilon_{ij3}\varepsilon_{12}. \end{aligned}$$

When the directions of material symmetry of which coincide with those of three principal stress directions, there occurs no rotation ($\omega_{ij} = 0$), and

$$\begin{aligned} u_{ij} = u_{ji} \quad \varepsilon_{ij} = u_{ij} = \partial u_i/\partial a_j \\ \lambda_i \delta_{ij} \equiv \frac{\partial x_i}{\partial a_j} = \delta_{ij}(1 + \varepsilon_{ij}) \quad (i \text{ fixed}) \quad \lambda_i = 1 + \varepsilon_{ii} \quad (i \text{ fixed}). \end{aligned} \quad (50)$$

λ_i above is called the principal stretch. With a solid medium of no rotation involved, both F_{ijkl} and F_{ijklmn} will be respectively replaced by S_{ijkl} and Q_{ijklmn} (see Eq. 42) and the subscript ω will henceforth be dropped in the notation of $[ijklmn]_{\omega}$ and $[ijklmnpq]_{\omega}$ appearing in Eqs. 28, 29, and 47-49. They reduce to

$$[ijklmn] \equiv S_{ijkl}S_{hlmn} - 2S_{ijkh}S_{hlmn} + (1/2)S_{ijklmn} \quad (51)$$

$$\begin{aligned}
[ijklmnpq] = & S_{ijkl} \left(\frac{3}{2} S_{ggmn} S_{hhpq} - 2 S_{ggmh} S_{hnpq} - S_{ghmn} S_{ghpq} + \frac{1}{2} S_{hhmnpq} \right) \\
& + S_{ijkq} \left(4 S_{glmh} S_{hnpq} - 4 S_{glmn} S_{hnpq} + 3 S_{ghmn} S_{hlpq} + S_{glmnpq} \right) . \\
& + S_{ijgh} S_{gkmn} S_{hlpq} + S_{hhpq} S_{ijklmn} - 2 S_{hlpq} S_{ijkhmn} + \frac{1}{6} S_{ijklmnpq}
\end{aligned} \tag{52}$$

η_{ij} , ε_{ij} , and λ_i in the absence of rotation become

$$\eta_{ij} = S_{ijkl} \sigma_{kl} + [ijklmn] \sigma_{kl} \sigma_{mn} + [ijklmnpq] \sigma_{kl} \sigma_{mn} \sigma_{pq} + \dots \tag{53}$$

$$\begin{aligned}
\varepsilon_{ij} = & S_{ijkl} \sigma_{kl} + \left([ijklmn] - \frac{1}{2} S_{hikl} S_{hjmn} \right) \sigma_{kl} \sigma_{mn} \\
& + \left([ijklmnpq] - \frac{1}{4} S_{hikl} Q_{hjmn} - \frac{1}{4} S_{hikl} Q_{himnpq} \right) \sigma_{kl} \sigma_{mn} \sigma_{pq} + \dots
\end{aligned} \tag{54}$$

$$\begin{aligned}
\lambda_i = 1 + \varepsilon_{ii} = & S_{iikl} \sigma_{kl} + \left([iiklmn] - \frac{1}{2} S_{hikl} S_{himn} \right) \sigma_{kl} \sigma_{mn} \\
& + \left([iiklmnpq] - \frac{1}{4} S_{hikl} Q_{himnpq} - \frac{1}{4} S_{hikl} Q_{himnpq} \right) \sigma_{kl} \sigma_{mn} \sigma_{pq} + \dots \quad (i \text{ fixed})
\end{aligned} \tag{55}$$

$$\lambda_i^2 = 1 + 2\eta_{ii} = 1 + S_{iikl} \sigma_{kl} + 2[iijklmn] \sigma_{kl} \sigma_{mn} + 2[iijklmnpq] \sigma_{kl} \sigma_{mn} \sigma_{pq} + \dots \quad (i \text{ fixed}), \tag{56}$$

where Q_{ijklmn} is given in Eq. 42b.

Let $H_{ik} \equiv (\partial a_i / \partial X_j) (\partial a_k / \partial X_j)$ represent the inverse of the Cauchy-Green tensor G_{ik} defined in Eq. 8 such that $G_{ik} H_{kj} = \delta_{ij}$. It is shown [10] that stretch $\lambda_{\mathbf{N}}$ and thickness shrink $f_{\mathbf{N}}$ in the direction \mathbf{N} (unit vector) at the natural stress-free state are given by

$$\lambda_{\mathbf{N}}^2 = G_{ik} N_i N_k \quad f_{\mathbf{N}}^2 = H_{ik} N_i N_k . \tag{57}$$

The angle θ_N between the current direction of the natural normal \mathbf{N} to a plane at the natural state and the current normal to the plane at the deformed state is

$$\cos \theta_N = 1/(\lambda_N f_N), \quad (58)$$

and the ratio of the planar separations of the deformed and natural configurations is

$$L_x/L_0 = 1/f_N. \quad (59)$$

The G_{ik} and H_{ik} have the same eigenvectors. If \mathbf{N} is an eigenvector of G_{ik} , then λ_N^2 is the corresponding eigenvalue equal to Eq. (50) and f_N^2 is its reciprocal. In this case the rotation angle θ_N is zero as already mentioned.

1. Nonlinear relations of thermodynamic and effective elastic coefficients to stresses

In the previous chapter, we described various nonlinear relations sufficiently necessary to derive the nonlinear relations of thermodynamic and effective elastic coefficients to stresses in this chapter. The derived equations for thermodynamic and effective elastic coefficients are expanded to the power of two in Cauchy stress and include the second, third, and fourth order elastic coefficients. The expansion to the higher than the power of two in Cauchy stress involves the contribution of the fifth order elastic constants, which are far beyond the capacity of the present art of precision measurement techniques and is therefore not attempted in this paper. For a clear distinction between thermodynamic and effective elastic coefficients, readers are referred to Refs. 9 and 12.

The effective elastic coefficients defined at a deformed state \mathbf{X} , $K_{ijkl}(\mathbf{X}) \equiv (\partial \sigma_{ij} / \partial \varepsilon_{ij})_{\mathbf{X}}$ and $Q_{ijkl}(\mathbf{X}) \equiv (\partial \varepsilon_{ij} / \partial \sigma_{kl})_{\mathbf{X}}$, indicate a measure of material strength at the very stressed state. $K_{ijkl}(\mathbf{X})$ is related to the thermodynamic elastic stiffness coefficient $C_{ijkl}^{S/T}(\mathbf{X})$, defined in Eqs. 14 and 15 by replacing the evaluation state \mathbf{x} by \mathbf{X} , by [9]

$$K_{ijkl}(\mathbf{X}) = C_{ijkl}(\mathbf{X}) - \sigma_{ij}(\mathbf{X})\delta_{kl} + (1/2)[\sigma_{ik}(\mathbf{X})\delta_{jl} + \sigma_{il}(\mathbf{X})\delta_{jk} + \sigma_{jk}(\mathbf{X})\delta_{il} + \sigma_{jl}(\mathbf{X})\delta_{ik}] \quad (60)$$

where δ denotes a Kronecker delta. The difference between K_{ijkl}^S and K_{ijkl}^T , C_{ijkl}^S and C_{ijkl}^T , and Q_{ijkl}^S and Q_{ijkl}^T is discussed in Ref. 12. As aforementioned, the full symmetry relations $C_{ijkl} = C_{jikl} = C_{ijlk} = C_{klij}$ is maintained in the thermodynamic elastic stiffness coefficients $C_{ijkl}(\mathbf{X})$. However, the full symmetry is lost in the effective elastic stiffness coefficients $K_{ijkl}(\mathbf{X})$ and $Q_{ijkl}(\mathbf{X})$ unless the stress acting on the specimen is hydrostatic, i.e., $\sigma_{ij} = s\delta_{ij}$, where s is a positive or negative scalar variable. K_{ijkl} coefficients obey the relations $K_{ijkl} = K_{jikl} = K_{ijlk}$ along with

$$K_{ijkl} - K_{klij} = \delta_{ij}\sigma_{kl} - \delta_{kl}\sigma_{ij}, \quad (61)$$

which is known as the Huang's conditions. When K_{ijkl} and Q_{ijkl} are written in the matrix form using the abbreviated Voigt indices α, β , and γ ($\alpha, \beta, \gamma = 1, 2, \dots, 6$), they satisfy the reciprocal relation $K_{\alpha\gamma}Q_{\gamma\beta} = \delta_{\alpha\beta}$ or $[Q_{\alpha\beta}] = [K_{\alpha\beta}]^{-1}$. The Huang's conditions impose ten constraints on the coefficients of $K_{\alpha\beta}$ and $Q_{\alpha\beta}$, which result in general 26 independent coefficients. It is readily shown from Eq. 60 that

$$[K_{\alpha\beta}] = \begin{bmatrix} C_{11} + \sigma_1 & C_{12} - \sigma_1 & C_{13} - \sigma_1 & C_{14} & C_{15} + \sigma_5 & C_{16} + \sigma_6 \\ C_{11} - \sigma_2 & C_{22} + \sigma_2 & C_{23} - \sigma_2 & C_{24} + \sigma_4 & C_{25} & C_{26} + \sigma_6 \\ C_{13} - \sigma_3 & C_{23} - \sigma_3 & C_{33} + \sigma_3 & C_{34} + \sigma_4 & C_{35} + \sigma_5 & C_{36} \\ C_{14} - \sigma_4 & C_{24} & C_{34} & C_{44} + (\sigma_2 + \sigma_3)/2 & C_{45} + \sigma_6/2 & C_{46} + \sigma_5/2 \\ C_{15} & C_{11} + \sigma & C_{35} & C_{45} + \sigma_6/2 & C_{44} + (\sigma_2 + \sigma_3)/2 & C_{56} + \sigma_4/2 \\ C_{16} & C_{26} & C_{36} - \sigma_6 & C_{46} + \sigma_5/2 & C_{56} + \sigma_4/2 & C_{44} + (\sigma_2 + \sigma_3)/2 \end{bmatrix} \quad (62)$$

where all the $K_{\alpha\beta}$ elements are evaluated at the stressed state \mathbf{X} . The *in situ* isothermal material strengths $K_{\alpha\beta}^T$ and $Q_{\alpha\beta}^T$ in an arbitrarily stressed state can be measured by very slow static

tension/compression and torsion tests, while $K_{\alpha\beta}^S$ and $Q_{\alpha\beta}^S$ can be obtained from the ultrasonic wavespeeds measurements [21].

We are first interested in finding expressions for $K_{ijkl}(\mathbf{X})$ and as a function of the Cauchy stress using the SOESC, TOESC and FOESC. Eqs. 60-61 indicate that at first we derive expressions for $C_{ijkl}(\mathbf{X})$. The thermodynamic elastic stiffness coefficient $C_{ijkl}(\mathbf{X})$ evaluated at and referred to the deformed state \mathbf{X} is written as [10]

$$\begin{aligned}
 C_{ijkl}(\mathbf{X}) &= \frac{1}{J} \frac{\partial X_i}{\partial a_m} \frac{\partial X_j}{\partial a_n} \frac{\partial X_k}{\partial a_p} \frac{\partial X_l}{\partial a_q} C_{mnpq}(\mathbf{X}; \mathbf{a}) \\
 &= \frac{1}{J} \frac{\partial X_i}{\partial a_m} \frac{\partial X_j}{\partial a_n} \frac{\partial X_k}{\partial a_p} \frac{\partial X_l}{\partial a_q} \left[C_{mnpq}(\mathbf{a}) + C_{mnpqrs}(\mathbf{a}) \eta_{rs}(\mathbf{X}; \mathbf{a}) + \frac{1}{2} C_{mnpqrstuv}(\mathbf{a}) \eta_{rs}(\mathbf{X}; \mathbf{a}) \eta_{tu}(\mathbf{X}; \mathbf{a}) \right] \quad (63)
 \end{aligned}$$

where $C_{mnpq}(\mathbf{X}; \mathbf{a})$ is a thermodynamic elastic stiffness coefficient evaluated at \mathbf{X} and referred to \mathbf{a} and $C_{mnpq}(\mathbf{a})$, $C_{mnpqrs}(\mathbf{a})$, and $C_{mnpqrstuv}(\mathbf{a})$ are respectively the second, third and fourth order elastic stiffness constants (SOESC, TOESC, and FOESC), all evaluated at and referred to the natural state \mathbf{a} . It is sufficient to make use of Eqs. 28, 38, and 46 to expand $C_{ijkl}(\mathbf{X})$ to the power of two in Cauchy stress. Substituting these relations into Eq. 61, one obtains after lengthy involved algebra

$$\begin{aligned}
C_{ijkl}(\mathbf{X}) = & C_{ijkl} + (C_{mjkl}F_{imab} + C_{inlk}F_{jnab} + C_{ijpl}F_{kpab} + C_{ijkq}F_{lqab} - C_{ijkl}S_{hhab} + C_{mjklmn}S_{mnab})\sigma_{ab} \\
& + \left[\begin{aligned}
& C_{mnkl}F_{imab}F_{jnab} + C_{mjpl}F_{imab}F_{kpab} + C_{mjkq}F_{imab}F_{lqab} + C_{inpl}F_{jnab}F_{kpab} \\
& + C_{inkq}F_{jnab}F_{lqab} + C_{ijpq}F_{kpab}F_{lqab} \\
& - S_{hhab} \left(C_{mjkl}F_{imcd} + C_{inlk}F_{jnab} + C_{ijpl}F_{kpab} + C_{ijkq}F_{lqab} \right) \\
& + C_{ijkl} \left(2S_{hhag}F_{gbcd} + 2S_{ghab}S_{ghcd} - \frac{1}{2}S_{ggab}F_{hhcd} - \frac{1}{2}S_{hhabcd} \right) \\
& + S_{rsab} \left(C_{mjklrs}F_{imcd} + C_{inlkrs}F_{jnab} + C_{ijplrs}F_{kpab} + C_{ijkqrs}F_{lqab} \right) \\
& + \frac{1}{2} \left(C_{mjkl}F_{imabcd} + C_{inlk}F_{jnabcd} + C_{ijpl}F_{kpabcd} + C_{ijkq}F_{lqabcd} \right) \\
& + C_{ijklmn} \left(\frac{1}{2}S_{mnabcd} - 2S_{mna}F_{hbcd} \right) + \frac{1}{2}C_{ijklmnpq}S_{mnab}S_{pqcd}
\end{aligned} \right] \sigma_{ab}\sigma_{cd} + \dots, \tag{64}
\end{aligned}$$

where all the elements except the Cauchy stress quantities at the right side of the above equation are evaluated at the natural state \mathbf{a} .

When the directions of acting principal stresses align with those of material symmetry of solids with orthotropic or higher symmetry, there is no rotation involved and Eqs. 39 and 42 hold valid. Using $\sigma_{ij} = \sigma_i\delta_{ij}$ (i fixed) and substituting Eqs. 39 and 42 into Eq. (62) result in

$$\begin{aligned}
C_{ijkl}(\mathbf{X}) = & C_{ijkl} + [C_{ijkl}(S_{iiaa} + S_{jja} + S_{kkaa} + S_{llaa} - S_{hhaa}) + C_{ijklmn}S_{mmaa}] \sigma_a - \\
& + \left[\begin{aligned}
& C_{ijkl} \left(S_{iiaa}S_{jjbb} + S_{iiaa}S_{kkbb} + S_{iiaa}S_{llbb} + S_{jja}S_{kkbb} + S_{jja}S_{llbb} + S_{kkaa}S_{llbb} \right) \\
& (1/2) \left(S_{iiaa}S_{iibb} + S_{jja}S_{jjbb} + S_{kkaa}S_{kkbb} + S_{llaa}S_{llbb} \right) + \\
& C_{ijkl}S_{hhaa} \left(2S_{aabb} + S_{hhbb} - \frac{1}{2}S_{ggbb} \right) + \\
& \left(C_{ijklmn}S_{mmaa} - 2C_{ijkl}S_{aabb} \right) \left(S_{iibb} + S_{jjbb} + S_{kkbb} + S_{llbb} \right) + \\
& (1/2) \left(S_{iiaabb} + S_{jjaabb} + S_{kkaabb} + S_{llaabb} - S_{hhaabb} \right) + \\
& C_{ijklmn} \left(\frac{1}{2}S_{mmaabb} - 2S_{mmaa}S_{aabb} \right) + (1/2)C_{ijklmnp}S_{mmaa}S_{ppbb}
\end{aligned} \right] \sigma_a\sigma_b \tag{65}
\end{aligned}$$

where σ_a and σ_b ($a, b = 1, 2, 3$) represent the principal stresses aligned in the directions of material symmetry.

In the case of uniaxial loading along the 3 direction, $\sigma_a = \sigma_3 \delta_{3a}$ and $\sigma_b = \sigma_3 \delta_{3b}$. As a result, Eq. 65 reduces to

$$C_{ijkl}(\mathbf{X}) = C_{ijkl} + [C_{ijkl}(S_{i3} + S_{j3} + S_{k3} + S_{l3} - S_{13} - S_{23} - S_{33}) + C_{ijkl1}S_{13} + C_{ijkl2}S_{23} + C_{ijkl3}S_{33}] \sigma_3$$

$$+ \left[\begin{aligned} & C_{ijkl}(S_{i3}S_{j3} + S_{is}S_{k3} + S_{is}S_{l3} + S_{js}S_{k3} + S_{js}S_{l3} + S_{ks}S_{l3}) - \frac{1}{2}C_{ijkl}(S_{i3}^2 + S_{j3}^2 + S_{k3}^2 + S_{l3}^2) + \\ & (1/2)C_{ijkl}(S_{13} + S_{23} + S_{33})^2 + 2C_{ijkl}(S_{33}^2 - 2S_{13}S_{23}) + \\ & (C_{ijkl1}S_{13} + C_{ijkl2}S_{23} + C_{ijkl3}S_{33} - 2S_{33}C_{ijkl})(S_{i3} + S_{j3} + S_{k3} + S_{l3}) + \\ & (1/2)C_{ijkl}(S_{i33} + S_{j33} + S_{k33} + S_{l33} - S_{133} - S_{233} - S_{333}) + \\ & (1/2)(C_{ijkl1}S_{133} + C_{ijkl2}S_{233} + C_{ijkl3}S_{333}) - 2S_{33}(S_{13}C_{ijkl1} + S_{23}C_{ijkl2} + S_{33}C_{ijkl3}) + \\ & (1/2) \left(S_{13}^2 C_{ijkl111} + S_{23}^2 C_{ijkl222} + S_{33}^2 C_{ijkl333} + 2S_{13}S_{23}C_{ijkl122} + 2S_{13}S_{33}C_{ijkl133} + \right. \\ & \left. S_{23}S_{33}C_{ijkl233} \right) \end{aligned} \right] \sigma_3^2 \quad (66)$$

The above equation can be expressed in terms of the first Piola-Kirchhoff stress π_3 , which refers to the original area at the natural state. σ_3 is related to π_3 by

$$\sigma_3 = \pi_3 - (S_{13} + S_{23})\pi_3^2, \quad (67)$$

on which may be substituted into Eq. (66) to write it in terms of π_3 . In an ordinary uniaxial loading an applied force is conveniently measured by a force transducer or a load cell. In absence of the dimensional measurements of the cross-sectional area, π_3 serves as a more useful quantity.

Under hydrostatic pressures $\sigma_{ij} = -p\delta_{ij}$, the pressure dependence of $C_{ijkl}(\mathbf{X})$ is found by replacing σ_a and $\sigma_a\sigma_b$ by $-p$ and p^2 in Eq. (65), respectively.

Eqs. (64)-(66) can be combined into Eq. 60 or Eq. 62 to yield $K_{\alpha\beta}(\mathbf{X})$ as a function of stress. Then, $Q_{\alpha\beta}(\mathbf{X})$ is taken equal to its inverse, and the corresponding $Q_{ijkl}(\mathbf{X})$ elements are utilized

to obtain the expressions for the Young's modulus, Poisson's ratio, and bulk modulus (compressibility) as a function of stress. However, from Eq. 49 one finds directly the relation

$$Q_{ijkl}(\mathbf{X}) \equiv \left(\frac{\partial \varepsilon_{ij}}{\partial \sigma_{kl}} \right)_{\mathbf{X}} = (2S_{ijkl}S_{hhmn} - 4S_{ijkh}F_{hlmn} - F_{hikl}F_{hjmn} + S_{ijklmn})\sigma_{mn} + [3[ijklmnpq]_{\omega} - (3/4)F_{hikl}F_{hjmn} - (3/4)F_{hikl}F_{himnpq}]\sigma_n\sigma_q + \dots \quad (68)$$

Conclusions

The author of this article stated in Ref. 22 that $Q_{ijkl}(\mathbf{X})$ of Eq. (68) indicates the measure of the material strength at finitely deformed state and in Ref. 23 he derived succinct formulas for nonlinear higher order stiffness coefficients at finite deformation.

Declaration of Generative AI and AI-assisted technologies in the writing process

This work was prepared by AI based on the author's published research. The author has reviewed and edited the content as needed and takes full responsibility for the content of the publication.

References

1. F.D. MURNAGHAN, *Am. J. Math.* **59** (1937) 235.
2. *Idem.*, “Finite Deformation of an Elastic Solid” (Dover, New York, 1951).
3. F. BIRCH, *Phys. Rev.* **71** (1947) 809.
4. *Idem.*, *J. Geophys. Res.* **57** (1952) 227.
5. K.BRUGGER, *Phys. Rev.* **133** (1964) A1611.
6. R.N.THURSTON AND K. BRUGGER, *ibid.* **133** (1964) A1604.
7. R.F.S. HEARMON, in “Numerical Data and Functional Relationships in Science and Technology,” edited by K.-H. Hellwege and A.M. Hellwege (Springer-Verlag, New York, 1979), Landolt- Börnstein , New Series, Group III, Vol. 11.
8. Every, A.G. and McCurdy, A.K. (2010), in *Numerical Data and Functional Relationships in Science and Technology*, Landolt-Börnstein, New Series, Group III, Vol. 29a, Editor: D.F. Nelson (Springer-Verlag, New York). Tables 9-11.
9. R.N. THURSTON, *J. Acoust. Soc. Am.* **37** (1965) 348.
10. R.N. THURSTON, in “Handbuch der Physik,” edited by S. Flügge (Springer-Verlag, New York, 1974), Vol. VIa/4, in “Mechanics of Solids,” Vol. IV, Volume editor: C. Truesdell (Springer-Verlag, New York, 1984) pp. 109-308.
11. D.C. WALLACE, “Thermodynamics of Crystals” (Wiley, New York, 1972).
12. K.Y. KIM, *Phys. Rev. B.* **54** (1996) 6245.
13. H.J. McSKIMIN, in “Physical Acoustics,” edited by W.P. Mason (Academic, New York, 1964) Vol. 1, pp. 271-334.
14. E.P. PAPADAKIS, in “Physical Acoustics,” edited by W.P. Mason and R.N. Thurston (Academic, New York, 1976), Vol. 12, pp. 277-374.
15. S.S. BRENNER, in “Growth and Perfection of Crystals,” edited by R.H. Doremus, B.W. Roberts, and D. Turnbull (John Wiley and Sons, New York, 1958), p. 157.
16. H. KOBAYASHI and Y. HIKI, *Phys. Rev. B.* **7** (1973) 594.
17. A.L. RUOFF, *J. Appl. Phys.* **49** (1978) 197.
18. *Idem.*, *Scripta Metall.* **15** (1981) 525.
19. K.Y. KIM and W. Sachse, *J. Mater. Sci.* **35** (2000) 3197.
20. “Standards on Piezoelectric Crystals,” *Proc. IRE.* **37** (1949) 1378.

21. K.Y. KIM and W. SACHSE, in “Handbook of Elastic Properties of Solids, Liquids, and Gases,” edited by Levy, Bass, and Stern (Academic, New York, 2001), Vol. 1, Chap. 19.
22. K.Y. KIM, “[Equivalence of \$D_{ijkl}\$ and \$Q_{ijkl}\$](#) ” (2023).
23. K.Y. KIM, “[Coefficients of Equation of State Expressed in Higher-Order Elastic Constants](#)” (2023).