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Fourth-Order Elastic Moduli for Polycrystal: Isotropic Aggregate of Hexagonal Single Crystals

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The investigation of non-linear elasticity of solids is performed. The Gibbs free energy of a polycrystalline solid at given pressure P and temperature T is decomposed over the invariants of the Lagrange finite deformations tensor, including fourth-order contributions of deformation components. Based on this, we define the fourth-order elastic moduli for polycrystal (fourth-order Lamé coefficients) under arbitrary pressure (the corresponding coefficients of the second and third order are well known). The linear invariants of the fourth-order elastic constants tensor are used to obtain the relations that define the fourth-order Lamé coefficients through the fourth-order elastic constants of single crystal grains with hexagonal symmetry, which form a polycrystal. The data for the second, third, and fourth-order Lamé coefficients for magnesium and erbium are obtained using the available data for the corresponding order elastic constants of single crystals.

Keywords: polycrystalline materials, non-linear elasticity, fourth-order Lamé coefficients.

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1. Introduction

Higher-order elastic constants play an important role in solid state physics. The higher-order elastic constants represent anharmonic effects from an atomic point of view, in the language of lattice dynamics. They can be used to calculate the derivative frequencies of phonons by deformation (Grüneisen parameters) and the scattering cross sections of these phonons. First of all, this concerns elastic constants of the third and fourth order (TOEC and FOEC, respectively).

On the other hand, TOEC and FOEC determine the nonlinear response of a solid to a finite deformation, the dependence of the speed of sound on the applied load [1,2], the distortion of the shape of an ultrasonic wave of finite amplitude during its propagation in a solid and the amplitudes of the second and third harmonics [3–6], also allow evaluating the ideal strength and ductility of metals [7]. They are important for understanding the patterns of wave propagation in materials under very high pressure, when the pressure becomes comparable to elastic constants. An example is the thermoelastic properties of the Earth's interior [8,9].

Polycrystalline materials are important from a practical point of view. The most convenient way to describe the elastic properties of these materials is to use an isotropic media model. Elastic moduli of a polycrystal (Lamé coefficients) can be obtained by averaging elastic constants of various orders of a single crystal over all orientations of single crystal grains [10–13]. The relations between the second-order Lamé coefficients (SOLC), third-order Lamé coefficients (TOLC) and elastic constants of a single crystal

at normal pressure are given in [11–15]. A generalization of these relations for a polycrystal at arbitrary hydrostatic pressure P is given in [16]. The relations between the fourth-order Lamé coefficients (FOLC) and FOEC of a single crystal with a cubic lattice at arbitrary pressure are obtained in [17].

These relations allow modeling the nonlinear elastic properties of polycrystals *ab initio*, since density functional theory (DFT) allows calculating the elastic constants of the highest order of a single crystal at arbitrary pressure [18–20]. We can determine the nonlinear elastic modulus of a polycrystal at a given P using the relations between the Lamé coefficients and higher-order elastic constants, which are important for understanding the structural behavior and physical properties of materials under load.

We consider in this paper the case when the monocrystalline grains forming a polycrystal have a hexagonal structure, since this structure, together with the cubic one, is characteristic of metals. The relations between FOLC and FOEC of a hexagonal crystal, which are important for technical applications, are given. The Lamé coefficients of the second, third and fourth orders of polycrystalline magnesium and erbium are calculated as an example.

2. Basic definitions and ratios

Let's consider a preloaded single crystal. We select the equilibrium state at temperature T and pressure P as the initial state. Given P and T , the state of the system is described by the Gibbs free energy G . Let the single crystal undergo a small but finite deformation described by

the Lagrange tensor with components η_{ij} . The isothermal effective elastic constants of the second and higher order of a preloaded crystal are determined by the ratio [18]:

$$C_{ijkl\dots} = \frac{1}{V_0} \left(\frac{\partial^n G}{\partial \eta_{ij} \partial \eta_{kl\dots}} \right)_T, \quad n \geq 2, \quad (1)$$

where V_0 — the volume in the initial state. Elastic constants (1) directly enter into the relations connecting the Cauchy stress tensor (true stresses) with the components η_{ij} [21]. When $P = 0$ the ratio (1) coincides with the standard definition of elastic constants of the n -the order of the unloaded crystal [22].

The value G for a deformed isotropic body given P and T is invariant with respect to rotations and displacements of the body as a whole, and does not depend on the choice of coordinate system. Therefore, the Gibbs free energy shall be a function of the invariants of the strain tensor. The strain tensor has three main invariants of the first, second and third degree in the components η_{ij} [1]:

$$I_1 = \text{tr}(\eta) = \eta_{11} + \eta_{22} + \eta_{33}, \quad (2a)$$

$$\begin{aligned} I_2 &= \frac{1}{2} [(\text{tr}\eta)^2 - \text{tr}\eta^2] = (\eta_{11}\eta_{22} - \eta_{12}^2) + (\eta_{11}\eta_{33} - \eta_{13}^2) \\ &\quad + (\eta_{22}\eta_{33} - \eta_{23}^2), \end{aligned} \quad (2b)$$

$$I_3 = \det \eta$$

$$= \eta_{11}\eta_{22}\eta_{33} + 2\eta_{23}\eta_{13}\eta_{12} - \eta_{11}\eta_{23}^2 - \eta_{22}\eta_{13}^2 - \eta_{33}\eta_{12}^2. \quad (2c)$$

Together with the main invariants, their combinations can be used:

$$\begin{aligned} \bar{I}_1 &= I_1, \quad \bar{I}_2 = \text{tr}\eta^2 = I_1^2 - 2I_2, \\ \bar{I}_3 &= \text{tr}\eta^3 = 3I_3 + I_1^3 - 3I_1I_2. \end{aligned} \quad (3)$$

Let's decompose G near the equilibrium state according to the invariants of the strain tensor, including fourth-order contributions by components η_{ij} . The coefficients of this decomposition are the Lamé coefficients of the corresponding order. The Gibbs free energy has a minimum in the equilibrium state, therefore $\partial G / \partial I_1|_0 = 0$, and the decomposition begins with a quadratic contribution over deformation. Two quadratic scalars (I_1^2, I_2), three cubic ones (I_1^3, I_1I_2, I_3) and four fourth order scalars ($I_1^4, I_1^2I_2, I_1I_3, I_2^2$) can be created from the main invariants (2). The same is possible for the invariants $\bar{I}_1, \bar{I}_2, \bar{I}_3$.

The decomposition of the energy of a deformed isotropic solid, taking into account third-order contributions, has been considered in a number of papers [1,4,10,23–25]. Various definitions of TOLC have been used, which is related to the decomposition of invariants (2) or (3). Here are the three most well-known definitions: Murnaghan (l, m, n — decomposition by invariants (2)); Toupin and Bernstein (ν_1, ν_2 and ν_3 invariant decomposition (3)); Landau and Lifshitz (A, B and C — invariant decomposition (3)). The definition of TOLC by Toupin and Bernstein is more convenient (ν_i coincide with the independent elastic constants

of an isotropic solid), therefore we will give ratios linking them with other definitions of TOLC [24,25]:

$$\left. \begin{aligned} \nu_1 &= 2(l-m) + n = 2C \\ \nu_2 &= m - 1/2n = B \\ \nu_3 &= n/4 = A/4 \end{aligned} \right\}. \quad (4)$$

The expression for the Gibbs free energy for the given P and T per unit volume V_0 in the initial state, taking into account the fourth-order contribution of η_{ij} , is represented as follows

$$\begin{aligned} \frac{\Delta G}{V_0} &= \frac{\lambda + 2\mu}{2} I_1^2 - 2\mu I_2 + \frac{\nu_1 + 6\nu_2 + 8\nu_3}{6} I_1^3 \\ &\quad - 2(\nu_2 + 2\nu_3) I_1 I_2 + 4\nu_3 I_3 + \frac{1}{24} \xi_1 I_1^4 \\ &\quad - \left(\frac{\xi_1 - \xi_2}{8} + \frac{\xi_4}{3} \right) I_1^2 I_2 + \left(\frac{\xi_1 - \xi_2}{8} + \frac{\xi_4}{3} - \xi_3 \right) I_1 I_3 + \frac{2}{3} \xi_4 I_2^2, \end{aligned} \quad (5)$$

where $\Delta G = G(P, T, \eta) - G(P, T, 0)$, λ and μ — SOLC, ν_i — TOLC, ξ_i — FOLC.

With this determination of the Lamé coefficients, they coincide with the elastic constants of an isotropic solid. For the second and third order [11,24]

$$\lambda = C_{12}^*, \mu = C_{44}^*, \nu_1 = C_{123}^*, \nu_2 = C_{144}^*, \nu_3 = C_{456}^*. \quad (6)$$

For the fourth order [17]

$$\xi_1 = C_{1111}^*, \xi_2 = C_{1122}^*, \xi_3 = C_{1144}^*, \xi_4 = C_{4444}^*. \quad (7)$$

The elastic constants of an isotropic solid are given here in the Vogt notation (11 — 1, 22 — 2, 33 — 3, 23 — 4, 13 — 5, 12 — 6).

The decomposition of the elastic strain energy, including the fourth-order contribution (η^4), is given in [6,26,27], where sound propagation in nonlinear isotropic solids was considered, and in [28,29] for estimating fourth-order shear modulus in metallic glasses. The invariants determined by equations (3) were used in this case. In the first case, FOLC were designated as E, F, G, H , in the second case they were designated as $\gamma_1, \gamma_2, \gamma_3, \gamma_4$. We present the relations linking these FOLC with the parameters $\xi_1, \xi_2, \xi_3, \xi_4$, which are used in this paper

$$\left. \begin{aligned} H &= \frac{1}{24} \gamma_1 = \frac{1}{6} \left(\frac{\xi_2}{4} - \xi_3 + \frac{\xi_4}{3} \right) \\ F &= \frac{1}{2} \gamma_2 = \frac{1}{2} \left(\xi_3 - \frac{2}{3} \xi_4 \right) \\ E &= \frac{4}{3} \gamma_3 = \frac{1}{3} \left(\frac{\xi_1 - \xi_2}{8} - \xi_3 + \frac{\xi_4}{3} \right) \\ G &= \frac{1}{2} \gamma_4 = \frac{1}{6} \xi_4 \end{aligned} \right\}. \quad (8)$$

It can be seen that the Lamé coefficients defined in these paper are expressed in terms of combinations of elastic

constants of an isotropic body, which is not convenient for practical application.

A polycrystal can be considered as an isotropic aggregate of monocrystalline grains. The grains are arbitrarily oriented, their sizes are infinitely small compared to the size of the sample, but large enough to have volume-elastic properties. The Vogt averaging method [12,13] is used to calculate the elastic constants of an isotropic material (Lamé coefficients). Following Vogt, we believe that all monocrystalline grains in a polycrystal are in the same deformed state, so the elastic constants of such a material are equal to the tensor of elastic constants averaged in all directions

$$C_{ijkl\dots}^V = (C_{ijkl\dots})_{Av}. \quad (9)$$

This procedure is usually called „homogenization“.

3. Calculation method and details

It is convenient to use the method of linear invariants of the tensor of elastic constants used in [12,14,30,31] for calculation of „the homogenized“ values (9) in calculating the Vogt averages for TOEC. The relations between ξ_i and FOEC for random grain orientation can be obtained from the condition of equality of linear invariants of two tensors representing a single crystal and a polycrystal.

The derivation of linear invariants of FOEC for a hexagonal crystal is given in the Appendix. Expressions for linear invariants of an isotropic body are given in [17] (formulas (19))

$$\left. \begin{aligned} L_1^{\text{is}} &= 3(3\xi_1 + 24\xi_2 - 24\xi_3 - 16\xi_4) \\ L_2^{\text{is}} &= 6\xi_1 + 3\xi_2 + 12\xi_3 + 28\xi_4 \\ L_3^{\text{is}} &= (33\xi_1 - 21\xi_2 - 24\xi_3 + 64\xi_4)/4 \\ L_4^{\text{is}} &= 3(57\xi_1 - 9\xi_2 + 24\xi_3 - 104\xi_4)/16 \end{aligned} \right\}. \quad (10)$$

Let's find the relations determining the Lamé coefficients of the fourth order through the FOEC of a hexagonal crystal from the resulting system of equations by equating the corresponding invariants (10) and (A3a)–(A3d) from the Appendix

$$\begin{aligned} \xi_1 &= \frac{1}{315} (64C_{1111} + 64C_{2222} + 35C_{3333} + 128C_{4444} \\ &+ 32C_{1113} + 48C_{1133} + 192C_{1155} + 40C_{1333} \\ &+ 384C_{1355} + 32C_{2223} + 192C_{2244} + 240C_{3344}), \end{aligned} \quad (11a)$$

$$\begin{aligned} \xi_2 &= \frac{1}{105} \left(C_{1111} + C_{2222} + C_{3333} + \frac{64}{3}C_{4444} - \frac{8}{3}C_{6666} \right. \\ &+ 22C_{1122} + 16C_{1113} + 24C_{1123} + 40C_{1133} + 48C_{1144} \\ &+ 8C_{1333} + 16C_{1344} - 48C_{1355} - 8C_{2223} - 16C_{2244} \\ &\left. - 8C_{3366} + 32C_{4466} \right), \end{aligned} \quad (11b)$$

$$\begin{aligned} \xi_3 &= \frac{1}{315} \left(0.5C_{1111} + 0.5C_{2222} + C_{3333} + 32C_{4444} \right. \\ &+ 16C_{6666} + 15C_{1122} - 14C_{1113} - 39C_{1123} + 9C_{1133} \\ &+ 2C_{1333} + 132C_{1144} + 72C_{1155} - 72C_{1355} + 72C_{1344} \\ &\left. + 25C_{2223} - 84C_{2244} + 6C_{3344} + 69C_{3366} - 24C_{4466} \right), \end{aligned} \quad (11c)$$

$$\begin{aligned} \xi_4 &= \frac{1}{105} \left(\frac{7}{8}C_{1111} + \frac{7}{8}C_{2222} + C_{3333} + 24C_{4444} + 23C_{6666} \right. \\ &- 0.75C_{1122} + 6C_{1133} - 2C_{1113} + 6C_{1144} - 24C_{1155} \\ &+ 3C_{1123} - 4C_{1333} - 12C_{1344} - 12C_{1355} - 5C_{2223} \\ &\left. + 30C_{2244} + 12C_{3344} + 6C_{3366} + 60C_{4466} \right). \end{aligned} \quad (11d)$$

4. Numerical results and discussion

The choice of materials with a hexagonal structure with a known complete set of FOEC is very limited. We analyze the homogenized moduli of the second, third and fourth order (Lamé coefficients) of magnesium and erbium with a random orientation of grains having a hexagonal structure with known necessary elastic constants. Data on elastic constants of the second-fourth order Mg and Er are given in [34]. The values of SOEC and TOEC were determined experimentally (normal pressure, room temperature). The complete set of FOEC is obtained from the analysis of data on elastic constants of the second and third order. The values of the elastic constants C_{2222} , C_{2244} , C_{3366} , C_{4466} , C_{6666} , which are not provided in [34], are found from the ratios between the FOEC of the hexagonal crystal (see [33]). The ratios for calculating the second and third order Lamé coefficients are given in [16] (formulas (24)–(28)), the formulas (11a)–(11d) are used for calculating the fourth order Lamé coefficients.

The results of our calculations using data of elastic constants Mg and Er are listed in Table 1.

The Lamé coefficients of the third order have mostly negative values, the Lamé coefficients of the fourth order have mostly positive values.

Table 1. The results of calculations of the Lamé coefficients

Metal	λ	μ	ν_1	ν_2	ν_3	ξ_1	ξ_2	ξ_3	ξ_4
Mg	23.7	17.4	-35.2	-20.2	-61	7665	861	311	659
Er	25.8	28.3	368	-253	102	6480	742	271	556

Note. All values are given in GPa ($P = 0$, $T = 300$ K).

Table 2. Values C_{11}^* , C_{111}^* and C_{1111}^* (GPa)

Metal	$C_{11}^* = \lambda + 2\mu$	$C_{111}^* = \nu_1 + 6\nu_2 + 8\nu_3$	$C_{1111}^* = \xi_1$
Mg	58.5	-644	7665
Er	82.4	-334	6480

The values of C_{11}^* , C_{111}^* and C_{1111}^* are listed in Table 2 for the analysis of the changes of the elastic constants of the second-fourth order of an isotropic solid.

It can be seen that during the transition from the second to the fourth order, the elastic constant modulus increases about an order of magnitude with each transition.

5. Conclusion

The determination of the fourth-order elastic modulus of polycrystals (fourth-order Lamé coefficients) at arbitrary pressure and temperature is given by decomposing the Gibbs free energy by invariants of the Lagrange finite strain tensor. The case of a polycrystal with arbitrarily oriented grains of hexagonal symmetry is considered. The relations determining the Lamé coefficients of the fourth order of such a polycrystal through the elastic constants of the fourth order of monocrystalline grains are obtained. The Lamé coefficients of the second, third and fourth orders of polycrystalline magnesium and erbium are calculated using available data on the elastic constants of single crystals of these materials.

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Appendix

Linear invariants of the FOEC tensor of a hexagonal crystal.

The components of the eighth-rank tensor are transformed according to the following law in case of transition from one orthonormal basis to another [32]:

$$C'_{ijklmnop} = a_{iq}a_{jr}a_{ks}a_{lt}a_{mu}a_{nv}a_{ow}a_{px}C_{qrstuvwxyz}, \quad (A1)$$

where a_{iq}, \dots — the guiding cosines between the coordinate axes. The FOEC tensor has four linear invariants that do not change with any orthogonal transformation of the vector basis. The rotation matrices in equation (A1) should be taken in pairs to obtain these invariants and their indices should be changed so that the product of each of these pairs becomes δ -Kronecker function ($\delta_{ij} = 1$ if $i = j$, and 0 if $i \neq j$) [32].

For example, if $i = j$, $k = l$, $m = n$ and $o = p$ is selected, the following is obtained

$$\begin{aligned} L_1 &= C'_{iikkmmpp} \\ &= (a_{iq}a_{ir})(a_{ks}a_{kt})(a_{mu}a_{mv})(a_{pw}a_{px})C_{qrstuvwxyz} \\ &= \delta_{qr}\delta_{st}\delta_{uv}\delta_{wx}C_{qrstuvwxyz}. \end{aligned} \quad (A2a)$$

We get the second invariant taking $i = m$, $j = n$, $k = o$ and $l = p$:

$$L_2 = C'_{ijkljkl} = \delta_{qu}\delta_{rv}\delta_{sw}\delta_{tx}C_{qrstuvwxyz}. \quad (A2b)$$

Then, putting $i = k$, $j = m$, $l = o$, $n = p$, we find

$$L_3 = C'_{ijiljlnl} = \delta_{qs}\delta_{ru}\delta_{tw}\delta_{vx}C_{qrstuvwxyz}, \quad (A2c)$$

and for $i = l$, $j = n$, $k = m$, $o = p$

$$L_4 = C'_{ijkikjoo} = \delta_{qt}\delta_{rv}\delta_{su}\delta_{wx}C_{qrstuvwxyz}. \quad (A2d)$$

The repeated indexes are summed from 1 to 3 as usual.

For the case of hexagonal symmetry (Laue group HI : point groups $\bar{6}2m$, $6mm$, 622 , $\frac{6}{m}\frac{2}{m}\frac{2}{m}$, 19 independent FOEC [33]) expressions for linear invariants (equations (A2)) have the following form

$$\begin{aligned} L_1^H &= 2(C_{1111} + C_{2222}) + C_{3333} + 12C_{1122} + 16C_{1113} \\ &+ 24C_{1133} + 24C_{1123} + 8C_{1333} - 8C_{2223} - 24C_{3366} - 16C_{6666}, \end{aligned} \quad (A3a)$$

$$\begin{aligned} L_2^H &= 1.5(C_{1111} + C_{2222}) + C_{3333} + \frac{32}{3}C_{4444} + \frac{20}{3}C_{6666} \\ &+ C_{1122} + 4C_{1133} + 8C_{1144} + 8C_{2244} \\ &+ 4C_{3366} + 8C_{3344} + 16C_{4466}, \end{aligned} \quad (A3b)$$

$$\begin{aligned} L_3^H &= \frac{7}{4}(C_{1111} + C_{2222}) + C_{3333} + \frac{16}{3}C_{4444} + \frac{10}{3}C_{6666} \\ &- 1.5C_{1122} - 4C_{1144} + 12C_{2244} - 4C_{1344} \\ &+ 12C_{1355} + 8C_{3344} + 8C_{4466}, \end{aligned} \quad (A3c)$$

$$\begin{aligned} L_4^H &= 2(C_{1111} + C_{2222}) + C_{3333} - 4C_{6666} + C_{1113} \\ &- 1.5C_{1123} + 2C_{1333} + 12C_{1155} + 3C_{1344} \\ &+ 15C_{1355} + 2.5C_{2223} + 6C_{3344} - 12C_{4466}. \end{aligned} \quad (A3d)$$

Here $C_{\alpha\beta\gamma\delta}$ — isothermal elastic constants of the fourth order with given P and T in the Vogt notation.

Conflict of interest

The authors declare that they have no conflict of interest.

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