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Elastic Properties of Polycrystalline Fe: The Pressure Effect

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The elastic moduli of a polycrystal (Lamé constants) of the second and third order at pressure P are expressed through derivatives of Gibbs free energy with respect to invariants of Lagrange's finite deformation tensor. Relations connecting these Lamé constants with corresponding elastic constants of monocrystalline grains having hexagonal lattice structure that constitute the polycrystal are presented. Based on data for second- and third-order elastic constants of ε-Fe, the respective Lamé constants of polycrystalline Fe were calculated over the pressure range from 50 to 340 GPa. Using the obtained results, an analysis was carried out on the intensity of various three-phonon scattering processes in iron and their dependence on pressure

Keywords: high pressures, Lamé constants of the second and third order, three-phonon scattering processes.

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1. Introduction

Iron is the main component of the Earth's inner core, where the pressure reaches 330−360 GPa. There is ample evidence for the stability of the epsilon phase of iron with a hexagonal close-packed (HCP) crystal lattice at such extremely high pressures and at low and moderate temperatures [1–4]. According to experimental data, the transition of Fe from a body-centered cubic (BCC) to a HCP structure at room temperature occurs at pressures of 10−18 GPa, and the HCP phase remains stable up to at least 400 GPa [3–5]. Knowledge of the elastic properties of this phase is important for many reasons. For example, the elastic properties of iron affect the propagation of seismic waves through the Earth's core, and knowledge of them is critical for interpreting seismological data and understanding the internal structure of our planet [6]. The study of the elastic properties of polycrystalline iron at ultrahigh pressures is a promising task.

The elastic properties of ε-Fe have been studied in various experimental and theoretical works. Second-order elastic constants (SOEC) of single crystals at various pressures were found in Refs. [7–10] from experiments with polycrystalline ε-Fe samples. The results of calculations of the SOEC of monocrystalline ε-Fe in the pressure range up to 400GPa in the framework of density functional theory (DFT) are given in Refs. [11–16]. The elastic constants of the third order (TOEC) ε-Fe were also calculated in Ref. [16] in the pressure range of 50−340 GPa. Along with SOECS, which characterize the linear elastic response of a material, third-order elastic constants determine the nonlinear response to finite deformation and are important for explaining the anharmonic properties of a solid, such as thermal expansion, the dependence of the speed of sound on temperature and applied load, and the generation of second harmonics during propagation of finite-amplitude ultrasonic waves in a solid [17–20].

In the case of ε-Fe, we are talking about the elastic constants of a solid body under very high pressure (the pressure is comparable to the magnitude of the volumetric modulus). Pressure P is a fundamental thermodynamic variable along with temperature T. However, unlike T, research at high pressures has long been hampered by the lack of the possibility of both obtaining static pressures in the megabar range (100 GPa) and measuring the properties of a highly compressed material. Currently, after the invention of the diamond anvil cell (DAC) and the advent of modern micro-nano-sensing technologies, it is possible to study the state of matter beyond three megabars. As a result, the number of studies on the properties of solids at high pressures is growing rapidly [21]

Pressure radically changes all the properties of solids, including elastic constants of various orders. The elastic constants n of the order $(n \geq 2)$ are defined as the corresponding partial derivatives of the thermodynamic potential of a thermoelastic medium subjected to a small but finite deformation [22]. The internal energy $U = U(S, V)$ is a thermodynamic potential in variables S (entropy) and V (volume), the free Helmholtz energy $F = F(T, V)$ is a thermodynamic potential in variables T and V. The standard definition of elastic constants n of the order $(n \geq 2)$ is given in Ref. [22] for these two cases (without pressure):

$$C^S_{ijkl\ldots} = \frac{1}{V_0}\left(\frac{\partial^n U}{\partial\eta_{ij}\partial\eta_{kl\ldots}}\right)_S, \tag{1}$$

$$C^T_{ijkl\ldots} = \frac{1}{V_0}\left(\frac{\partial^n F}{\partial\eta_{ij}\partial\eta_{kl\ldots}}\right)_T, \tag{2}$$

Here $C^S_{ijkl\ldots}$ and $C^T_{ijkl\ldots}$ are adiabatic and isothermal elastic constants, respectively, at $P = 0$, η_{ij} are components of the

Lagrange finite strain tensor [17], V_0 is the volume in the initial (undeformed) state. The derivatives are calculated at constant entropy S and temperature T.

The enthalpy $H(S,P)$ is the thermodynamic potential in variables S and P, and Gibbs free energy $G(T,P)$ is the thermodynamic potential in variables T and P. Therefore, the adiabatic and isothermal elastic constants of various orders of a preloaded crystal (at pressure P) can be defined as the corresponding derivatives of enthalpy H and Gibbs potential G in terms of components η_{ij} [23–25]:

$$\tilde{C}^S_{ijkl\ldots} = \frac{1}{V_0}\left(\frac{\partial^n H}{\partial\eta_{ij}\partial\eta_{kl\ldots}}\right)_S, \tag{3}$$

$$\tilde{C}^T_{ijkl\ldots} = \frac{1}{V_0}\left(\frac{\partial^n G}{\partial\eta_{ij}\partial\eta_{kl\ldots}}\right)_T, \tag{4}$$

These constants are called „effective“ elastic constants to emphasize that they are determined not only by the interatomic interaction (derivatives of the nth order of free (internal) energy with respect to the components of the strain tensor η_{ij}), but also directly with respect to the external load [23,25]. They fully determine the elastic properties of a loaded crystal: the ratio of Cauchy stress (true stress) — strain η_{ij}, the equation of small vibrations, and stability conditions [17,26]. In the case of hydrostatic pressure, the constants (3) and (4) have complete Voigt symmetry to the permutation of the indices [17]. At $P = 0$, the elastic constants (3) and (4) coincide with the constants for an unloaded crystal (1) and (2). Further, when talking about elastic constants, we will keep in mind the constants (4).

In the polycrystalline state, the material can be considered as an isotropic aggregate of monocrystalline grains. Accordingly, the elastic modules of a polycrystal (Lamé constants) can be obtained by averaging a tensor of elastic constants of the appropriate order over all orientations of single crystal grains. Relations linking the Lamé constants of the second (SOLC) and third order (TOLC) with elastic constants of the 2nd and 3rd order of a crystal of arbitrary symmetry (without pressure) were obtained in Refs. [27–30], and a special case of monocrystalline grains of cubic symmetry was also considered.

In this paper, expressions for the Lamé constants of the 2nd and 3rd order are given in terms of the elastic constants of monocrystalline grains of hexagonal symmetry that make up a polycrystal under high pressure. SOLC and TOLC of polycrystalline Fe were calculated over a wide pressure range (50−340 GPa) based on these relations, using data on elastic constants ε-Fe [16]. The influence of various three-phonon scattering processes on the anharmonic behavior of ε-Fe is also estimated. The results obtained are important for the interpretation of seismic studies, allow for a deeper understanding of the mechanisms of phase transitions occurring at high pressures, and contribute to the development of new materials capable of withstanding extreme external influences such as high pressures and temperatures.

2. Basic definitions and relations

Let us consider an isotropic solid (polycrystal) in an equilibrium state at pressure P and temperature T. Given P and T, the state of the system is described by the Gibbs free energy G. Let the polycrystal undergo a small but finite deformation described by the Lagrange tensor with components η_{ij}. The value G should not depend on the choice of the coordinate system. It is invariant with respect to the rotation and displacement of the deformed body as a whole. This is possible if G is a function of invariants of the strain tensor. The main invariants of the strain tensor have the form [19]

$$I_1 = \mathrm{tr}(\eta) = \eta_{11} + \eta_{22} + \eta_{33}, \tag{5a}$$

$$\begin{aligned} I_2 &= \frac{1}{2}\left[(\mathrm{tr}\eta)^2 - \mathrm{tr}\eta^2\right] \\ &= (\eta_{11}\eta_{22} - \eta_{12}^2) + (\eta_{11}\eta_{33} - \eta_{13}^2) + (\eta_{22}\eta_{33} - \eta_{23}^2), \end{aligned} \tag{5b}$$

$$\begin{aligned} I_3 &= \det\eta \\ &= \eta_{11}\eta_{22}\eta_{33} + 2\eta_{23}\eta_{13}\eta_{12} - \eta_{11}\eta_{23}^2 - \eta_{22}\eta_{13}^2 - \eta_{33}\eta_{12}^2. \end{aligned} \tag{5c}$$

Since the deformations are small, we decompose G into a series by invariants (5) near the undeformed state. Since the undeformed state is considered to be in equilibrium, then $(\frac{\partial G}{\partial I_1})_0 = 0$. Therefore, the decomposition begins with quadratic terms. The coefficients of this decomposition are the Lamé coefficients of the 2nd and 3rd order.

Two quadratic scalars (I_1^2, I_2) and three cubic ones (I_1^3, I_1I_2, I_3) can be created from the invariants (5). By analogy with Ref. [19], we obtain

$$\left.\begin{array}{c} \mu = -\frac{1}{2}\left(\frac{\partial G}{\partial I_2}\right)_0, \\ \lambda + 2\mu = \left(\frac{\partial^2 G}{\partial I_1^2}\right)_0 \\ \left(\frac{\partial G}{\partial I_3}\right)_0 = n = A,\ \left(\frac{\partial^2 G}{\partial I_1\partial I_2}\right)_0 = -4m = -2A - 4B \\ \left(\frac{\partial^3 G}{\partial I_1^3}\right)_0 = 4m + 2l = 2A + 6B + 2C \end{array}\right\}. \tag{6}$$

Here λ and μ are the Lamé constants of the 2nd order; l, m, n are the Lamé constants of the 3rd order in the Murnaghan definition [31], A, B, C in the definition of Landau–Lifshitz [32]. The Lamé constants (6) are derivatives of the Gibbs potential of the corresponding order, and not the Helmholtz free energy, as in Refs. [31,32]. Of course, these definitions match for $P = 0$.

The expression for Gibbs free energy, related to the unit volume, taking into account the contribution of the 3rd order, has the form

$$\frac{\Delta G}{V_0} = \frac{1}{2}(\lambda + 2\mu)I_1^2 - 2\mu I_2 + \frac{1}{3}(l + 2m)I_1^3 - 2mI_1I_2 + nI_3, \tag{7}$$

where $\Delta G = G(P,T,\eta) - G(P,T,0)$, V_0 is the volume of the body in an undeformed state. In this approximation, the elastic properties of an isotropic body are characterized by five constants. Most often, similar quantities are used as Lamé constants of the 3rd order in the definition of Toupin and Bernstein — ν_1, ν_2, ν_3 [33], associated with l, m, n relations [28]:

$$\left.\begin{aligned} \nu_1 &= 2(l-m)+n = 2C \\ \nu_2 &= m - \tfrac{1}{2}n = B \\ \nu_3 &= \tfrac{1}{4}n = \tfrac{1}{4}A \end{aligned}\right\}. \qquad (8)$$

With this choice of Lamé constants, they all coincide with the independent elastic constants of an isotropic solid [28]:

$$\lambda = \tilde{C}^*_{12},\ \mu = \tilde{C}^*_{44},\ \nu_1 = \tilde{C}^*_{123},\ \nu_2 = \tilde{C}^*_{144},\ \nu_3 = \tilde{C}^*_{456}. \quad (9)$$

The elastic constants of an isotropic solid are give here in the Voigt notation (1 1 − 1, 2 2 − 2, 3 3 − 3, 2 3 − 4, 1 3 − 5, 1 2 − 6).

3. Calculation method and details

A polycrystalline iron sample at high pressure is a collection of a large number of monocrystalline grains with a HCP structure. The grains are arbitrarily oriented, their dimensions are negligible compared to the sample size, but large enough to have volume-elastic properties. Following Voigt [23], we believe that all single-crystal grains in a polycrystal are in the same deformed state, therefore, the elastic constants of such a material are described by a tensor of elastic constants of the appropriate order, averaged in all directions.

To find the isotropic average values of the elastic constants of the 2nd and 3rd order of polycrystalline ε-Fe, we use the results of the recently published paper [34]. It develops numerical algorithms for symbolic calculations of effective elastic constants of the second and sixth order for polycrystalline aggregates having symmetry of any class, including complete isotropy. The previous isotropic averages were limited to the 4th order (see Ref. [35]), therefore, the results obtained in Ref. [34] significantly expand the possibilities of studying the nonlinear elasticity of polycrystalline materials [36].

Appendix D (see Ref. [34]) contains expressions for isotropic averages of elastic constants in the case of arbitrary symmetry of crystallites. Taking into account the relations between the elastic constants for the hexagonal structure (classes 622, $6\bar{2}m$, $6/mmm$) (Appendix A, [34]), we obtain for the Lamé constants (9) of polycrystalline iron at a pressure P and temperature T:

$$\lambda = \tilde{C}^*_{12} = \frac{1}{15}\left(\tilde{C}_{11} + 5\tilde{C}_{12} + 8\tilde{C}_{13} + \tilde{C}_{33} - 4\tilde{C}_{44}\right); \quad (10)$$

$$\mu = \tilde{C}^*_{44} = \frac{1}{15}\left(3.5\tilde{C}_{11} - 2.5\tilde{C}_{12} - 2\tilde{C}_{13} + \tilde{C}_{33} + 6\tilde{C}_{44}\right); \quad (11)$$

Table 1. Results of calculations of Lamé constants (GPa) in polycrystalline iron

P, GPa	K	λ	μ	ν_1	ν_2	ν_3
54.27	521.3	331.8	284.3	−348.4	−622.3	−623.0
90.67	664.3	438.2	339.1	−401.8	−772.1	−736.2
126.6	799.6	540.0	389.5	−451.1	−910.2	−840.9
162.1	929.7	638.5	436.9	−500.5	−1041	−940
197.4	1056	734.8	481.3	−554.6	−1163	−1028
232.4	1178	829.0	523.7	−602.8	−1280	−1117
267.3	1298	921.6	564.2	−647.0	−1393	−1203
302.1	1415	1013	602.9	−687.5	−1504	−1290
319.4	1473	1059	621.7	−709.5	−1560	−1333
336.6	1531	1104	640.5	−729.4	−1616	−1375

$$\nu_1 = \tilde{C}^*_{123} = \frac{1}{105}\left(-\tilde{C}_{111} + 10\tilde{C}_{112} + 3\tilde{C}_{113} + 11\tilde{C}_{122} + 63\tilde{C}_{123} + 18\tilde{C}_{133} - 84\tilde{C}_{144} + 12\tilde{C}_{155} + \tilde{C}_{333} - 12\tilde{C}_{344}\right); \quad (12)$$

$$\nu_2 = \tilde{C}^*_{144} = \frac{1}{105}\left(2.5\tilde{C}_{111} + 3\tilde{C}_{112} + 13.5\tilde{C}_{113} + 0.5\tilde{C}_{122} - 24.5\tilde{C}_{123} + 4\tilde{C}_{133} + 56\tilde{C}_{144} - 16\tilde{C}_{155} + \tilde{C}_{333} + 2\tilde{C}_{344}\right); \quad (13)$$

$$\nu_3 = \tilde{C}^*_{456} = \frac{1}{210}\left(8.5\tilde{C}_{111} - \tilde{C}_{112} - 15\tilde{C}_{113} - 9.5\tilde{C}_{122} + 21\tilde{C}_{123} - 6\tilde{C}_{133} - 63\tilde{C}_{144} + 45\tilde{C}_{155} + 2\tilde{C}_{333} + 18\tilde{C}_{344}\right). \quad (14)$$

Here $\tilde{C}_{\alpha\beta\cdot}$ are elastic constants (4) (the dot means that elastic constants of both the 2nd and 3rd order are possible here).

Formulas (10)−(14) completely coincide with the previously obtained relations for the Lamé constants of the 2nd and 3rd order of isotropic aggregates of hexagonal crystals [37](formulas (24)−(28)), considering that $C_{222} = C_{111} + C_{112} - C_{122}$ (Appendix A, [34]).

It should be noted that the analogous relations for the Lamé constants of the 4th order of polycrystals with grains of cubic and hexagonal symmetry, which we obtained earlier in [35] and [38], also completely coincide with the relations following from Appendix B, [34].

4. Calculation results and their discussion

Elastic constants of the 2nd and 3rd order were calculated in Ref. [16] within the framework of density functional theory (DFT) (see the formula (4)) HCP of iron in the pressure range of 50−340 GPa at $T = 0$ K. Using these data and the relations (10)−(14), we find the values of the Lamé constants in the specified pressure range. The results obtained are shown in Figures 1 and 2, and some of them are shown in Table 1.

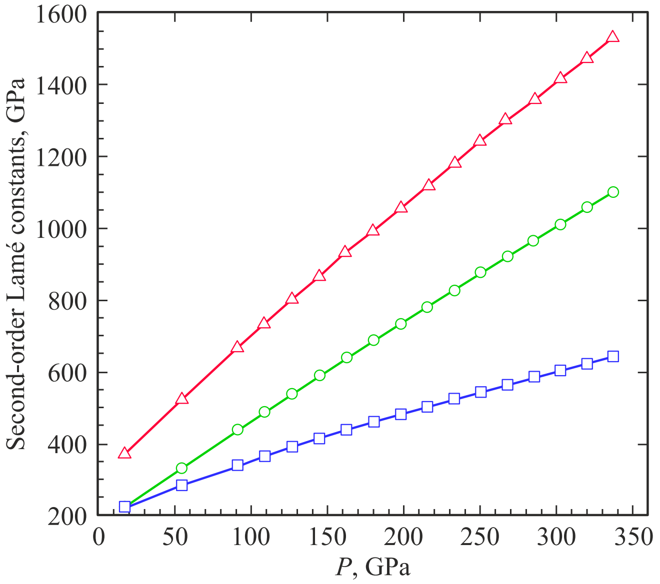

Figure 1. Pressure dependence of the all-round compression module $K = \lambda + (2/3)\mu$, as well as λ and μ. Triangles — K; circles — λ; squares — μ.

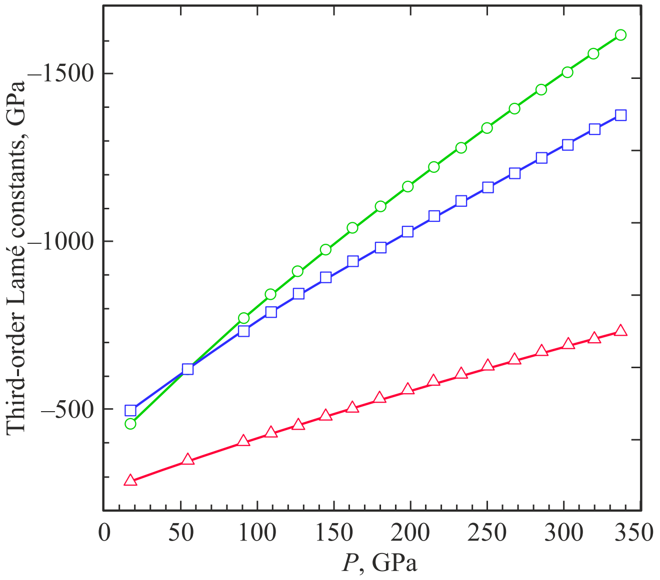

Figure 2. Pressure dependence of the Lamé constants of the 3rd order. Triangles — ν_1; circles — ν_2; squares — ν_3.

Table 2. Relative intensity of three-phonon scattering processes in polycrystalline iron

P, GPa	d/c	d/b	b/c
54.27	401	151.0	2.65
126.6	381	148.4	2.57
197.4	369	147.5	2.50
267.3	361	147.4	2.45
336.6	355	147.3	2.41

It can be seen that in the entire pressure range studied, the constants λ and μ are positive, and all the Lamé constants of the 3rd order have negative values. With increasing pressure, all constants of the 2nd and 3rd order increase in absolute magnitude. At the same time, the dependence of all Lamé constants on pressure is close to linear.

Three-phonon scattering processes play a key role in explaining a number of properties of solids, such as thermal conductivity, temperature dependences of heat capacity and speed of sound. Using the relations (A1)−(A4) (see Appendix), let us estimate the intensity of these processes in polycrystalline iron at high pressures. Table 2 shows the intensity ratios of various three-phonon scattering processes in the pressure range 50−340 GPa.

It can be seen that the process d (A4), $(\mathrm{L}+\mathrm{L}\leftrightarrow\mathrm{L})$ is crucial in explaining the anharmonic properties of iron at high pressures. Two other processes such as b (A2) and c (A3) are much less effective.

With pressure increase, the intensity of three-phonon scattering b, c and d monotonously increase to approximately the same degree:

$$\frac{b(337)}{b(54)} = 5.2; \quad \frac{c(337)}{c(54)} = 5.7; \quad \frac{d(337)}{d(54)} = 5.0.$$

5. Conclusion

The Lamé constants of the 2nd and 3rd order at arbitrary pressure P are determined by the Gibbs free energy decomposition method using invariants of the Lagrange finite strain tensor. For an isotropic aggregate of monocrystalline grains with a hexagonal structure, relations have been obtained relating the Lamé constants of the 2nd and 3rd order to the corresponding elastic constants of the crystallites. The Lamé constants of the 2nd and 3rd order of polycrystalline iron in the pressure range from 50 to 340 GPa were calculated using data on elastic constants ε-Fe. With pressure, all constants of the 2nd and 3rd order increase in absolute magnitude according to a law close to linear. In this pressure range, the contribution of various three-phonon scattering processes to the anharmonic properties of polycrystalline iron is also estimated.

The results obtained in this work are important for the interpretation of geophysical research and contribute to the development of new materials suitable for operation in extreme conditions (high pressures and temperatures).

Appendix

The cubic anharmonic term in the expansion of elastic energy can be associated with the three-phonon scattering strengths $\overline{\left[A^{ss's''}\right]^2}$ [39], which describe the following possible acoustic phonon scattering processes in the isotropic continuum model [39]:

$$\mathrm{T}+\mathrm{T} \leftrightarrow \mathrm{L};\quad \mathrm{T}+\mathrm{L} \leftrightarrow \mathrm{L};\quad \mathrm{T}+\mathrm{T} \leftrightarrow \mathrm{T};\quad \mathrm{L}+\mathrm{L} \leftrightarrow \mathrm{L},$$

where T and L are transverse and longitudinal acoustic phonons, respectively.

In terms of isotropic elastic constants, the values $\overline{|A^{ss's''}|}^2$ have the form [39]

$$a = \overline{|A^{\mathrm{TTT}}|}^2 = 0, \tag{A1}$$

$$b = \overline{|A^{\mathrm{TLL}}|}^2 = 0.0255(A+3\mu)^2 + 0.1333(\lambda+2B)^2 + 0.0593(A+3\mu)(\lambda+2B), \tag{A2}$$

$$c = \overline{|A^{\mathrm{TTL}}|}^2 = 0.1333[\lambda^2 + 1.5B(\lambda+B)] + (A+4\mu) \times [0.0124(A+4\mu) + 0.0222(\lambda+2B)], \tag{A3}$$

$$d = \overline{|A^{\mathrm{LLL}}|}^2 = [3\lambda + 6\mu + 2(A+3B+C)]^2. \tag{A4}$$

Here A, B and C are the 3rd-order Lamé constants in the Landau-Lifshitz definition related to ν_1, ν_2, ν_3 by the ratio (8). It follows from (A1) that in the isotropic continuum model, the scattering intensity for the process T+T↔T is zero.

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Conflict of interest

The authors declare that they have no conflict of interest.

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