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# An Iterative Scheme to Estimate the Values of Elastic Constants of a Solid at High Pressures from the Sound Wave Velocity Measurements[1]

By

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The transit time measurements of sound wave velocities in a solid as a function of pressure contain all the possible information about the mechanical changes brought about in the solid due to the application of pressure. The present work gives a procedure to estimate accurately the values of the elastic constants of a solid from the transit time measurements as a function of pressure without *a priori* knowledge of the compressibility of the solid. When the transit time measurements are made as a function of pressure at more than two temperatures the procedure developed here also gives estimates for (i) the pressure derivative of the thermal expansion coefficient, (ii) the temperature derivative of the thermal expansion coefficient, and (iii) the pressure derivative of the specific heat, all as a function of pressure.

Messungen der Durchgangszeiten von Schallwellen in einem Festkörper als Funktion des Druckes enthalten alle möglichen Informationen über mechanische Veränderungen, die durch Anwendung von Druck im Festkörper bewirkt werden. Die vorliegende Arbeit gibt ein Verfahren an, um die Werte der elastischen Konstanten eines Festkörpers aus Messungen der Durchgangszeiten als Funktion des Druckes ohne a-priori-Kenntnisse der Kompressibilität genau abzuschätzen. Werden Messungen der Durchgangszeiten als Funktion des Druckes bei mehr als zwei Temperaturen durchgeführt, liefert das Verfahren außerdem: 1. die Druckableitung des thermischen Ausdehnungskoeffizienten, 2. die Temperaturableitung des thermischen Ausdehnungskoeffizienten, 3. die Druckableitung der spezifischen Wärme, alle als Funktion des Druckes.

## 1. Introduction

This paper presents an iterative procedure to estimate the values of the elastic constants of a crystalline solid at high pressures from the sound wave velocity measurements under the assumption that the concomitant compressibility measurements are either unavailable or unreliable. This procedure resembles the procedure developed by Cook [1] with regard to the use of the principle of self-consistent integration but differs with regard to the estimation of $\Delta(l, m, n, P, T)$ (cf. equation (7)). The iterative procedure presented here requires no restrictive assumptions when the sonic or ultrasonic measurements are made as a function of pressure at more than two temperatures. If, however, these measurements are made as a function of pressure at two temperatures, then we assume that the temperature derivatives of the linear thermal expansion coefficients are independent of pressure and they may be represented by their respective values at some lower pressure where they are known. Lastly, if the ultrasonic measurements are made as a function of pressure at only one tem-

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perature then an additional assumption must be made, i.e. the temperature derivatives of isothermal linear compressibilities are independent of pressure and they may be represented by their respective values at some lower pressure where they are known. For simplicity of presentation the analysis given in this paper refers to measurements of the transit-time of an elastic wave propagated in an anisotropic medium. No attempt is made to establish convergence of the procedure because it is generally difficult to do so for a numerical procedure unambiguousy since no explicit analytic expression can be obtained *a priori*. However, the iterative procedure presented in this paper predicts appropriate elastic constants whenever the data being analyzed are internally consistent; three examples discussed later confirm the utility of this procedure.

## 2. Conventions and Notations

For convenience, the elastic constants of a solid refer to the coordinate system $(x, y, z)$ related to the crystal axes $(a, b, c)$ as defined by the IRE Standards Committee [2]. The number of elastic constants $(C_{ijkt})$ necessary to characterize the elastic property of a solid depends upon the crystal class to which the solid belongs. The subscripts of these constants were contracted to $(C_{pq})$ by following the usual convention of writing the subscript $ij$, $kt = 11, 22, 33, 23, 12, 13$ by $p, q = 1, 2, 3, 4, 5, 6$ so as to represent these $C_{pq}$ by a $6 \times 6$ matrix, denoted by $[C_{pq}]$. The corresponding $6 \times 6$ matrix of the elastic compliances $S_{pq}$ may be obtained from $C_{pq}$ by using the matrix relation between them, namely

$$[C_{pq}] \times [S_{pq}] = [I] , \tag{1}$$

where $[I]$ is a $6 \times 6$ unit matrix. The $C_{pq}$-matrices for the different crystal classes may be found in [3].

### *Notations*

| | |
|---|---|
| $P$ | Pressure |
| $T$ | Temperature |
| $\varrho(P, T)$ | Density of a solid at pressure $P$ and temperature $T$ |
| $\chi(l, m, n, P, T)$ | Linear compressibility of the solid in the direction whose direction cosines are given by $l$, $m$, and $n$ at $P$ and $T$ |
| $\beta(l, m, n, P, T)$ | Linear thermal expansion of the solid in the direction whose direction cosines are given by $l$, $m$, and $n$ at $P$ and $T$ |
| $\chi(P, T)$ | Volume compressibility of the solid at $P$ and $T$ |
| $\beta(P, T)$ | Volume thermal expansion of the solid at $P$ and $T$ |
| $L(l, m, n, J, P, T)$ | Width of the specimen used to measure the $J$th velocity mode in the solid in the direction such that $l$, $m$, $n$ determine the direction cosines at $P$ and $T$ |
| $\tau(l, m, n, J, P, T)$ | Transit time of the $J$th wave velocity mode corresponding to $L(l, m, n, J, P, T)$ |
| $V(l, m, n, J, P, T)$ | The $J$th velocity in the solid in the direction whose direction cosines are $l$, $m$, and $n$ at $P$ and $T$ |
| $\lambda(l, m, n, J, P, T)$ | $= \dfrac{L(l, m, n, J, P_1, T)}{L(l, m, n, J, P, T)}$, where $P_1 < P$ and $P$ equal to unity indicates one atmosphere pressure |
| $C_P(P, T)$ | Specific heat of the solid at constant $P$ and $T$. |

The superscripts $T$ and $S$ attached to a quantity indicate its isothermal and adiabatic values, respectively.

## 3. Preliminaries

The various relations used in the iterative procedure to be described originate either in the theory of elastic wave propagation in a solid or in thermodynamics theory. These general relations are presented with brief introductory remarks drawn from the two theories and appended only to clarify the material of this paper.

### *3.1 Relations obtained from the theory of elastic wave propagation*

The elastic constants of a solid are determined by measuring the velocities with which elastic waves are propagated along several directions in the solid. The number of velocity measurements needed to understand the elastic property of the solid depends on the crystallographic class to which it belongs. Christoffel's equations (e.g. equation (2)) which are applicable to any crystalline system give the relationship between measured velocities and elastic constants. In general, for a plane wave propagated in a crystal having direction cosines $l$, $m$, $n$, the three possible wave velocities $V$ may be found in terms of the elastic constant $C_{pq}$ from the roots of Christoffel's equations.

$$\begin{vmatrix} A_{11} - \varrho V^2 & A_{12} & A_{13} \\ A_{12} & A_{22} - \varrho V^2 & A_{23} \\ A_{13} & A_{23} & A_{33} - \varrho V^2 \end{vmatrix} = 0 \,, \tag{2}$$

where

$$\begin{aligned} A_{ij} = l^2 C_{1i1j} + m^2 C_{2i2j} + n^2 C_{3i3j} + l\, m\, (C_{1i2j} + C_{2i1j}) + \\ + l\, n\, (C_{1i3j} + C_{3i1j}) + m\, n\, (C_{2i3j} + C_{3i2j}) \,. \end{aligned} \tag{3}$$

It follows that $\varrho V^2$ is related to $C_{ijkt}$ in a manner determined by the direction in which a wave is propagated. Three different velocities of propagation imply that the three displacement vectors associated with these velocities are mutually perpendicular and hence independent. Usually the three waves are mixed; one is predominantly longitudinal and the other two are predominantly shear. Pure waves may be propagated only in a few special crystallographic directions. The location of the pure mode directions in crystals of various symmetries have been investigated exhaustively by Borgnis [4] and Brugger [5].

### *3.2 Thermodynamic relations*

These relations serve two purposes:

(i) To evaluate the pressure derivatives of the specific heat and the linear thermal expansions in the three principal directions of the solid at pressure $P$ and temperature $T$; and

(ii) To convert the adiabatic quantities into their isothermal counterparts at pressure $P$ and temperature $T$.

The adiabatic and isothermal elastic compliances are related by

$$S^S_{ijkt}(P, T) - S^T_{ijkt}(P, T) = -\beta_{ij}(P, T)\, \beta_{kt}(P, T)\, T[C_P(P, T)\, \varrho(P, T)]^{-1} \,. \tag{4}$$

When expressed in our notation the following relation between $\chi^S(l, m, n, P, T)$ and $\chi^T(l, m, n, P, T)$ results:

$$\chi^T(l, m, n, P, T) = \chi^S(l, m, n, P, T) + \varDelta(l, m, n, P, T) \,, \tag{5}$$

where

$$\chi(l, m, n, P, T) = (S_{11} + S_{12} + S_{13})\, l^2 + (S_{12} + S_{22} + S_{23})\, m^2 + \\ + (S_{13} + S_{23} + S_{33})\, n^2 , \qquad (6)$$

$$\Delta(l, m, n, P, T) = \frac{\beta(P, T)\, T}{\varrho(P, T)\, C_P(P, T)} \{\beta(1, 0, 0, P, T)\, l^2 + \\ + \beta(0, 1, 0, P, T)\, m^2 + \beta(0, 0, 1, P, T)\, n^2\} , \qquad (7)$$

and

$$\beta(P, T) = \beta(1, 0, 0, P, T) + \beta(0, 1, 0, P, T) + \beta(0, 0, 1, P, T) . \qquad (8)$$

From the definition of isothermal linear compressibility, we have

$$\chi^T(l, m, n, P, T) = -\frac{1}{L(l, m, n, P, T)} \left(\frac{\partial L(l, m, n, P, T)}{\partial P}\right)_T \\ = \frac{1}{\lambda(l, m, n, P, T)} \left(\frac{\partial \lambda(l, m, n, P, T)}{\partial P}\right)_T . \qquad (9)$$

Since the suffix $J$ has no significance in the above relation, it has been omitted from $L(l, m, n, J, P, T)$. By integrating equation (9) with respect to pressure, we obtain

$$\lambda(l, m, n, P, T) = \lambda(l, m, n, P_1, T) \exp \{(P - P_1)\, \chi^T(l, m, n, P, T)\} , \qquad (10)$$

by taking into account definition (9) which implies that $\chi^T(l, m, n, P, T)$ remains constant in the range of integration $P$ to $P_1$. Again the temperature derivative of linear compressibility is related to the pressure derivative of the linear thermal expansion of a material by

$$\left(\frac{\partial \chi^T(l, m, n, P, T)}{\partial T}\right)_P = -\left(\frac{\partial \beta(l, m, n, P, T)}{\partial P}\right)_T \qquad (11)$$

and the pressure derivative of specific heat may be written as

$$\left(\frac{\partial C_P(P, T)}{\partial P}\right)_T = -\frac{T}{\varrho(P, T)} \left\{\beta^2(P, T) + \left(\frac{\partial \beta(P, T)}{\partial T}\right)_P\right\} . \qquad (12)$$

Use of the above set of relations enables one to estimate the values of the elastic constants of a solid at high pressure without *a priori* knowledge of the compressibility of the substance.

## 4. General Iterative Scheme

In general the iterative scheme proposed here attempts to obtain self-consistent estimates of $\lambda(l, m, n, P, T)$ in the three principal directions, i.e. $\lambda(1, 0, 0, P, T)$, $\lambda(0, 1, 0, P, T)$, and $\lambda(0, 0, 1, P, T)$ at pressure $P$ and temperature $T$. The scheme presented below assumes the following:

(i) The temperature dependence of the linear expansion coefficients are known at one atmosphere.

(ii) The value of specific heat is known as a function of temperature at one atmosphere.

(iii) An adequate number of velocity measurements are made to extract information about the elastic property of the solid as a function of pressure at more than two temperatures.

At $P = 1$ and within the experimental range of temperature, all the quantities in the above set of 12 relations are known; no iteration is required to estimate the values of the elastic constants of a solid as a function of temperature at one atmosphere.

At the next higher pressure all the quantities in the above set of relations are known except those involving $\lambda(l, m, n, P, T)$. The iterative procedure developed here yields a self-consistent estimate of the elastic constants of a solid and also the pressure and temperature derivatives of the linear thermal expansion and of the specific heat of the solid at a pressure $P$ and temperature $T$. To initiate the procedure, we set

$$\left(\frac{\partial\chi^T(l, m, n, P, T)}{\partial T}\right)_P = \left(\frac{\partial\chi^T(l, m, n, P_1, T)}{\partial T}\right)_{P_1} \tag{13}$$

and

$$\left(\frac{\partial\beta(l, m, n, P, T)}{\partial T}\right)_P = \left(\frac{\partial\beta(l, m, n, P_1, T)}{\partial T}\right)_{P_1}, \tag{14}$$

where $P_1$ is the preceding value of pressure. These enable us to compute $\beta(l, m, n, P, T)$ and $C_P(P, T)$.

Next we set

$$\lambda(l, m, n, P, T) = \lambda(l, m, n, P_1, T)\,, \tag{15}$$

enabling us to compute $\varrho(P, T)$ from the relation

$$\frac{1}{\varrho(P, T)}\left(\frac{\partial\varrho(P, T)}{\partial P}\right)_T = \chi^T(P, T) = \chi^T(1, 0, 0, P, T) + \chi^T(0, 1, 0, P, T) + \\ + \chi^T(0, 0, 1, P, T)\,. \tag{16}$$

Equation (16) is obtained by expressing $\chi^T(P, T)$ in terms of the three principal isothermal linear compressibilities through equation (6). $L(l, m, n, P, T)$ is computed with the help of both equation (15) and the definition of $\lambda(l, m, n, P, T)$ given in the section dealing with notation. These with the known values of $\tau(l, m, n, J, P, T)$ enable us to estimate $C^S_{pq}(P, T)$ from equations (2). The use of relations (1), (4), (11), and (12) together with the assumptions (13) and (14) provide estimates of the values of $S^T_{pq}(P, T)$. From these estimates of $S^T_{pq}(P, T)$ we obtain $\chi^T(l, m, n, P, T)$, which by relation (10) yields a new estimate of $\lambda(l, m, n, P, T)$. If the new values of $\lambda(l, m, n, P, T)$ in the three principal directions agree with their respective values assumed at the beginning of the iteration, the estimated values of the elastic constants are correct and consistent with the assumptions represented by relations (13) and (14). If these values of $\lambda(l, m, n, P, T)$ do not agree with the previously assumed values iteration is repeated with these new values of $\lambda(l, m, n, P, T)$ as starting values and all the related quantities are recalculated. This process repeats until two consecutive estimates of $\lambda(l, m, n, P, T)$ in the three respective principal directions are equal in magnitude at the pressure $P$ and a temperature $T$. The complete procedure is carried out at the pressure $P$ and at all the temperatures at which the travel-time measurements were made. When all the elastic constants of

Table 1

A flow chart of the iterative scheme to estimate the values of the elastic constants of a solid as a function of pressure and temperature.

$A(l, m, n, P, T) = [\partial\chi^{\mathrm{T}}(l, m, n, P, T)/\partial T]_P$ and $B(l, m, n, P, T) = [\partial\beta(l, m, n, P, T)/\partial T]_P$

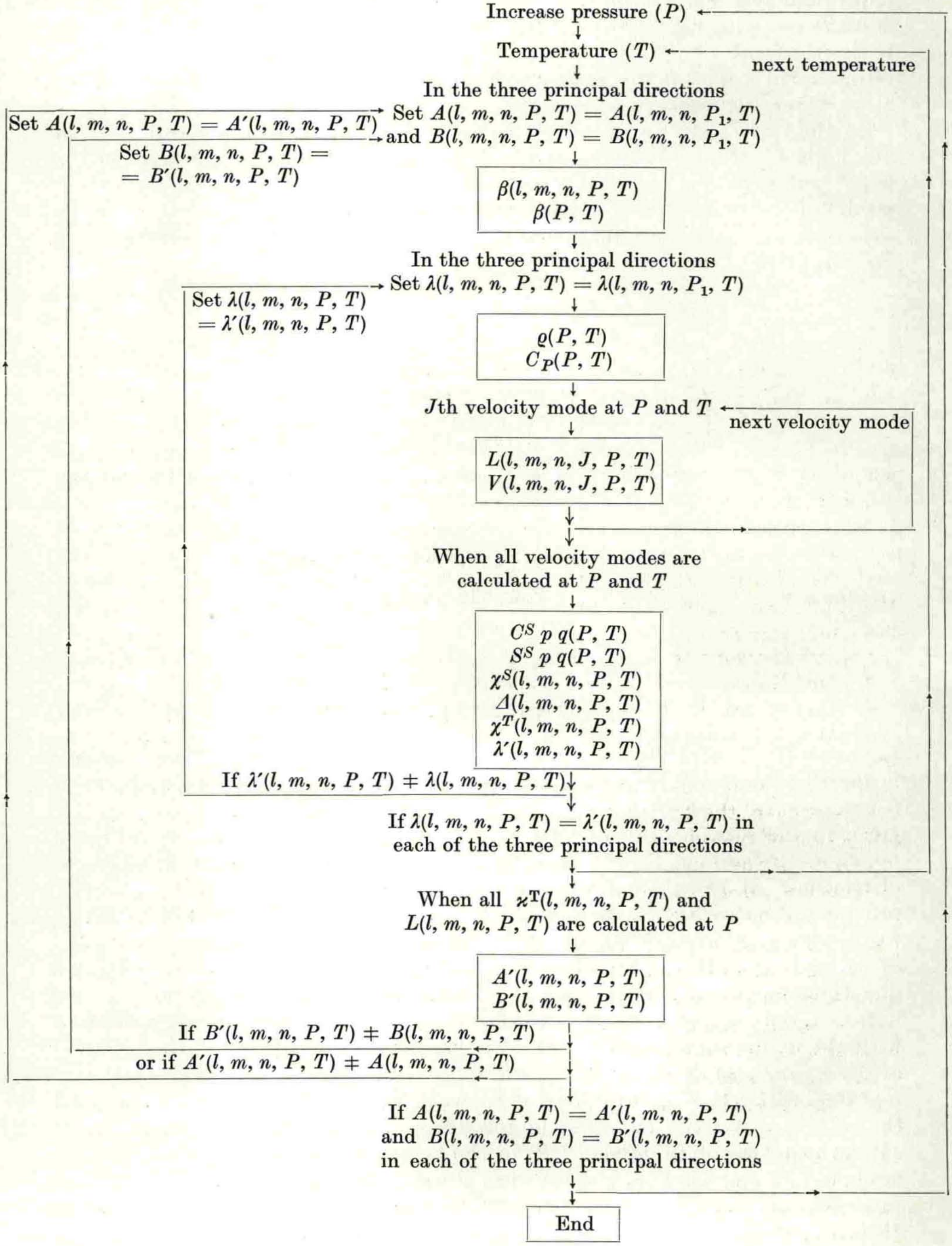

Table 2

Pressure derivatives of the adiabatic elastic constants of lead at 296 and 195 °K as obtained by Miller and Schuele (M & S) and by the iterative procedure (IP)

| | 296 °K | | 195 °K | |
|---|---|---|---|---|
| | M&S | IP | M&S | IP |
| $C_{11}$ | 5.94 | 6.02 | 5.82 | 5.90 |
| $C_{12}$ | 5.33 | 5.40 | 5.26 | 5.34 |
| $C_{44}$ | 2.06 | 2.10 | 1.97 | 2.01 |
| $B_S$ | 5.53 | 5.61 | 5.45 | 5.50 |

a solid are estimated in the above manner at the pressure $P$ as a function of temperature, we obtain new estimates of $[\partial\chi^T(l, m, n, P, T)/\partial T]_P$ and $[\partial\beta(l, m, n, P, T)/\partial T]_P$. If these new values of $[\partial\chi^T(l, m, n, P, T)/\partial T]_P$ and $[\partial\beta(l, m, n, P, T)/\partial T]_P$ in the three principal directions agree with their respective assigned values at the beginning of the calculation the estimated values of the elastic constants at the pressure $P$ and all the temperatures are considered correct. If these new values of $[\partial\chi^T(l, m, n, P, T)/\partial T]_P$ and/or $[\partial\beta(l, m, n, P, T)/\partial T]_P$ do not agree with their previously assigned values the iterative procedure is repeated all over again with these new values as the starting point, until two consecutive estimates of $[\partial\chi^T(l, m, n, P, T)/\partial T]_P$ and $[\partial\beta(l, m, n, P, T)/\partial T]_P$ agree with their respective values in all the three principal directions. (See the flow chart in Table 1).

The net outcome includes

(i) the values of the elastic constants as a function of pressure and temperature,

(ii) the values of the partial temperature and pressure derivatives of the linear thermal expansion coefficient as a function of pressure and temperature, and

(iii) the pressure derivative of the specific heat as a function of temperature.

If, however, the transit time of the elastic waves in a solid is measured as a function of pressure at only two temperatures, then the values of the elastic constants of the solid may be estimated at high pressure by interpreting $P_1$ in relation (14) to signify some lower pressure at which these quantities are known, and by omitting the iterations on $[\partial\beta(l, m, n, P, T)/\partial T]_P$ in the above scheme. Lastly, if transit times of the elastic waves in a solid are measured as a function of pressure at only one temperature, then the values of the elastic constants may be estimated at high pressures by interpreting $P_1$ in both relations (13) and (14) to signify some lower pressures at which these quantities are known, and by omitting the iteration on $[\partial\chi^T(l, m, n, P, T)/\partial T]_P$ and $[\partial\beta(l, m, n, P, T)/\partial T]_P$ in the above scheme.

We have applied the iterative procedure described above to estimate the values of the elastic constants from the ultrasonic data for lead (a cubic solid) at 296 and 195 °K to 3 kbar [6], for rutile (a tetragonal solid) at 298 °K to 7.5 kbar [7], and for calcite (a trigonal solid) at 298 °K to 6 kbar [8]. Table 2 compares the values of the pressure derivatives of adiabatic elastic constants of lead at 296 and 195 °K as calculated by Miller and Schuele and by the itera-

Table 3

Adiabatic elastic constants (in units of $10^{11}$ dyn/cm$^2$) of calcite with pressure at 298 °K as obtained by Dandekar (D) [8] and as obtained by the iterative procedure (IP) from the same ultrasonic data

| Pressure (kbar) | $C_{11}$ | | $C_{33}$ | | $C_{44}$ | | $C_{66}$ | | $C_{14}$ | | $C_{13}$ | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | D | IP | D | IP | D | IP | D | IP | D | IP | D | IP |
| | ±0.05 | | ±0.05 | | ±0.02 | | ±0.07 | | ±0.06 | | ±0.33 | |
| 0.001 | 14.626 | 14.626 | 8.531 | 8.531 | 3.405 | 3.405 | 4.328 | 4.328 | −2.076 | −2.076 | 5.076 | 5.076 |
| 2.0 | 14.650 | 14.651 | 8.526 | 8.527 | 3.453 | 3.453 | 4.335 | 4.336 | −2.078 | −2.080 | 5.190 | 5.179 |
| 4.0 | 14.674 | 14.676 | 8.522 | 8.522 | 3.468 | 3.468 | 4.342 | 4.343 | −2.101 | −2.100 | 5.419 | 5.402 |
| 6.0 | 14.697 | 14.700 | 8.518 | 8.516 | 3.449 | 3.448 | 4.349 | 4.350 | −2.137 | −2.138 | 5.757 | 5.734 |

Sources of the values of thermodynamic parameters used in the I.P: Thermal expansion coefficients from [10], temperature derivative of the compressibilities at one atmosphere from [11], specific heat value from [12].

Table 4

Adiabatic elastic constants (in kbar) of single crystal rutile with pressure at 298 °K obtained by Manghanani (M) [7] and by the iterative procedure (IP) from Manghanani's data

| Pressure (kbar) | $C_{11}$ | | $C_{33}$ | | $C_{44}$ | | $C_{66}$ | | $C_{12}$ | | $C_{13}$ | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | M | IP | M | IP | M | IP | M | IP | M | IP | M | IP |
| 0.001 | 2714.3 | 2714.3 | 4839.5 | 4839.5 | 1244.3 | 1244.3 | 1947.7 | 1947.7 | 1779.6 | 1779.6 | 1495.7 | 1495.7 |
| 1.25 | 2722.4 | 2722.4 | 4849.9 | 4850.8 | 1245.7 | 1245.2 | 1955.7 | 1955.7 | 1791.0 | 1791.0 | 1502.0 | 1503.4 |
| 2.50 | 2730.5 | 2730.4 | 4860.3 | 4862.1 | 1247.1 | 1246.3 | 1963.8 | 1963.7 | 1802.3 | 1802.3 | 1508.3 | 1509.6 |
| 5.00 | 2746.7 | 2746.6 | 4881.2 | 4884.7 | 1249.8 | 1248.4 | 1979.9 | 1979.7 | 1825.1 | 1825.1 | 1520.8 | 1522.2 |
| 7.50 | 2762.8 | 2762.8 | 4902.0 | 4907.3 | 1252.6 | 1250.5 | 1996.0 | 1995.9 | 1847.8 | 1847.8 | 1533.4 | 1534.8 |

Sources of the values of thermodynamic parameters: Same as those used by Manghanani [7], i.e. thermal expansion coefficients from [13], specific heat value from [14], and temperature derivatives of the compressibilities at 1 atm [7].

tive procedure. The required thermodynamic data used in the calculations here are those quoted in Table 1 of the paper by Miller and Schuele [6]. The differences in the values of the pressure derivatives are small but real. These differences can become significant at high pressures. In the course of calculation it was also found that the temperature derivatives of the volume compressibility of lead did not change significantly with pressure to 3 kbar. For example, the values at 1 bar and 3 kbar are $0.00013 \times 10^{-11}$ and $0.00012 \times 10^{-11}$ cm$^2$/dyn deg. The pressure derivatives of the specific heat at both 296 and 195 °K are similarly negligible.

Tables 3 and 4 present the adiabatic elastic constants of calcite and rutile as a function of pressure at 298 °K. These tables indicate that the two sets of estimates for these constants are in good agreement. Hence, they suggest that the ultrasonic travel-time data as a function of pressure at 298 °K, thermal expansion coefficients data, and the ultrasonic data as a function of temperature at one atmosphere and the specific heat value at 298 °K on the one hand, and Bridgman's compressibility measurements on calcite and rutile on the other, are consistent. The values of $a$ and $b$ appear in Bridgman's equation

$$\frac{V - V_0}{V_0} = -a\,P + b\,P^2 \,. \tag{17}$$

We present the values of $a$ and $b$ for calcite as obtained from the iterative procedure and those determined by Bridgman [9] in Table 5. The differences in the estimates of $b$ are evident. Since at low pressures their effects are negligible, the two sets of elastic constants values in Table 3 agree remarkably with each other. A similar statement may be made for rutile.

These calculations demonstrate that the iterative procedure presented in this paper accurately estimates the values of the elastic constants of a solid at high pressure from the ultrasonic measurements when the concomitant compressibility measurements are either unavailable or unreliable. Moreover, the procedure provides an indirect method by which to compute the values of specific heats and linear thermal expansion coefficients as a function of pressure and temperature provided the ultrasonic measurements are made as a function of pressure at more than two temperatures. The accuracy of these estimates is limited only by the precision of the sound measurements as a function of pressure and temperature and of linear thermal expansion coefficients and specific heat measurements as a function of temperature at one atmosphere. If and when it becomes

Table 5

The coefficients $a$ and $b$ in Bridgman's equation of state (17) (units of $a$ are $10^{-12}$ cm$^2$/dyn and units of $b$ are $10^{-24}$ cm$^2$/dyn) for calcite at 298 °K

| Compressibility | Present work | | Bridgman | |
|---|---|---|---|---|
| | $a$ | $b$ | $a$ | $b$ |
| 100 | 0.277 | 0.60 | 0.273 | 0.24 |
| 001 | 0.847 | 3.6 | 0.822 | 2.9 |
| Volume | 1.401 | 5.98 | 1.367 | 3.9 |

possible to measure the values of linear thermal expansion coefficients and specific heats of a solid at high pressure with precision, the present procedure would continue to provide us with a check for the consistency of these measurements with the other relevant data on the solid.

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