

and

$$T'_{12} = 2C_{44}\beta_{12} \quad (18)$$

after having gone through a long algebra, we find the expressions for the effective second-order elastic constants as follows:

$$\begin{aligned} C_{11} = & c_{11} + \eta(2c_{11} + 2c_{12} + c_{111} + 2c_{112}) \\ & + \eta^2(-\frac{3}{2}c_{11} - 2c_{12} + \frac{3}{2}c_{111} + 5c_{112} + c_{123} \\ & + \frac{1}{2}c_{1111} + 2c_{1112} + c_{1122} + c_{1123}) \end{aligned} \quad (19)$$

$$\begin{aligned} C_{12} = & c_{12} + \eta(-c_{11} - c_{12} + 2c_{112} + c_{123}) \\ & + \eta^2(c_{11} + \frac{3}{2}c_{12} - \frac{1}{2}c_{111} - c_{112} \\ & + c_{1112} + c_{1122} + \frac{5}{2}c_{1123}) \end{aligned} \quad (20)$$

$$\begin{aligned} C_{44} = & c_{44} + \eta(c_{11} + 2c_{12} + c_{44} + c_{144} + 2c_{166}) \\ & + \eta^2(-c_{11} - 2c_{12} - \frac{1}{2}c_{44} + \frac{1}{2}c_{111} \\ & + 3c_{112} + c_{123} + c_{144} + 2c_{166} \\ & + \frac{1}{2}c_{1144} + c_{1155} + 2c_{1255} + c_{1266}). \end{aligned} \quad (21)$$

Where $c_{\mu\nu}$, $c_{\mu\nu\lambda}$ and $c_{\mu\nu\lambda\xi}$ are the second-, third- and fourth-order elastic constants of crystal in Voigt's notation, respectively, and they are expressed in accordance with the thermodynamic definition[3].

It may be noted that, in equations (19–21), the coefficients of the terms in η with the second- and third-order elastic constants are the conventional expressions for the effective elastic constants[1, 4–6] and they agree with those derived initially by Birch[1] when the third-order elastic constants in Birch's definition are converted into those of more general thermodynamic definition.* However, the coefficients of the terms in η^2 in equations (20) and (21) are at variance with ones given by

Ghate[6]. In light of the present analysis, the writer believes that the *minus* signs of the quantities c_{11} and c_{12} found in the η^2 term of Ghate's equation (23) should have been *plus* signs. And, as for the expression for C_{44} , the quantity $(+\frac{1}{2}c_{1144})$ should be found in the η^2 term of Ghate's equation (24).

3. THE ULTRASONIC EFFECTIVE ELASTIC CONSTANTS

The expressions of the effective elastic constants as given by equations (19–21) can be either the adiabatic or isothermal expressions, and the proper designation of these is obviously done by adding the proper superscript either 's' or 'T' to all the elastic constants. The acoustic data resulting from the usual acoustic experiments with pressure are neither thermodynamically adiabatic nor thermodynamically isothermal quantities, but they are 'thermodynamically mixed' isothermal-adiabatic quantities[7]. Thus, in this section, we seek for the expressions of the effective elastic constants that may be resulting from the ultrasonic-pressure experiments at high pressures.

Recalling the usual behaviors of ultrasonic wave velocities in the medium of a cubic crystal[8, 9], we note that a longitudinal stiffness c_{11} and shear stiffness c_{44} result directly from measurements of the longitudinal and transverse wave velocities in the [001] direction of the crystal, respectively. If one measures a transverse wave velocity in [110] polarized in the $[1\bar{1}0]$ direction, the resulting stiffness constant is $(c_{11} - c_{12})/2$. Thus, from this, one finds immediately the elastic constant c_{12} as a typical procedure. Following exactly the same procedure as the above but subjected to hydrostatic pressure, we find the *ultrasonic* effective elastic constants of cubic crystals as:

$$\begin{aligned} C_{11(\text{ultrasonic})} = & c_{11}^s + \eta(c_{11}^s + C_a^m + 3B^T) \\ & + \eta^2(-\frac{1}{2}c_{11}^s + C_a^m + \frac{1}{2}C_a^m \\ & + C_e^m + \frac{1}{2}C_a^T + C_b^T - 3B^T) \end{aligned} \quad (22)$$

*The relations between the $c_{\mu\nu\lambda}$ defined by Brügger ($c_{\mu\nu\lambda}^{Br}$) and those by Birch ($c_{\mu\nu\lambda}^{Bi}$) are: $c_{111}^{Br} = 6c_{111}^{Bi}$, $c_{112}^{Br} = 2c_{112}^{Bi}$, $c_{123}^{Br} = c_{123}^{Bi}$, $c_{456}^{Br} = \frac{1}{2}c_{456}^{Bi}$, $c_{144}^{Br} = \frac{1}{2}c_{144}^{Bi}$, and $c_{166}^{Br} = \frac{1}{2}c_{166}^{Bi}$. It is noted that the relation between Birch's c_{456} and Brügger's c_{456} should be as given in this paper, provided c_{456} term in the expression of the strain energy is $[\frac{1}{2}c_{456}(\eta_{12}\eta_{23}\eta_{31} + \eta_{21}\eta_{32}\eta_{13})]$. However, if the term in the expression of the strain energy is $[c_{456}(\eta_{12}\eta_{23}\eta_{31} + \eta_{21}\eta_{32}\eta_{13})]$ as in Birch's original paper (e.g. equation 12 of [1]), the relation should be $c_{456}^{Br} = \frac{1}{4}c_{456}^{Bi}$.

$$C_{12}(\text{ultrasonic}) = c_{12}^s + \eta(c_{12}^s + C_b^m - 3B^T) + \eta^2(-\frac{1}{2}c_{12}^s + C_b^m + C_e^m + \frac{3}{2}c_{1123}^m - \frac{1}{2}C_a^T - C_b^T + 3B^T) \quad (23)$$

$$C_{44}(\text{ultrasonic}) = c_{44} + \eta(c_{44} + C_c^m + 3B^T) + \eta^2(-\frac{1}{2}c_{44} + C_c^m + \frac{1}{2}C_f^m + C_g^m + \frac{1}{2}C_a^T + C_b^T - 3B^T). \quad (24)$$

Where $B^T = (c_{11}^T + 2c_{12}^T)/3$ and the C_j are

$$C_a = c_{111} + 2c_{112} \quad (25)$$

$$C_b = 2c_{112} + c_{123} \quad (26)$$

$$C_c = c_{144} + 2c_{166} \quad (27)$$

$$C_d = c_{1111} + 2c_{1112} \quad (28)$$

$$C_e = c_{1112} + c_{1122} + c_{1123} \quad (29)$$

$$C_f = c_{1144} + 2c_{1155} \quad (30)$$

$$C_g = 2c_{1255} + c_{1266}. \quad (31)$$

The superscripts 's', 'T' and 'm' designate *thermodynamically adiabatic*, *thermodynamically isothermal*, and *thermodynamically mixed* elastic constants, respectively. Since the C_j are related to the pressure derivatives of the linear elastic constants $c_{\mu\nu}$, these relationships are to be found.

4. RELATION OF PRESSURE DERIVATIVES OF THE EFFECTIVE ELASTIC CONSTANTS TO PARTIAL CONTRACTIONS OF THE HIGHER-ORDER ELASTIC CONSTANTS

The pressure-dependent second-order elastic constants are [7, 10]

$$C_{ijkl}^s(P) = \frac{\lambda}{V^0} \left[\frac{\partial^2 U(V^0, S, \tilde{\eta})}{\partial \eta_{ij} \partial \eta_{kl}} \right]_{\substack{S = \text{const.} \\ \tilde{\eta} = \epsilon^1}} + PD_{ijkl} \quad (32)$$

where $D_{ijkl} = \delta_{ij}\delta_{kl} - \delta_{ik}\delta_{jl} - \delta_{il}\delta_{jk}$. V denotes the volume of crystal at reference state characterized by the hydrostatic pressure P , and $\tilde{\eta}$ is the strain tensor corresponding to an arbitrarily deformed state characterized by that pressure P . V^0 is defined by the relation $(V/V^0) = \lambda^3$, where λ is a factor given by the

coordinates of a material point in two reference states a_i and a_i^0 according to $(a_i/a_i^0) = \lambda$. The Lagrangian strain tensors corresponding to these two reference states are η_{ij} and η_{ij}^0 , and they are related by

$$\eta_{ij}^0 = \lambda^2 \eta_{ij} + \epsilon \delta_{ij}$$

where $\epsilon = \frac{1}{2}(\lambda^2 - 1)$. Since from thermodynamics $(\partial/\partial P)_T = -(V/B_T)(\partial/\partial V)_T$ and $(\partial\lambda/\partial V)_0 = \frac{1}{3}V^0$, we find by differentiating equation (32) that

$$\left(\frac{\partial C_{ijkl}^s}{\partial P} \right)_T = -\frac{1}{3B^T} \left[\frac{1}{V^0} \left\{ \frac{\partial^2 U(V^0, S, \tilde{\eta})}{\partial \eta_{ij} \partial \eta_{kl}} \right\}_{\substack{V^0 \\ S = \text{const.} \\ \tilde{\eta} = 0}} + \frac{1}{V^0} \left\{ \frac{\partial}{\partial \eta_{mm}} \left(\frac{\partial^2 U(V^0, S, \tilde{\eta})}{\partial \eta_{ij} \partial \eta_{kl}} \right) \right\}_{\substack{V^0 \\ S = \text{const.} \\ \tilde{\eta} = 0}} \right] + D_{ijkl}. \quad (33)$$

Note that the first term in equation (33) is by definition the zero-pressure second-order elastic constants. The second term is, however, thermodynamically mixed third-order elastic constants at zero pressure. Thus, from equation (33), it follows that [7]

$$\left(\frac{\partial C_{ijkl}^s}{\partial P} \right)_T = -\frac{1}{3B^T} [C_{ijkl}^s + C_{ijklmm}^m] + D_{ijkl} \quad (34)$$

where

$$C_{ijklmm}^m = \frac{1}{A} \left[C_{ijklmm}^s + T\gamma_G \left\{ -\beta C_{ijkl}^s + 3 \left(\frac{\partial C_{ijkl}^s}{\partial T} \right) \right\} \right]. \quad (35)$$

γ_G is the Grüneisen constant, β is the coefficient of volume expansion, and A is the ratio of the adiabatic bulk modulus to the isothermal bulk modulus and it is given by $A = 1 + T\beta\gamma_G$. The quantities given by equation (35) are the primary experimental quantities which result from the usual ultrasonic-pressure experiments at low pressures. For cubic crystals, equation (35) reduces to: