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SOME INVARIANCE RELATIONS FOR ELASTIC WAVES IN CRYSTALS*

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Second-order and third-order elastic coefficients of crystals can be determined by using the experimental data of the mass density and of the ultrasonic velocities and their changes by hydrostatic pressure or uniaxial compression, respectively. The correct values of the elastic coefficients can be determined only in the case when the experimental values of the ultrasonic velocities and their changes fulfil the invariance relations. In the present paper it is shown for which propagation and polarization directions invariance relations are fulfilled for crystals of cubic, tetragonal and hexagonal symmetry.

НЕКОТОРЫЕ ИНВАРИАНТНЫЕ СООТНОШЕНИЯ ДЛЯ УПРУГИХ ВОЛН В КРИСТАЛЛАХ

Коэффициенты упругости 2-го и 3-его порядков для кристаллов могут быть определены на основе экспериментальных данных о плотности, скоростях упругих волн и их изменений в образцах, подвергнутых гидростатическому давлению или одноосному сжатию. Правильные значения коэффициентов упругости 2-го и 3-его порядков можно определить только в случае, когда экспериментальные значения скоростей и их изменения удовлетворяют инвариантным соотношениям. В работе показано, для каких направлений распространения и поляризации упругих волн в кристаллах с кубической, тетрагональной и гексагональной решетками выполняются инвариантные соотношения.

I. INTRODUCTION

The determination of second-order elastic coefficients (S.O.E. coefficients) in crystals by using ultrasonic methods requires that we know the mass density and ultrasonic velocities in selected propagation and polarization directions in the natural state [1]. The results obtained in this way are reported by a number of authors and often for the same kind of crystals; however, there are disagreements between [2] and [3].

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Thurston and Brugger [4] developed relations the mass density, natural velocity and their changes by hydrostatic pressure or uniaxial compression and the third-order elastic coefficients (T.O.E. coefficients). These relations are expressed in [5] for the seven Laue groups: O, TI, CII, CI, RI, HII and HI. The relations are applied in [6] for NaCl and in [7] for $LiNbO_3$.

With respect to the Christoffel equation and to the results of [1] and [5] we show that in crystals of cubic, tetragonal and hexagonal symmetry there exist some invariants for S.O.E. coefficients and T.O.E. coefficients.

Our results published in [3] and [8] are extended in the present paper for the case of uniaxial compression.

II. BASIC RELATIONS

We consider an acoustic wave in the natural state of the crystal in the form

$$U_i = U_{i0} \exp\left\{j\omega\left(t - \frac{X_k N_k}{W}\right)\right\}, \tag{1}$$

where t is time, U_{i0} is the amplitude, ω is the angular frequency, X_k are coordinates, N_k are components of the unit vector in the propagation direction $\boldsymbol{N}$ and W is the ultrasonic natural phase velocity. (In relations (1) and in the following summation on repeated subscription is implied from 1 to 3).

Substitution of a plane wave (1) into the equation of motion gives the well-known Christoffel's equation in the form

$$(w_{ik} - \varrho_0 W^2 \delta_{ik}) U_{ik} = 0, \tag{2}$$

where δ_{ik} is Kronecker's tensor,

$$w_{ik} = C^S_{ijkl} N_j N_l \tag{3}$$

is the symmetric tensor of the second rank, ϱ_0 is mass density in the natural state and C^S_{ijkl} are isentropic S.O.E. coefficients in the tensor notation.

For any directions $\boldsymbol{N}$ equation (2) represents three equations for $i = 1, 2, 3$. The non-trivial roots of these equations are given by the equation

$$\begin{vmatrix} w_{11} - \varrho_0 W^2 & w_{12} & w_{13} \\ w_{21} & w_{22} - \varrho_0 W^2 & w_{23} \\ w_{31} & w_{32} & w_{33} - \varrho_0 W^2 \end{vmatrix} = 0 \tag{4}$$

For each propagation direction $\boldsymbol{N}$ we obtain from equation (4) three eigenvalues $\varrho_0 W_L^2$, $\varrho_0 W_{T_1}^2$ and $\varrho_0 W_{T_2}^2$, where W_L, W_{T_1} and W_{T_2} are one quasilongitudinal and two quasitransversal velocities of acoustic waves.

The first basic invariant of the symmetric tensor (3) in the direction $\boldsymbol{N}$ is

$$(w_{11} + w_{22} + w_{33})_N = (w_{ii})_N = \varrho_0 (W_L^2 + W_{T_1}^2 + W_{T_2}^2)_N. \tag{5}$$

We can show by using (3) and (5) that in crystals of cubic, tetragonal and hexagonal symmetry there exist propagation directions satisfying the equation

$$(w_{11} + w_{22} + w_{33})_{N_1} = (w_{11} + w_{22} + w_{33})_{N_2} \tag{6}$$

or

$$(W_L^2 + W_{T_1}^2 + W_{T_2}^2)_{N_1} = (W_L^2 + W_{T_1}^2 + W_{T_2}^2)_{N_2}, \tag{6a}$$

although in general for $\boldsymbol{N}_1 \neq \boldsymbol{N}_2$ there is fulfilled

$$(W_L)_{N_1} \neq (W_L)_{N_2}, \quad (W_{T_1})_{N_1} \neq (W_{T_1})_{N_2}, \quad (W_{T_2})_{N_1} \neq (W_{T_2})_{N_2}.$$

In the following the relations (6) and (6a) will be called invariances for S.O.E. coefficients.

The formulae of Thurston and Brugger [4] conveniently express T.O.E. coefficients from the stress derivatives of $\varrho_0 W^2$-eigenvalues at zero static stress. Their expression is

$$(\varrho_0 W^2)'_0 = \frac{\partial}{\partial p} (\varrho_0 W^2)_{p=0} \sim 2\varrho_0 W \frac{\partial W}{\partial p}, \tag{7}$$

depending only on a single parameter p-stress. Their general formula for the cases of hydrostatic pressure (indices HP) is

$$-(\varrho_0 W^2)'_0 = 1 + 2wF_{HP} + G_{HP} \tag{8}$$

and for uniaxial compression (indices UC)

$$-(\varrho_0 W^2)'_0 = \boldsymbol{N} \cdot \boldsymbol{M} + 2wF_{UC} + G_{UC}, \tag{9}$$

where

$$F_{HP} = S^T_{aars} U_r U_s \tag{8a}$$
$$G_{HP} = S^T_{aauv} C_{uvprqs} N_p N_q U_r U_s \tag{8b}$$
$$F_{UC} = S^T_{abrs} M_a M_b U_r U_s \tag{9a}$$
$$G_{UC} = S^T_{abuv} C_{uvprqs} M_a M_b N_p N_q U_r U_s \tag{9b}$$
$$w = (\varrho_0 W^2)_0 = C^S_{prqs} N_r N_s U_p U_q, \tag{10}$$

where S^T_{abrs} are the second-order isothermal compliances, C_{uvprqs} are T.O.E. Coefficients defined as the isothermal strain derivatives of isentropic S.O.E. coefficients, $\boldsymbol{M}$, $\boldsymbol{N}$ and $\boldsymbol{U}$ are unit vectors along the directions of compression, propagation and polarization in the absence of a static stress or static compression — in the natural state.

By using equations (8) and (9) we can get in deformed crystals by hydrostatic pressure or uniaxial compression the directions of propagation $\boldsymbol{N}_1 \neq \boldsymbol{N}_2$ for which there is valid

$$[(\varrho_0 W_L^2)' + (\varrho_0 W_{T_1}^2)' + (\varrho_0 W_{T_2}^2)']_{N_1} = [(\varrho_0 W_L^2)' + (\varrho_0 W_{T_1}^2) + (\varrho_0 W_{T_2}^2)']_{N_2} \quad (11)$$

or

$$\left[W_L \frac{\partial W_L}{\partial p} + W_{T_1} \frac{\partial W_{T_1}}{\partial p} + W_{T_2} \frac{\partial W_{T_2}}{\partial p}\right]_{N_1} =$$

$$= \left[W_L \frac{\partial W_L}{\partial p} + W_{T_1} \frac{c W_{T_1}}{\partial p} + W_{T_2} \frac{\partial W_{T_2}}{\partial p}\right]_{N_2}, \quad (11a)$$

although in general for $N_1 \neq N_2$ there are fulfilled

$$\left[W_L \frac{\partial W_L}{\partial p}\right]_{N_1} \neq \left[W_L \frac{\partial W_L}{\partial p}\right]_{N_2} \quad (12)$$

$$\left[W_{T_1} \frac{\partial W_{T_1}}{\partial p}\right]_{N_1} \neq \left[W_{T_1} \frac{\partial W_{T_1}}{\partial p}\right]_{N_2}$$

and

$$\left[W_{T_2} \frac{\partial W_{T_2}}{\partial p}\right]_{N_1} \neq \left[W_{T_2} \frac{\partial W_{T_2}}{\partial p}\right]_{N_2}.$$

In the following the relations (11) and (11a) will be called invariances for T.O.E. coefficients.

III. EXPLICIT SOLUTION OF INVARIANCES FOR SECOND-ORDER ELASTIC COEFFICIENTS

a) The Cubic system — the Laue groups CII and CI

For the first invariant of a symmetric tensor (3) in crystals of the cubic symmetry according to [3] we have

$$[w_{ii}]_N = \varrho_0 [W_L^2 + W_{T_1}^2 + W_{T_2}^2]_N = C_{11}^S + 2C_{44}^S, \quad (13)$$

where C_{11}^S and C_{44}^S are isentropic S.O.E. coefficients in the Voigt notation.

The relation (13) is correct for all crystals of cubic symmetry and its value does not depend on the propagation direction of the ultrasonic wave. From (13) it follows that equations (6) and (6a) in the case of cubic crystals are fulfilled for an arbitrary direction.

b) The tetragonal system — the Laue group TI

For the first invariant of a symmetric tensor (3) in crystals of the Laue group *TI* we have according to [3]

$$[w_{ii}]_{N\perp[001]} = \varrho_0 [W_L^2 + W_{T_1}^2 + W_{T_2}^2]_{N\perp[001]} = C_{11}^S + C_{44}^S + C_{66}^S. \quad (14)$$

The relation (14) is correct for all crystals of the Laue group for all directions of propagation perpendicular to the tetragonal axis. From (14) it follows that equations (6) and (6a) in crystals of the Laue group TI are fulfilled for an arbitrary direction perpendicular to the tetragonal axis.

c) The hexagonal system — the Laue groups HII and HI

For the first invariant of a symmetric tensor (3) in crystals of the Laue groups *H*II and *H*I we have according to [3]

$$[w_{ii}]_{N\perp[001]} = \varrho_0 [W_L^2 + W_{T_1}^2 + W_{T_2}^2]_{N\perp[001]} = \tfrac{1}{2}(3C_{11}^S - C_{12}^S + 2C_{44}^S). \quad (15)$$

The relation (15) is correct for all directions of propagation perpendicular to the hexagonal axis. From (15) it follows that equations (6) and (6a) in crystals of the Laue groups *H*II and *H*I are fulfilled in an arbitrary direction perpendicular to the hexagonal axis.

IV. EXPLICIT SOLUTION OF INVARIANCES FOR THIRD-ORDER ELASTIC COEFFICIENTS

a) The cubic system — the Laue groups CII and CI

For cubic crystals of the Laue group *C*II deformed by hydrostatic pressure we get using (8), (8a), (8b) and (10) according to [8] and [9] for the three mutual perpendicular modes U_L, U_{T_1} and U_{T_2} in the propagation direction N

$$[(\varrho_0 W_L^2)' + (\varrho_0 W_{T_1}^2)' + (\varrho_0 W_{T_2}^2)]_N' =$$

$$= -3 - \frac{1}{3B_0^T}[2(C_{11}^S + 2C_{44}^S) + C_{111} + C_{112} + C_{113} + 2(C_{144} + C_{155} + C_{166})] = C_1 \quad (16)$$

and for the Laue group CI

$$[(\varrho_0 W_L^2)' + (\varrho_0 W_{T_1}^2)' + (\varrho_0 W_{T_2}^2)']_N =$$

$$= -3 - \frac{1}{3B_0^T}[2(C_{11}^S + 2C_{44}^S) + C_{111} + 2C_{112} + 2C_{144} + 4C_{166}] = C_2 \quad (17)$$

where $B_0^T = \tfrac{1}{3}(C_{11}^T + 2C_{12}^T)$ is the isothermal bulk modulus at zero pressure, C_1 and C_2 are constants.

On the right-hand side of relations (16) and (17) there are all the quantities constant and these do not depend on the propagation direction. Therefore the left-hand side of relations (16) is equal to the constant C_1 for an arbitrary propagation direction of the Laue group *CII* and to the constant C_2 for the Laue group *CI*.

It follows from (16) and (17) that in cubic crystals deformed by hydrostatic pressure the relations (11) and (11a) are fulfilled in an arbitrary propagation direction.

In the case of uniaxial compression in the direction $\boldsymbol{M} \equiv [001]$ and all the propagation directions $\boldsymbol{N}$ perpendicular to $\boldsymbol{M}$ we have for cubic crystals

$$\begin{gathered} \boldsymbol{N} \equiv [N_1 N_2 0], \quad \boldsymbol{M} \equiv [001], \quad \boldsymbol{N} \perp \boldsymbol{M}, \quad (\boldsymbol{N} \cdot \boldsymbol{M}) = 0 \\ \boldsymbol{U}_L \equiv [U_{1L} U_{2L} 0] \\ \boldsymbol{U}_{T_1} \equiv [U_{1T_1} U_{2T_1} 0], \quad \boldsymbol{U}_{T_2} \equiv [001] \\ U_{1L} U_{2L} + U_{1T_1} U_{2T_1} = 0, \quad U_{1L}^2 + U_{2L}^2 = 1 \\ U_{1T_1}^2 + U_{2T_1}^2 = 1, \quad U_{1L}^2 + U_{1T_1}^2 = 1 \end{gathered} \tag{18}$$

and $U_{2L}^2 + U_{2T_1}^2 = 1$.

Putting conditions (18) into (9a), (9b) and (10) we get for the Laue group *CI*

$$(F_{UC})_L = S_{12}^T, \quad (F_{UC})_{T_1} = S_{12}^T, \quad (F_{UC})_{T_2} = S_{11}^T \tag{19}$$

$$[2w_L(F_{UC})_L + 2w_{T_1}(F_{UC})_{T_1} + 2w_{T_2}(F_{UC})_{T_2}]_{N \perp [001]} = 2[S_{12}^T(C_{11}^S + C_{44}^S) + S_{11}^T C_{44}^S]$$

$$\begin{aligned} (G_{UC})_L = S_{12}^T[(C_{111} + C_{112})(N_1^2 U_{1L}^2 + N_2^2 U_{2L}^2) + \\ + 2C_{155}(N_1^2 U_{2L}^2 + N_2^2 U_{1L}^2) + 4(C_{112} + C_{155}) N_1 N_2 U_{1L} U_{2L}] + \\ + S_{11}^T[C_{112}(N_1^2 U_{1L}^2 + N_2 U_{2L}^2) + 2(C_{144} + C_{123}) N_1 N_2 U_{1L} U_{2L} + C_{144}(N_1^2 U_{2L}^2 + N_2^2 U_{1L}^2)] \end{aligned} \tag{19a}$$

$$\begin{aligned} (G_{UC})_{T_1} = S_{12}^T[(C_{111} + C_{112})(N_1^2 U_{1T_1}^2 + N_2^2 U_{2T_1}^2) + \\ + 2C_{155}(N_1^2 U_{2T_1}^2 + N_2^2 U_{1T_1}^2) + 4(C_{112} + C_{155}) N_1 N_2 U_{1T_1} U_{2T_1}] + \\ + S_{11}^T[C_{112}(N_1^2 U_{1T_1}^2 + N_2^2 U_{2T_1}^2) + 2(C_{144} + C_{123}) N_1 N_2 U_{1T_1} U_{2T_1} + \\ + C_{144}(N_1^2 U_{2T_1}^2 + N_2^2 U_{1T_1}^2)] \end{aligned} \tag{19b}$$

and

$$(G_{UC})_{T_2} = S_{12}^T(C_{155} + C_{144}) + S_{11}^T C_{155}. \tag{19c}$$

Substituting (19), (19a), (19b) and (19c) into (9) and by their summation we get

$$\begin{aligned} [(\varrho_0 W_L^2)' + (\varrho_0 W_{T_1}^2)' + (\varrho_0 W_{T_2}^2)']_{N \perp [001]} = -2[S_{12}^T(C_{11}^S + C_{44}^S) + S_{11}^T C_{44}^S] - \\ - S_{12}^T(C_{111} + C_{112} + C_{144} + 3C_{155}) - S_{11}^T(C_{112} + C_{144} + C_{155}). \end{aligned} \tag{20}$$

It follows from (20) that the relations (11) and (11a) are satisfied.

Applying the data of Table 1 we can show that relation (20) is satisfied for .

$$\sum_{i=1}^{3} (\varrho_0 W_i^2)' = \sum_{i=4}^{6} (\varrho_0 W_i^2)' = \sum_{i=7}^{9} (\varrho_0 W_i^2)'.$$

The explanation of $\varrho_0 W_i^2$, where subscription $i = 1, 2, 3, 4, 5, 6, 7, 8, 9$, is listed in Table 1.

Table 1

$(\varrho_0 W^2)'$ for Laue group *CI*

i	$\boldsymbol{M}$	$\boldsymbol{N}$	$\boldsymbol{U}$	$(\varrho_0 W_i^2)'$
1	[001]	[100]	[100]	$-2S_{12}^T C_{11}^S - S_{12}^T C_{111} - PC_{112}$
2			[010]	$-2S_{12}^T C_{44}^S - S_{11}^T C_{144} - 2S_{12}^T C_{155}$
3			[001]	$-2S_{11}^T C_{44}^S - S_{12}^T C_{144} - PC_{155}$
4		[110]	[110]	$-QS_{12}^T - \frac{1}{2}[S_{12}^T C_{111} + RC_{112} + S_{11}^T(C_{123} + 2C_{144}) + 4S_{12}^T C_{155}]$
5			[110]	$-S_{12}^T(C_{11}^S - C_{12}^S) - \frac{1}{2}[S_{12}^T C_{111} + (S_{11}^T - S_{12}^T)C_{112} - S_{11}^T C_{123}]$
6			[001]	$-2S_{11}^T C_{44} - S_{12}^T C_{144} - PC_{155}$
7	[110]	[001]	[001]	$-2S_{12}^T C_{11}^S - S_{12}^T C_{111} - PC_{112}$
8			[100]	$-(S_{11}^T + S_{12}^T)C_{44}^S - \frac{1}{2}[RC_{155} + PC_{144}]$
9			[010]	as $(\varrho_0 W_8^2)'$

$P = S_{11}^T + S_{12}^T$; $Q = C_{11}^S + C_{12}^S + 2C_{44}^S$; $R = S_{11}^T + 3S_{12}^T$

b) *The tetragonal system — the Laue group TI*

For tetragonal crystals of the Laue group *TI* deformed by hydrostatic pressure we get by using (8), (8a), (8b) and (10) according to [9] for the three mutual perpendicular modes $\boldsymbol{U}_L$, $\boldsymbol{U}_{T_1}$ and $\boldsymbol{U}_{T_2} \equiv [001]$ in the propagation direction $\boldsymbol{N} \equiv [001]$

$$\begin{aligned} [(\varrho_0 W_L^2)' + (\varrho_0 W_{T_1}^2)' + (\varrho_0 W_{T_2}^2)']_{N \perp [001]} = \\ = -3 - (S_{11}^T + S_{12}^T + S_{13}^T)[2(C_{11}^S + C_{66}^S) + C_{111} + C_{112} + C_{144} + C_{155} + 2C_{166}] - \\ - (2S_{13}^T + S_{33}^T)(2C_{44}^S + C_{113} + C_{344} + C_{366}). \end{aligned} \tag{21}$$

For the tetragonal crystals of the Laue group *TI* deformed by uniaxial compression in the direction $\boldsymbol{M} \equiv [001]$ and the propagation direction $\boldsymbol{N} \equiv [N_1 N_2 0]$ we get by using (9), (9a), (9b) and (10) or the relation in Table 2

Table 2

$(\varrho_0 W_i^2)'$ for Laue group *TI*

i	$\boldsymbol{M}$	$\boldsymbol{N}$	$\boldsymbol{U}$	$(\varrho_0 W_i^2)'$
1	[001]	[100]	[100]	$-2S_{13}^T C_{11}^S - S_{13}^T(C_{111} + C_{112}) - S_{33}^T C_{113}$
2			[010]	$-2S_{13}^T C_{66}^S - 2S_{13}^T C_{166} - S_{33}^T C_{366}$
3			[001]	$-2S_{33}^T C_{44}^S - S_{13}^T(C_{144} + C_{155}) - S_{33}^T C_{344}$
4		[110]	[110]	$-S_{13}^T A - \frac{1}{2}[S_{13}^T(C_{111} + 3C_{112} + 4C_{166} + S_{33}^T(C_{113} + C_{123} + 2C_{366})]$
5			[$\bar{1}$10]	$-S_{13}^T(C_{11}^S - C_{12}^S) - \frac{1}{2}[S_{13}^T(C_{111} - C_{112}) + S_{33}^T(C_{113} - C_{123})]$
6			[001]	as $(\varrho_0 W_3^2)'$

$A = C_{11}^S + C_{12}^S + 2C_{66}^S$

$$\sum_{i=1}^{3}(\varrho_0 W_i^2)' = \sum_{i=4}^{6}(\varrho_0 W_i^2)' = -2[S_{13}^T(C_{11}^S + C_{66}^S) + S_{33}^T C_{44}^S] - \tag{22}$$
$$- S_{13}^T(C_{111} + C_{112} + C_{144} + C_{155} + 2C_{116}) - S_{33}^T(C_{113} + C_{366} + C_{344}).$$

From (22) it follows that for crystals of the Laue group *TI* the relation (11) is fulfilled.

c) The hexagonal system — the Laue group HI

For hexagonal crystals of the Laue group *HI* deformed by hydrostatic pressure we get by using (8), (8a), (8b) and (10) according to [9] for the three mutual perpendicular modes $\boldsymbol{U}_L$, $\boldsymbol{U}_{T_1}$ and $\boldsymbol{U}_{T_2} \equiv [001]$ in the propagation direction $\boldsymbol{N} \perp [001]$

$$[(\varrho_0 W_L^2)' + (\varrho W_{T_1}^2)' + (\varrho_0 W_{T_2}^2)']_{N\perp[001]} = \tag{23}$$
$$= -3 - (S_{11}^T + S_{12}^T + S_{13}^T)[3C_{11}^S - C_{12}^S + C_{111} + \tfrac{1}{2}(C_{112} + C_{222}) + C_{144} + C_{155}] -$$
$$- (2S_{13}^T + S_{33}^T)[2C_{44}^S + C_{344} + \tfrac{1}{2}(3C_{113} - C_{123})].$$

In a way similar to that in relation (18) one can show that for crystals of the hexagonal symmetry of the Laue group *HI* deformed by uniaxial compression in the direction $\boldsymbol{M} \equiv [001]$ we get for all propagation directions perpendicular to the hexagonal axis

$$[(\varrho_0 W_L^2)' + (\varrho_0 W_{T_1}^2)' + (\varrho_0 W_{T_2}^2)']_{N\perp[001]} = \tag{24}$$
$$= -S_{13}^T[3C_{11}^S - C_{12}^S + C_{111} + C_{144} + C_{155} +$$
$$+ \tfrac{1}{2}(C_{112} + C_{222})] - S_{33}^T[2c_{44}^S + \tfrac{1}{2}(3C_{113} - C_{123}) + C_{344}].$$

V. CONCLUSION

The invariance relations reported in the present paper give a possibility to clasify experimental data of ultrasonic velocities and their changes with hydrostatic pressure or uniaxial compression with respect to their applicability in calculations of S.O.E. and T.O.E. coefficients. The classification can be used in the two following situations.

1) Experimental data of the velocity and its changes with pressure measured in different crystalographic directions must satisfy relations (5) and (6) and relations (11) and (11a). If this does not occur, the measurements of the velocity and its changes with pressure are considered as distorted by experimental errors.

2) Various experimental values of the invariances reported by different authors for the same crystals in identical propagation and polarization directions, satisfying, however, formulae (5) and (6) for ultrasonic velocities and (11) and (11a) for their changes indicate various elastic properties of the investigated crystals.

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