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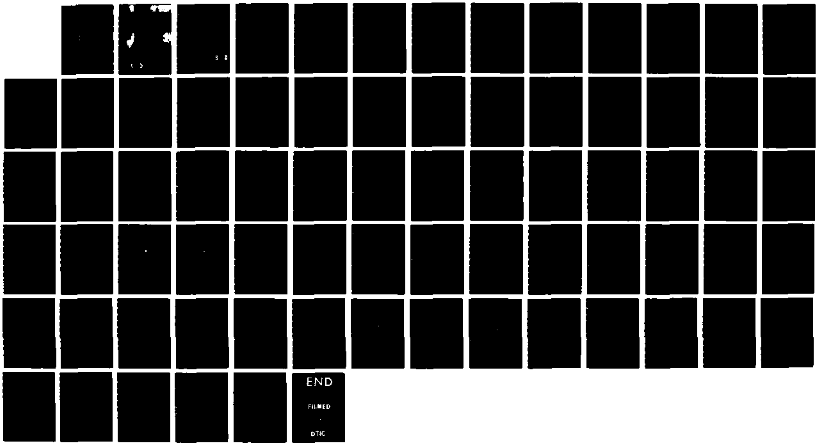

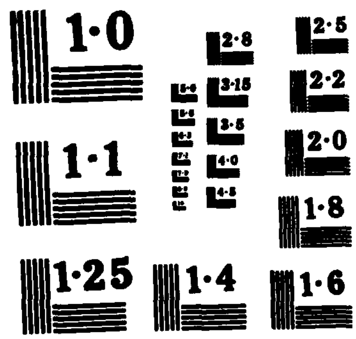
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THEORY OF THE PROPAGATION OF FINITE AMPLITUDE ULTRASONIC WAVES IN PURE MODE DIRECTIONS IN HEXAGONAL AND TRIGONAL CRYSTALS

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The nonlinear theory describing the propagation of finite amplitude ultrasonic waves in pure mode directions in hexagonal and trigonal crystals is developed. ~~in general.~~ By evaluating the coefficient of the term which couples the longitudinal wave of finite amplitude to the transverse modes, one finds the pure mode directions by requiring this coefficient to vanish. The result is that for hexagonal crystals the directions along the symmetry axis and in the basal plane ~~(studied in Technical Report No. 22)~~ are pure longitudinal mode directions. ~~In addition,~~ pure mode directions are also found for longitudinal waves → over

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propagating tangential to a cone whose apex is centered about the symmetry axis and whose apex angle is a function of the second-order elastic constants of the sample under consideration. The third-order elastic constants which determine the magnitude of the second harmonic of an initially sinusoidal ultrasonic wave are evaluated for these additional directions.

For finite amplitude longitudinal ultrasonic waves in trigonal crystals we have specialized to nonpiezoelectric crystals and have evaluated the third-order elastic constants which determine the magnitude of the second harmonic of an initially sinusoidal wave propagating in the pure mode directions: ~~The pure mode directions are:~~ (1) along the symmetry axis (the c-direction); (2) along the a-direction in the basal plane; (3) along the direction in the basal plane that makes an angle of 60° with the a-direction; and (4) along a direction in the b-c plane whose angle with respect to the b-axis is a function of the second-order elastic constants of the crystal under consideration. Keywords;

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I. INTRODUCTION

In Technical Report No. 22 we presented a theory of the propagation of finite amplitude ultrasonic waves in hexagonal crystals. In that report we presented a unique representation of the anisotropy of the nonlinearity in the basal plane of a hexagonal crystal and observed that pure mode propagation occurs along any direction in the basal plane as well as along the symmetry axis. However, in presenting the expressions for the nonlinearity parameters for wave propagation along these directions we overlooked the fact that an additional set of directions exist in which pure mode propagation can be observed. These directions are tangential to a cone whose apex angle is bisected by the symmetry axis, and whose apex angle is dependent on the second-order elastic constants. The relative complexity of these statements, the desire to present a means of defining the criteria for pure mode propagation, and the desire to analyze the nonlinear wave equation for propagation in all pure mode directions in crystals of hexagonal symmetry as well as trigonal symmetry, have necessitated our beginning this report with the fundamental definitions, in most cases the same fundamental definitions used in Technical Report No. 22. We hope that this repetition will add to clarity and make the present technical report an independent document. Although we do repeat most of the results of Technical Report No. 22 in new tabular form, we use a new perturbation approach to the derivation of the nonlinear terms, and we do not repeat the detailed equations. Thus, even though we consider the present technical report a complete document, we do not consider it a substitute for Technical Report No. 22 for scientists interested in the propagation of ultrasonic waves of finite amplitude in crystals of hexagonal symmetry. The two technical reports, then, provide information in sufficient detail for a complete understanding of nonlinearity in all pure mode directions in crystals of hexagonal symmetry as well as nonpiezoelectric pure mode directions in crystals of trigonal symmetry.

We begin the discussion with a generally applicable theory, one capable of describing crystals of any symmetry. As a matter of fact, at this point one could include virtually any physical process such as piezoelectricity or any other process, by including the appropriate terms in the expression for the strain energy. Our first simplification is to include only mechanical nonlinearities in our strain energy expression. This specializes our consideration to nonpiezoelectric materials. The second simplification is to specialize to crystals of hexagonal and trigonal symmetry. This means that our equations are somewhat more complicated than those previously used for cubic symmetry, but they still are not so complicated as to be unmanageable. The purpose of the present technical report is to develop the theory appropriate to this situation.

II. GENERAL THEORY OF NONLINEAR WAVE PROPAGATION IN CRYSTALS

Consider a point P in the medium with coordinates $a_i(a,b,c)$ in the unstrained state. Let P move to P' with coordinates $x_i(x,y,z)$ in the deformed state. The components of the displacement can then be written as

$$U = x - a$$
$$V = y - b \qquad \text{(II-1)}$$
$$W = z - c \ .$$

In the Lagrangian formulation, the strain is described in the initial, or undeformed, state and the initial coordinates of the material particle a_i are taken as independent variables. The Lagrangian formulation is used exclusively in the theory described in this technical report.

The Lagrangian strain parameters which are components of the finite strain tensor are given by Murnaghan (1) as follows:

$$\eta = \frac{1}{2}\,(J^*J - \delta) \qquad \text{(II-2)}$$

where J and J* are the Jacobian matrix and its transpose; δ is the unit matrix. They may be expressed, respectively, as follows:

$$J = \begin{vmatrix} 1 + U_a & U_b & U_c \\ V_a & 1 + V_b & V_c \\ W_a & W_b & 1 + W_c \end{vmatrix} , \qquad \text{(II-3)}$$

$$J^* = \begin{vmatrix} 1 + U_a & V_a & W_a \\ U_b & 1 + V_b & W_b \\ U_c & V_c & 1 + W_c \end{vmatrix} , \qquad \text{(II-4)}$$

and

$$\delta = \begin{vmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{vmatrix} , \qquad \text{(II-5)}$$

where $U_a = \frac{\partial U}{\partial a}$, $U_b = \frac{\partial U}{\partial b}$, etc.

The strain parameters can be written in terms of displacement gradients if we substitute (II-3)-(II-5) into (II-2). Expansion and simplification give the following expressions for η_{ij}:

$$\eta_{11} = U_a + \frac{1}{2} (U_a{}^2 + V_a{}^2 + W_a{}^2)$$

$$\eta_{12} = \eta_{21} = \frac{1}{2} (U_b + V_a + U_aU_b + V_aV_b + W_aW_b)$$

$$\eta_{13} = \eta_{31} = \frac{1}{2} (U_c + W_a + U_aU_c + V_aV_c + W_aW_c) \qquad \text{(II-6)}$$

$$\eta_{22} = V_b + \frac{1}{2} (U_b{}^2 + V_b{}^2 + W_b{}^2)$$

$$\eta_{23} = \eta_{32} = \frac{1}{2}(V_c + W_b + U_bU_c + V_bV_c + W_bW_c)$$

$$\eta_{33} = W_c + \frac{1}{2}(U_c^{\,2} + V_c^{\,2} + W_c^{\,2}) .$$

Let $\phi(\eta)$ be the strain energy per unit of undeformed volume. The properties of the crystalline medium enter into the theory through the strain energy density $\phi(\eta)$ which can be expanded and expressed as a sum of terms of different degrees in the elements of η as follows:

$$\phi = \phi_0 + \phi_1 + \phi_2 + \phi_3 + \dots . \tag{II-7}$$

In terms of the elastic constants, Eq. (II-7) can be written as

$$\phi = \phi_0 + k_1C_{ij}\eta_{ij} + k_2C_{ijk\ell}\eta_{ij}\eta_{k\ell} + k_3C_{ijk\ell mn}\eta_{ij}\eta_{k\ell}\eta_{mn} + \dots , \tag{II-8}$$

where the C's are the elastic constants and k_n ($n = 1,2,3 \dots$) is a constant factor depending on the definition of elastic constants. One does not lose generality by setting the first two terms in (II-8) equal to zero since ϕ_0 is the energy of the undeformed medium and ϕ_1 corresponds to displacement without deformation. Thus, the expression for the strain energy density becomes:

$$\phi = \phi_2 + \phi_3 + \dots = k_2C_{ijk\ell}\eta_{ij}\eta_{k\ell} + k_3C_{ijk\ell mn}\eta_{ij}\eta_{k\ell}\eta_{mn} . \tag{II-9}$$

As is well known, $C_{ijk\ell}$ are the second-order elastic (SOE) constants and $C_{ijk\ell mn}$ are the third-order elastic (TOE) constants.

In accordance with Brugger's thermodynamic definition of elastic constants (2), the factor kn is 1/n!. Therefore, based on that definition we have $k_2 = \frac{1}{2}$ and $k_3 = \frac{1}{6}$. Using a contracted-subscript notation for the elastic constants as well as using Brugger's definition of elastic constants, the general expression for ϕ can be rewritten as:

$$\phi = \frac{1}{2} C_{\lambda\mu}\eta_{ij}\eta_{k\ell} + \frac{1}{6} C_{\lambda\mu\nu}\eta_{ij}\eta_{k\ell}\eta_{mn} + \text{higher order terms} \,, \tag{II-10}$$

where $\lambda \rightarrow ij$, $\mu \rightarrow k\ell$ and $\nu \rightarrow mn$. For example, we write 1 for the pair of indices 11, 2 for 22, 3 for 33, 4 for 23 and 32, 5 for 31 and 13, 6 for 12 and 21. Summation over repeated indices is understood.

The SOE constants $C_{\lambda\mu}$ form a fourth rank tensor containing 81 components, of which 21 are independent for the most unsymmetrical triclinic crystal and the TOE constants $C_{\lambda\mu\nu}$ form a sixth rank tensor with 729 components, of which 56 are independent for the triclinic crystal. The number of elastic constants decreases considerably for crystals of higher symmetry. Eq. (II-10) for the strain energy density takes the appropriate form for crystals of different symmetry. The number of elastic constants in different crystal classes has been worked out by various authors and has been presented in a tabular form by Hearmon (3).

The equations of motion for an elastic medium, in fact, are a restatement of Newton's second law. For convenience one can introduce the stress tensor T, which is not symmetric, as

$$T = J\left(\frac{\partial\phi}{\partial\eta}\right) \tag{II-11}$$

where

$$\frac{\partial\phi}{\partial\eta} = \begin{vmatrix} \partial\phi/\partial\eta_{11} & \partial\phi/\partial\eta_{12} & \partial\phi/\partial\eta_{13} \\ \partial\phi/\partial\eta_{21} & \partial\phi/\partial\eta_{22} & \partial\phi/\partial\eta_{23} \\ \partial\phi/\partial\eta_{31} & \partial\phi/\partial\eta_{32} & \partial\phi/\partial\eta_{33} \end{vmatrix} . \tag{II-12}$$

Thus, the equa`ions of motion in Lagrangian coordinates are written as (4):

$$\frac{\partial T_{ij}}{\partial a_j} = \rho_0 \ddot{U}_i \ . \tag{II-13}$$

These equations of motion take particular forms along the a, b and c axes of the crystal:

$$\begin{aligned} \rho_0\ddot{U} &= \frac{\partial T_{11}}{\partial a} + \frac{\partial T_{12}}{\partial b} + \frac{\partial T_{13}}{\partial c} \\ \rho_0\ddot{V} &= \frac{\partial T_{21}}{\partial a} + \frac{\partial T_{22}}{\partial b} + \frac{\partial T_{23}}{\partial c} \ . \\ \rho_0\ddot{W} &= \frac{\partial T_{31}}{\partial a} + \frac{\partial T_{32}}{\partial b} + \frac{\partial T_{33}}{\partial c} \end{aligned} \tag{II-14}$$

According to Eq. (II-11), the stress matrix T can be written as:

$$\begin{vmatrix} T_{11} & T_{12} & T_{13} \\ T_{21} & T_{22} & T_{23} \\ T_{31} & T_{32} & T_{33} \end{vmatrix} = \begin{vmatrix} J_{11} & J_{12} & J_{13} \\ J_{21} & J_{22} & J_{23} \\ J_{31} & J_{32} & J_{33} \end{vmatrix} \begin{vmatrix} \frac{\partial\phi}{\partial\eta_{11}} & \frac{\partial\phi}{\partial\eta_{12}} & \frac{\partial\phi}{\partial\eta_{13}} \\ \frac{\partial\phi}{\partial\eta_{21}} & \frac{\partial\phi}{\partial\eta_{22}} & \frac{\partial\phi}{\partial\eta_{23}} \\ \frac{\partial\phi}{\partial\eta_{31}} & \frac{\partial\phi}{\partial\eta_{32}} & \frac{\partial\phi}{\partial\eta_{33}} \end{vmatrix} . \tag{II-15}$$

From (II-15) we can write down the components as:

$$T_{11} = J_{11} \frac{\partial\phi}{\partial\eta_{11}} + J_{12} \frac{\partial\phi}{\partial\eta_{21}} + J_{13} \frac{\partial\phi}{\partial\eta_{31}}$$

$$T_{12} = J_{11} \frac{\partial\phi}{\partial\eta_{12}} + J_{12} \frac{\partial\phi}{\partial\eta_{22}} + J_{13} \frac{\partial\phi}{\partial\eta_{32}}$$

$$T_{13} = J_{11} \frac{\partial\phi}{\partial\eta_{13}} + J_{12} \frac{\partial\phi}{\partial\eta_{23}} + J_{13} \frac{\partial\phi}{\partial\eta_{33}}$$

$$T_{21} = J_{21} \frac{\partial\phi}{\partial\eta_{11}} + J_{22} \frac{\partial\phi}{\partial\eta_{21}} + J_{23} \frac{\partial\phi}{\partial\eta_{31}}$$

$$T_{22} = J_{21} \frac{\partial\phi}{\partial\eta_{12}} + J_{22} \frac{\partial\phi}{\partial\eta_{22}} + J_{23} \frac{\partial\phi}{\partial\eta_{32}} \qquad \text{(II-16)}$$

$$T_{23} = J_{21} \frac{\partial\phi}{\partial\eta_{13}} + J_{22} \frac{\partial\phi}{\partial\eta_{23}} + J_{23} \frac{\partial\phi}{\partial\eta_{33}}$$

$$T_{31} = J_{31} \frac{\partial\phi}{\partial\eta_{11}} + J_{32} \frac{\partial\phi}{\partial\eta_{21}} + J_{33} \frac{\partial\phi}{\partial\eta_{31}}$$

$$T_{32} = J_{31} \frac{\partial\phi}{\partial\eta_{12}} + J_{32} \frac{\partial\phi}{\partial\eta_{22}} + J_{33} \frac{\partial\phi}{\partial\eta_{32}}$$

$$T_{33} = J_{31} \frac{\partial\phi}{\partial\eta_{13}} + J_{32} \frac{\partial\phi}{\partial\eta_{23}} + J_{33} \frac{\partial\phi}{\partial\eta_{33}}$$

The components of the Jacobin matrix in terms of displacement gradients can be expressed from (II-3):

$$J_{11} = 1 + U_a = 1 + \partial U/\partial a$$

$$J_{12} = U_b = \partial U/\partial b$$

$$J_{13} = U_c = \partial U/\partial c$$

$$J_{21} = V_a = \partial V/\partial a$$

$$J_{22} = 1 + V_b = 1 + \partial V/\partial b \qquad \text{(II-17)}$$

$$J_{23} = V_c = \partial V/\partial c$$

$$\begin{aligned}
\phi = {} & \frac{1}{2} C_{11}(\eta_{11}^2 + \eta_{22}^2 + \eta_{12}^2 + \eta_{21}^2) + C_{12}(\eta_{11}\eta_{22} - \frac{1}{2}\eta_{12}^2 - \frac{1}{2}\eta_{21}^2) \\
& + C_{13}(\eta_{11}\eta_{33} + \eta_{22}\eta_{33}) + \frac{1}{2} C_{33}\eta_{33}^2 + C_{44}(\eta_{13}^2 + \eta_{31}^2 + \eta_{23}^2 + \eta_{32}^2) \\
& + \frac{1}{6} C_{111}(\eta_{11} + \eta_{22})^3 + \frac{1}{2} C_{113}\eta_{33}(\eta_{11} + \eta_{22})^2 + \frac{1}{2} C_{133}\eta_{33}^2(\eta_{11} + \eta_{22}) \\
& + C_{144}(\eta_{11} + \eta_{22})(\eta_{23}^2 + \eta_{32}^2 + \eta_{13}^2 + \eta_{31}^2) \\
& + \frac{1}{2} C_{166}(\eta_{11} + \eta_{22})[(\eta_{11} - \eta_{22})^2 + 2(\eta_{12}^2 + \eta_{21}^2)] \qquad \text{(III-1)} \\
& + \frac{1}{6} C_{266}[(\eta_{11} - \eta_{22})^3 - 6(\eta_{12}^2 + \eta_{21}^2)(\eta_{11} - \eta_{22})] \\
& + \frac{1}{6} C_{333}\eta_{33}^3 + C_{344}\eta_{33}(\eta_{23}^2 + \eta_{32}^2 + \eta_{13}^2 + \eta_{31}^2) \\
& + \frac{1}{2} C_{366}\eta_{33}[(\eta_{11} - \eta_{22})^2 + 2(\eta_{12}^2 + \eta_{21}^2)] \\
& + 2C_{456}[(\eta_{22} - \eta_{11})(\eta_{23}^2 + \eta_{32}^2 - \eta_{13}^2 - \eta_{31}^2) + 4(\eta_{12}\eta_{23}\eta_{31} + \eta_{21}\eta_{32}\eta_{13})].
\end{aligned}$$

In this expression C_{166}, C_{266}, C_{366} and C_{456} are combinations of TOE constants given by

$$\begin{aligned}
C_{166} &= \frac{1}{4}(-2C_{111} - C_{112} + 3C_{222}) \\
C_{266} &= \frac{1}{4}(2C_{111} - C_{112} - C_{222}) \qquad \text{(III-2)} \\
C_{366} &= \frac{1}{2}(C_{113} - C_{123}) \\
C_{456} &= \frac{1}{2}(-C_{144} + C_{155}) .
\end{aligned}$$

Einspruch and Manning (8) derived an expression by using Birch's definition of TOE constants. That expression can easily be converted

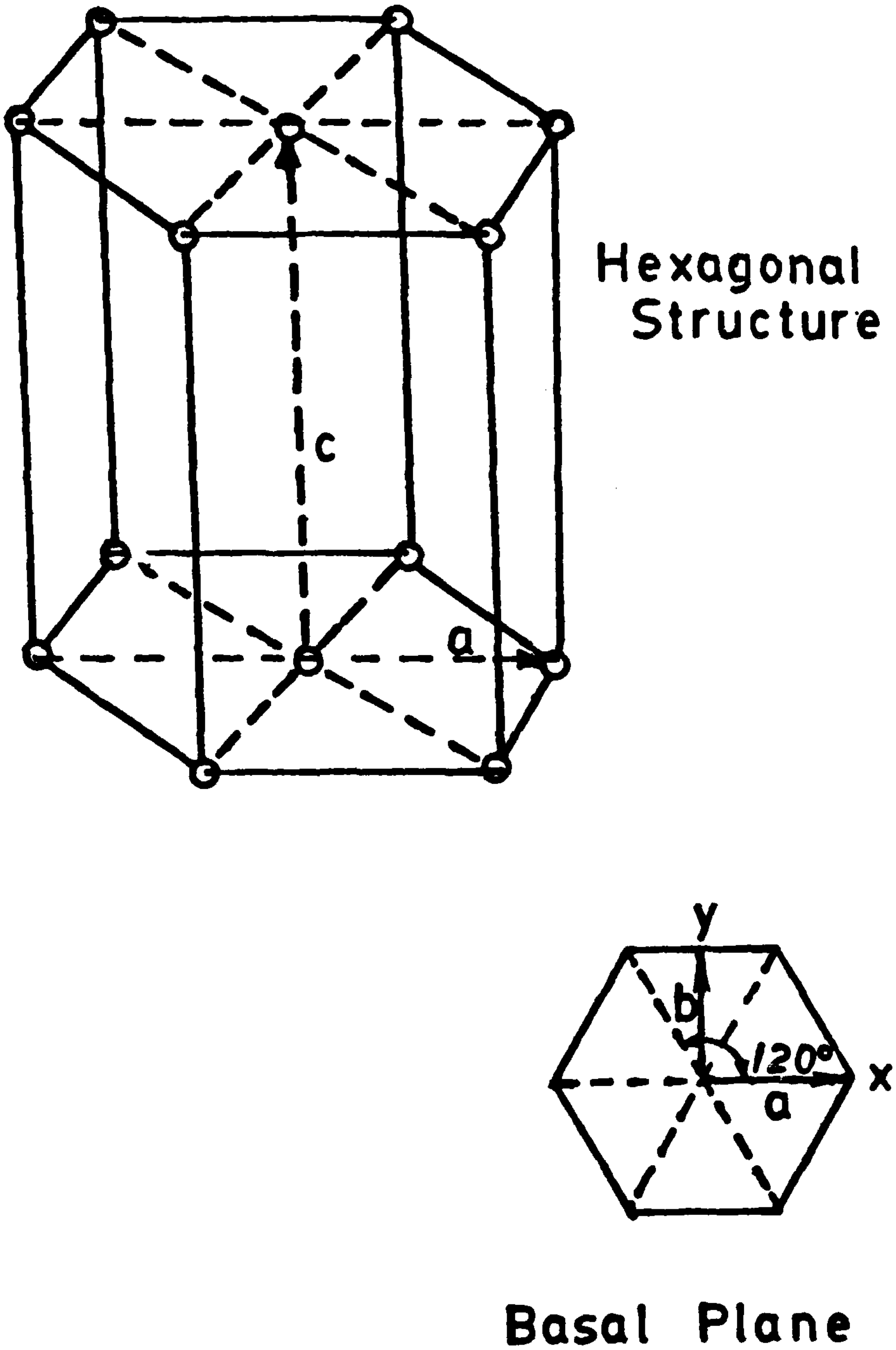

Figure III-1. Hexagonal close packed structure.

$$\rho_0\ddot{U}'' = \frac{\partial}{\partial a''}\left[\frac{\partial\phi_2}{\partial\eta''_{11}} + \left(\frac{\partial\phi_2}{\partial\eta''_{11}}\frac{\partial U}{\partial a''} + \frac{\partial\phi_3}{\partial\eta''_{11}}\right)\right]$$

$$\rho_0\ddot{V}'' = \frac{\partial}{\partial a''}\left[\frac{\partial\phi_2}{\partial\eta''_{21}} + \left(\frac{\partial\phi_2}{\partial\eta''_{11}}\frac{\partial V''}{\partial a''} + \frac{\partial\phi_3}{\partial\eta''_{21}}\right)\right] \qquad \text{(II-41)}$$

$$\rho_0\ddot{W}'' = \frac{\partial}{\partial a''}\left[\frac{\partial\phi_2}{\partial\eta''_{31}} + \left(\frac{\partial\phi_2}{\partial\eta''_{11}}\frac{\partial W''}{\partial a''} + \frac{\partial\phi_3}{\partial\eta''_{31}}\right)\right]$$

These equations are to be examined so one can define the directions and evaluate nonlinearity parameters for finite amplitude ultrasonic waves propagating in hexagonal and trigonal crystals.

III. PURE MODE PROPAGATION IN HEXAGONAL CRYSTALS

There are two classes of crystals with hexagonal symmetry, one with the Hermann-Mauguin symbol 6, $\bar{6}$, 6/m with 5 second-order and 12 third-order elastic constants and the other with the symbol 622, 6 mm, $\bar{6}$m2, 6/mmm with 5 second-order and 10 third-order elastic constants. Since most of the crystals with hexagonal symmetry belong to the second class. We will confine our attention to it. The hexagonal close packed structure is shown in Figure III-1. A drawing of the coordinate frame and the basal plane are also given in the figure. The five SOE constants are C_{11}, C_{12}, C_{13}, C_{33} and C_{44} and the ten TOE constants are C_{111}, C_{113}, C_{116}, C_{133}, C_{144}, C_{226}, C_{333}, C_{334}, C_{366} and C_{456} (3). By using Brugger's definition of third-order elastic constants, the expression for the strain energy density for hexagonal crystals is given by (7):

where ϕ_2 and ϕ_3 represent the separate parts of the strain energy density containing only SOE constants and only TOE constants, respectively.

Now as soon as the expression of the strain energy density in terms of strain components is acquired for specific crystals under investigation, we will be able to get the wave equations for propagating along any a"-direction in the crystals by using the above equations.

These expressions of the strain energy are written out in more explicit terms for crystals belonging to hexagonal and trigonal symmetries in the next sections.

Combining (II-37), (II-38) and (II-39), we may have the following stress components in which the strain terms involving SOE constants are separated from those involving TOE constants:

$$T_{11} = \frac{\partial\phi_2}{\partial\eta''_{11}} + \left(\frac{\partial\phi_2}{\partial\eta''_{11}}\frac{\partial U''}{\partial a''} + \frac{\partial\phi_3}{\partial\eta''_{11}}\right)$$

$$T_{21} = \frac{\partial\phi_2}{\partial\eta''_{21}} + \left(\frac{\partial\phi_2}{\partial\eta''_{11}}\frac{\partial V''}{\partial a''} + \frac{\partial\phi_3}{\partial\eta''_{21}}\right) \qquad \text{(II-40)}$$

$$T_{31} = \frac{\partial\phi_2}{\partial\eta''_{31}} + \left(\frac{\partial\phi_2}{\partial\eta''_{11}}\frac{\partial W''}{\partial a''} + \frac{\partial\phi_3}{\partial\eta''_{31}}\right) .$$

Therefore, we may write a set of equations of motion for plane sound waves propagating along the a"-axis direction in crystals:

Moreover, the components of the Jacobian matrix is terms of displacement gradients can be reduced to

$$J_{11} = 1 + \partial U''/\partial a''$$
$$J_{21} = \partial V''/\partial a''$$
$$J_{31} = \partial W''/\partial a'' \qquad \text{(II-37)}$$
$$J_{22} = J_{33} = 1$$

and

$$J_{12} = J_{13} = J_{23} = J_{32} = 0 \ .$$

Thus, the stress components involved in Equations (II-19) also can be reduced to:

$$T_{11} = J_{11} \frac{\partial \phi}{\partial \eta''_{11}}$$
$$T_{21} = J_{21} \frac{\partial \phi}{\partial \eta''_{11}} + \frac{\partial \phi}{\partial \eta''_{21}} \qquad \text{(II-38)}$$
$$T_{31} = J_{31} \frac{\partial \phi}{\partial \eta''_{11}} + \frac{\partial \phi}{\partial \eta''_{31}} \ .$$

Obviously, under the above rotated coordinate system, the strain energy density ϕ can be expressed as a function not only of the double primed strain components η''_{ij} but also the rotation angles ϕ and θ; i.e., it can be written as:

$$\phi(\eta_{ij}) = \phi(\eta''_{ij},\phi,\theta)$$
$$= \phi_2(\eta''_{ij},\phi,\theta) + \phi_3(\eta_{ij},\phi,\theta) \ , \qquad \text{(II-39)}$$

$$\eta_{11} = \eta''_{11}\cos^2\phi\cos^2\theta - (\eta''_{13} + \eta''_{31})\cos^2\phi\cos\theta\sin\theta$$
$$- (\eta''_{12} + \eta''_{21})\cos\phi\sin\phi\cos\theta$$

$$\eta_{12} = \eta''_{11}\cos\phi\sin\phi\cos^2\theta - (\eta''_{13} + \eta''_{31})\cos\phi\sin\phi\cos\theta\sin\theta$$
$$+ (\eta''_{12}\cos^2\phi - \eta''_{21}\sin^2\phi)\cos\theta$$

$$\eta_{13} = \eta''_{11}\cos\phi\cos\theta\sin\theta + (\eta''_{13}\cos^2\theta - \eta''_{31}\sin^2\theta)\cos\phi$$
$$- \eta''_{21}\sin\phi\sin\theta$$

$$\eta_{21} = \eta''_{11}\cos\phi\sin\phi\cos^2\theta - (\eta''_{13} + \eta''_{31})\sin\phi\cos\phi\sin\theta\cos\theta$$
$$- \eta''_{12}\sin^2\phi\cos\theta + \eta''_{21}\cos^2\phi\cos\theta$$

$$\eta_{22} = \eta''_{11}\sin^2\phi\cos^2\theta - (\eta''_{13} + \eta''_{31})\sin^2\phi\sin\theta\cos\theta \qquad \text{(II-36)}$$
$$+ (\eta''_{12} + \eta''_{21})\sin\phi\cos\phi\cos\theta$$

$$\eta_{23} = \eta''_{11}\sin\phi\cos\theta\sin\theta + (\eta''_{13}\cos^2\theta - \eta''_{31}\sin^2\theta)\sin\phi$$
$$+ \eta''_{21}\cos\phi\sin\theta$$

$$\eta_{31} = \eta''_{11}\cos\phi\sin\theta\cos\theta - (\eta''_{13}\sin^2\theta - \eta''_{31}\cos^2\theta)\cos\phi$$
$$- \eta''_{12}\sin\phi\sin\theta$$

$$\eta_{32} = \eta''_{11}\sin\phi\sin\theta\cos\theta - (\eta''_{13}\sin^2\theta - \eta''_{31}\cos^2\theta)\sin\phi$$
$$+ \eta''_{12}\cos\phi\sin\theta$$

$$\eta_{33} = \eta''_{11}\sin^2\theta + (\eta''_{13} + \eta''_{31})\sin\theta\cos\theta \ .$$

$$\eta''_{11} = U''_a + \frac{1}{2}(U''^2_a + V''^2_a + W''^2_a)$$

$$\eta''_{12} = \eta''_{21} = \frac{1}{2} V''_a \qquad \text{(II-34)}$$

$$\eta''_{13} = \eta''_{31} = \frac{1}{2} W''_a \, .$$

Thus, Eq. (II-32) can be simplified as:

$$\eta'_{11} = \cos\theta(\eta''_{11}\cos\theta - \eta''_{13}\sin\theta) - \eta''_{31}\sin\theta\cos\theta$$

$$\eta'_{12} = \eta''_{12}\cos\theta$$

$$\eta'_{13} = (\eta''_{11}\sin\theta + \eta''_{13}\cos\theta) - \eta''_{31}\sin^2\theta$$

$$\eta'_{21} = \eta''_{21}\cos\theta$$

$$\eta'_{22} = 0 \qquad \text{(II-35)}$$

$$\eta'_{23} = \eta''_{21}\sin\theta$$

$$\eta'_{31} = (\eta''_{11}\cos\theta - \eta''_{13}\sin\theta)\sin\theta + \eta''_{31}\cos^2\theta$$

$$\eta'_{32} = \eta''_{12}\sin\theta$$

$$\eta'_{33} = (\eta''_{11}\sin\theta + \eta''_{13}\cos\theta)\sin\theta + \eta''_{31}\cos\theta\sin\theta$$

Putting (II-35) into (II-31), we have the following expressions for the strain components in the original frame expressed in the terms of the strain components in the twice-rotated or double-primed coordinate system:

$$\eta_{23} = \sin\phi\ \eta'_{13} + \cos\phi\ \eta'_{23}$$

$$\eta_{31} = \eta'_{31}\cos\phi - \eta'_{32}\sin\phi$$

$$\eta_{32} = \eta'_{31}\sin\phi + \eta'_{32}\cos\phi$$

$$\eta_{33} = \eta'_{33}$$

and

$$\eta'_{11} = \cos\theta(\eta''_{11}\cos\theta - \eta''_{13}\sin\theta) - \sin\theta(\eta''_{31}\cos\theta - \eta''_{33}\sin\theta)$$

$$\eta'_{12} = \eta''_{12}\cos\theta - \eta''_{32}\sin\theta$$

$$\eta'_{13} = (\eta''_{11}\sin\theta + \eta''_{13}\cos\theta)\cos\theta - (\eta''_{31}\sin\theta + \eta''_{33}\cos\theta)\sin\theta$$

$$\eta'_{21} = \eta''_{21}\cos\theta - \eta''_{23}\sin\theta$$

$$\eta'_{22} = \eta''_{22} \qquad \text{(II-32)}$$

$$\eta'_{23} = \eta''_{21}\sin\theta + \eta''_{23}\cos\theta$$

$$\eta'_{31} = (\eta''_{11}\cos\theta - \eta''_{13}\sin\theta)\sin\theta + (\eta''_{31}\cos\theta - \eta''_{33}\sin\theta)\cos\theta$$

$$\eta'_{32} = \eta''_{12}\sin\theta + \eta''_{32}\cos\theta$$

$$\eta'_{33} = (\eta''_{11}\sin\theta + \eta''_{13}\cos\theta)\sin\theta + (\eta''_{31}\sin\theta + \eta''_{33}\cos\theta)\cos\theta\ .$$

If only the propagation of the plane wave along the a"-axis is accounted for, the displacement components will be a function of only the coordinate a" (see Eq. (II-27)). In this case, we will have:

$$\eta''_{22} = \eta''_{33} = \eta''_{23} = \eta''_{32} = 0\ , \qquad \text{(II-33)}$$

and the other nonvanishing displacement gradients will be simplified as follows:

$$(\eta) = (R_1^*)(\eta')(R_1)$$

and (II-28)

$$(\eta') = (R_2^*)(\eta'')(R_2)$$

to arrive at the following expressions:

$$\eta = \begin{vmatrix} \cos\phi & -\sin\phi & 0 \\ \sin\phi & \cos\phi & 0 \\ 0 & 0 & 1 \end{vmatrix} \begin{vmatrix} \eta'_{11} & \eta'_{12} & \eta'_{13} \\ \eta'_{21} & \eta'_{22} & \eta'_{23} \\ \eta'_{31} & \eta'_{32} & \eta'_{33} \end{vmatrix} \begin{vmatrix} \cos\phi & \sin\phi & 0 \\ -\sin\phi & \cos\phi & 0 \\ 0 & 0 & 1 \end{vmatrix} \quad \text{(II-29)}$$

and

$$\eta' = \begin{vmatrix} \cos\theta & 0 & -\sin\theta \\ 0 & 1 & 0 \\ \sin\theta & 0 & \cos\theta \end{vmatrix} \begin{vmatrix} \eta''_{11} & \eta''_{12} & \eta''_{13} \\ \eta''_{21} & \eta''_{22} & \eta''_{23} \\ \eta''_{31} & \eta''_{32} & \eta''_{33} \end{vmatrix} \begin{vmatrix} \cos\theta & 0 & \sin\theta \\ 0 & 1 & 0 \\ -\sin\theta & 0 & \cos\theta \end{vmatrix} .$$

(II-30)

Multiplication and simplification lead to the following expressions for the strain components expressed in terms of the strain components in the twice-rotated system.

$$\eta_{11} = \cos\phi(\eta'_{11}\cos\phi - \eta'_{12}\sin\phi) - \sin\phi(\eta'_{21}\cos\phi - \eta'_{22}\sin\phi)$$

$$\eta_{12} = \cos\phi(\eta'_{11}\sin\phi + \eta'_{12}\cos\phi) - \sin\phi(\eta'_{21}\sin\phi + \eta'_{22}\cos\phi)$$

$$\eta_{13} = \eta'_{13}\cos\phi - \eta'_{23}\sin\phi$$

$$\eta_{21} = \sin\phi(\eta'_{11}\cos\phi - \eta'_{12}\sin\phi) + \cos\phi(\eta'_{21}\cos\phi - \eta'_{22}\sin\phi)$$

$$\eta_{22} = \sin\phi(\eta'_{11}\sin\phi + \eta'_{12}\cos\phi) + \cos\phi(\eta'_{21}\sin\phi + \eta'_{22}\cos\phi) \quad \text{(II-31)}$$

Then we may rotate the above rotated basal plane (a'b') by an angle θ about the b' axis (Figure II-1) so that the new coordinate frame formation can be expressed as

$$\begin{vmatrix} a'' \\ b'' \\ c'' \end{vmatrix} = (R_2) \begin{vmatrix} a' \\ b' \\ c' \end{vmatrix} , \qquad \text{(II-24)}$$

where the rotation matrix (R_2) is given by

$$(R_2) = \begin{vmatrix} \cos\theta & 0 & \sin\theta \\ 0 & 1 & 0 \\ -\sin\theta & 0 & \cos\theta \end{vmatrix} , \qquad \text{(II-25)}$$

and its corresponding transpose (R_2^*) is

$$(R_2^*) = \begin{vmatrix} \cos\theta & 0 & -\sin\theta \\ 0 & 1 & 0 \\ \sin\theta & 0 & \cos\theta \end{vmatrix} . \qquad \text{(II-26)}$$

Let us define the displacement components propagating along the a"-axis in the new coordinate system as

$$\begin{aligned} U'' &= U''(a'',t) \\ V'' &= V''(a'',t) \qquad \text{(II-27)} \\ W'' &= W''(a'',t) . \end{aligned}$$

Using the transformation matrix as given by (II-22), (II-23), (II-25) and (II-26) one can transform the strain components by (1)

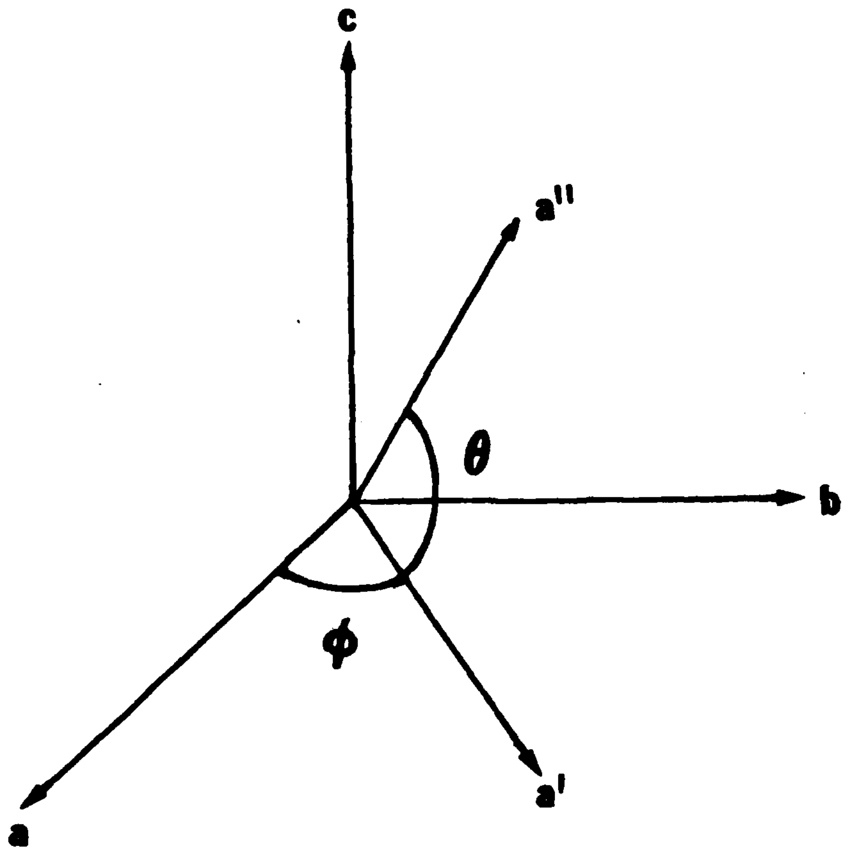

Figure II-1. Rotation of coordinate.

The procedure for accomplishing this is developed in the next section of this technical report.

We have shown the strain energy density ϕ in the original frame can be expressed as a function of strain components,

$$\phi = \phi(\eta_{ij}) \; . \qquad \text{(II-20)}$$

Now, suppose we want to consider the propagation of the plane finite amplitude wave along some arbitrary direction instead of the a-axis under original consideration. We can rotate the basal plane in the original frame by an angle ϕ about the c-axis (Figure II-1) first so that the rotated coordinate frame formation can be written as

$$\begin{vmatrix} a' \\ b' \\ c' \end{vmatrix} = (R_1) \begin{vmatrix} a \\ b \\ c \end{vmatrix} \qquad \text{(II-21)}$$

where the rotation matrix (R_1) is given by

$$(R_1) \begin{vmatrix} \cos\phi & \sin\phi & 0 \\ -\sin\phi & \cos\phi & 0 \\ 0 & 0 & 1 \end{vmatrix} \qquad \text{(II-22)}$$

and its corresponding transpose (R_1^*) is

$$(R_1^*) = \begin{vmatrix} \cos & -\sin\phi & 0 \\ \sin\phi & \cos\phi & 0 \\ 0 & 0 & 1 \end{vmatrix} . \qquad \text{(II-23)}$$

that keeping higher order terms is not justified. The terms kept in the theory to be presented are sufficient to account for most of the observed nonlinear properties.

III. PURE MODE DIRECTIONS

In an anisotropic medium there are only certain directions along which elastic waves can propagate as the pure longitudinal modes which are of primary interest in the application of the harmonic generation technique. Associated with any propagation direction there are possibly three independent waves, the displacements of which form a mutually orthogonal set (coupling terms). In general, none of the three displacement vectors coincides with the vector which is normal to the wavefront; i.e., in general, the waves are neither pure longitudinal nor pure transverse. The specific directions in which pure mode longitudinal waves propagate have been determined qualitatively by Borgnis (5) and Brugger (6) for crystals with cubic, hexagonal and trigonal symmetry, etc. In order to derive the nonlinear wave equation of a pure longitudinal mode in specific directions and then to determine the third-order elastic constants from the harmonic generation technique, it is necessary to know quantitatively these pure mode directions. In addition, some unique pure mode directions even depend upon the SOE constants for the crystals under consideration.

In order to determine the pure longitudinal mode directions, one must develop a way to rotate the coordinates and observe the directions in which the coupling terms in the wave equations vanish.

$$J_{31} = W_a = \partial W/\partial a$$

$$J_{32} = W_b = \partial W/\partial b$$

$$J_{33} = 1 + W_c = 1 + \partial W/\partial c$$

We will consider the case of plane finite amplitude waves propagating along the axes of the medium under consideration. For plane waves propagating along the a-axis the displacements become:

$$U = U(a,t)$$
$$V = V(a,t) \qquad \text{(II-18)}$$
$$W = W(a,t) \ .$$

As the a-axis can be rotated into alignment with any axis of the medium, the equations of motion in general can be written

$$\rho_0 \ddot{U} = \partial T_{11}/\partial a$$

$$\rho_0 \ddot{V} = \partial T_{21}/\partial a \qquad \text{(II-19)}$$

$$\rho_0 \ddot{W} = \partial T_{31}/\partial a \ ,$$

for propagation along the a-direction.

These equations are written out in more explicit terms and solved for crystals belonging to hexagonal and trigonal symmetry in the following sections. The expressions of elastic strain energy density appropriate to different crystal symmetries are obtained with only terms up to third-order since to date, experimental uncertainty is great enough into Eq. (III-1), which was Brugger's definition. The relation between Birch and Brugger TOE constants can be obtained from the relation:

$$C_{\lambda\mu\nu}^{(\mathrm{Birch})} = M C_{\lambda\mu\nu}^{(\mathrm{Brugger})}/6 \qquad \text{(III-3)}$$

where M is the possible number of ways in which $C_{\lambda\mu\nu}$ can be expressed in tensor notation.

Now we substitute (II-36) for (III-1) to give the expression for the strain energy density in the double primed coordinate system,

$$\phi(\eta_{ij}) = \phi_2(\eta''_{ij},\phi,\theta) + \phi_3(\eta''_{ij},\phi,\theta) \qquad \text{(III-4)}$$

where

$$\begin{aligned}
\phi_2 = {} & \frac{1}{2} C_{11}[\eta''^2_{11}\cos^4\theta + (\eta''^2_{12} + \eta''^2_{21})\cos^2\theta \\
& - 2\eta''_{11}(\eta''_{13} + \eta''_{31})\sin\theta\cos^3\theta + (\eta''_{13} + \eta''_{31})^2\cos^2\theta\sin^2\theta] \\
& - \frac{1}{2} C_{12}(\eta''^2_{12} + \eta''^2_{21})\cos^2\theta + C_{13}[\eta''^2_{11}\cos^2\theta\sin^2\theta \\
& + \eta''_{11}(\eta''_{13} + \eta''_{13})\cos\theta\sin\theta(\cos^2\theta - \sin^2\theta) - (\eta''_{13} + \eta''_{31})^2\cos^2\theta\sin^2\theta] \\
& + \frac{1}{2} C_{33}[\eta''^2_{11}\sin^4\theta + 2\eta''_{11}(\eta''_{13} + \eta''_{31})\sin^3\theta\cos\theta \\
& + (\eta''_{13} + \eta''_{31})^2\sin^2\theta\cos^2\theta] + C_{44}[2\eta''^2_{11}\cos^2\theta\sin^2\theta \\
& + (\eta''^2_{12} + \eta''^2_{21})\sin^2\theta + 2\eta''_{11}(\eta''_{13} + \eta''_{31})\cos\theta\sin\theta(\cos^2\theta - \sin^2\theta) \\
& + (\eta''_{13} + \eta''_{31})^2(\cos^4\theta + \sin^4\theta) - 4\eta''_{13}\eta''_{31}\cos^2\theta\sin^2\theta]
\end{aligned} \qquad \text{(III-5)}$$

and

$$\phi_3 = \frac{1}{6} C_{111}[\eta''^3_{11}\cos^6\theta - 3\eta''^2_{11}(\eta''_{13} + \eta''_{31})\cos^5\theta\sin\theta + 3\eta''_{11}(\eta''_{13} + \eta''_{31})^2\cos^4\theta\sin\theta$$

$$- (\eta''_{13} + \eta''_{31})^3\cos^3\theta\sin^3\theta] + \frac{1}{2} C_{113}[\eta''^3_{11}\sin^2\theta\cos^4\theta + \eta''^2_{11}(\eta''_{13} + \eta''_{31})$$

$$\cdot \sin\theta\cos^3\theta(\cos^2\theta - 2\sin^2\theta) + \eta''_{11}(\eta''_{13} + \eta''_{31})^2\cos^2\theta\sin^2\theta(\sin^2\theta - 2\cos^2\theta)$$

$$+ (\eta''_{13} + \eta''_{31})^3\cos^3\theta\sin^3\theta] + C_{133}[\eta''^3_{11}\sin^4\theta\cos^2\theta + \eta''^2_{11}(\eta''_{13} + \eta''_{31})$$

$$\cdot \cos\theta\sin^3\theta(2\cos^2\theta - \sin^2\theta) + \eta''_{11}(\eta''_{13} + \eta''_{31})^2 \cos^2\theta\sin^2\theta(\cos^2\theta - 2\sin^2\theta)$$

$$- (\eta''_{13} + \eta''_{31})^3\cos^3\theta\sin^3\theta] + C_{144}[\eta''^3_{11}(2\cos^4\theta\sin^2\theta)$$

$$+ \eta''_{11}(\eta''^2_{12} + \eta''^2_{21})\cos^2\theta\sin^2\theta + 2\eta''^2_{11}(\eta''_{13} + \eta''_{31})\cos^3\theta\sin\theta(\cos^2\theta - 2\sin^2\theta)$$

$$- (\eta''_{13} + \eta''_{31})(\eta''^2_{12} + \eta''^2_{21})\cos\theta\sin^3\theta + \eta''_{11}(\eta''^2_{13} + \eta''^2_{31})\cos^6\theta$$

$$+ 3\eta''_{11}(\eta''^2_{13} + \eta''^2_{31})\sin^4\theta\cos^2\theta - 2\eta''_{11}(\eta''^2_{13} + \eta''^2_{31})\cos^4\theta\sin^2\theta$$

$$+ 4\eta''_{11}\eta''_{13}\eta''_{31}\cos^2\theta\sin^2\theta(\sin^2\theta - 2\cos^2\theta) - (\eta''_{13} + \eta''_{31})$$

$$\cdot (\eta''^2_{13} + \eta''^2_{31})\cos\theta\sin\theta(\cos^4\theta + \sin^4\theta) + 4\eta''_{13}\eta''_{31}(\eta''_{13} + \eta''_{31})\cos^3\theta\sin^3\theta]$$

$$+ \frac{1}{2} C_{166}[\eta''^3_{11}\cos^6\theta - 3\eta''^2_{11}(\eta''_{13} + \eta''_{31})\cos^5\theta\sin\theta + 3\eta''_{11}(\eta''_{13} + \eta''_{31})^2$$

$$\cdot \cos^4\theta\sin^2\theta - (\eta''_{13} + \eta''_{31})^3\cos^3\theta\sin^3\theta + 2\eta''_{11}(\eta''^2_{12} + \eta''^2_{21})\cos^4\theta$$

$$- 2(\eta''_{13} + \eta''_{31})(\eta''^2_{12} + \eta''^2_{21})\cos^3\theta\sin\theta] + \frac{1}{6} C_{266}[\eta''^3_{11}\cos^6\theta\cos(2\phi)$$

$$\cdot (\cos^2(2\phi) - 3\sin^2(2\phi)) + 3\eta''^2_{11}(\eta''_{13} + \eta''_{31})\cos^5\theta\sin\theta\cos(2\phi)$$

$$\cdot (3\sin^3(2\phi) - \cos^2(2\phi)) + 3\eta''_{11}(\eta''_{13} + \eta''_{31})^2\cos^4\theta \sin^2\theta\cos(2\phi)$$

$$\cdot (\cos^2(2\phi) - 3\sin^2(2\phi) + 3\eta''^2_{11}(\eta''_{12} + \eta''_{21})\cos^5\theta\sin(2\phi)(\sin^2(2\phi)$$

$$- 3\cos^2(2\phi)) + 9\eta''_{11}(\eta''_{12} + \eta''_{21})^2\cos^4\theta\cos(2\phi)\sin^2(2\phi)$$

$$- \eta''_{11}(\eta''^2_{12} + \eta''^2_{21})\cos^4\theta\cos(2\phi)(\cos^4\phi + \sin^4\phi) + 6\eta''_{11}(\eta''_{12} + \eta''_{21})$$

$$\cdot (\eta''_{13} + \eta''_{31})\cos^4\theta\sin\theta\sin(2\phi)(3\cos^2(2\phi) - \sin^2(2\phi))$$

$$+ 6\eta''_{11}\eta''_{12}\eta''_{21}\cos^4\theta\sin^2(2\phi)\cos(2\phi) + (\eta''_{13} + \eta''_{31})^3\cos^3\theta\sin^3\theta\cos(2\phi)$$

$$\cdot (\cos^2(2\phi) + 3\sin(2\phi)) - (\eta''_{12} + \eta''_{21})^3\cos^3\theta\sin^3(2\phi)$$

$$+ 3(\eta''_{12} + \eta''_{21})(\eta''_{13} + \eta''_{31})^2\cos^3\theta\sin^2\theta\sin(2\phi)(\sin^2(2\phi) - 3\cos^2(2\phi))$$

$$- 9(\eta''_{12} + \eta''_{21})^2(\eta''_{13} + \eta''_{31})\cos^3\theta\sin\theta\sin^2(2\phi)\cos(2\phi)$$

$$+ (\eta''_{13} + \eta''_{31})(\eta''^2_{12} + \eta''^2_{21})\cos^3\theta\sin\theta\cos(2\phi)(\cos^4\phi + \sin^4\phi)$$

$$+ (\eta''_{12} + \eta''_{21})(\eta''^2_{12} + \eta''^2_{21})\cos^3\theta\sin(2\phi)(\cos^4\phi + \sin^4\phi)$$

$$- 6\eta''_{12}\eta''_{21}(\eta''_{13} + \eta''_{31})\sin\theta\cos^3\theta\cos(2\phi)\sin^2(2\phi)$$

$$- 6\eta''_{12}\eta''_{21}(\eta''_{12} + \eta''_{21})\cos^3\theta\sin^3(2\phi)] + \frac{1}{6}C_{333}[\eta''^3_{11}\sin^6\theta + 3\eta''^2_{11}$$

$$\cdot (\eta''_{13} + \eta''_{31})\sin^5\theta\cos\theta + 3\eta''_{11}(\eta''_{13} + \eta''_{31})^2 \sin^4\theta\cos^2\theta$$

$$+ (\eta''_{13} + \eta''_{31})^3\sin^3\theta\cos^3\theta] + C_{344}[2\eta''^3_{11}\cos^2\theta\sin^4\theta + \eta''_{11}(\eta''^2_{12} + \eta''^2_{21})$$

$$\cdot \sin^4\theta + 2\eta''^2_{11}(\eta''_{13} + \eta''_{31})\cos\theta\sin^3\theta(2\cos^2\theta - \sin^2\theta)$$

$$+ \eta''_{11}(\eta''^2_{13} + \eta''^2_{31})\sin^2\theta(\cos^4\theta + \sin^4\theta) - 4\eta''_{11}\eta''_{13}\eta''_{31}\cos^2\theta\sin^4\theta$$

$$+ (\eta''^2_{12} + \eta''^2_{21})(\eta''_{13} + \eta''_{31})\sin^3\theta\cos\theta + 2\eta''_{11}(\eta''_{13} + \eta''_{31})^2\sin^2\theta\cos^2\theta$$

$$\cdot (\cos^2\theta - \sin^2\theta) + (\eta''_{13} + \eta''_{31})(\eta''^2_{13} + \eta''^2_{31})\sin\theta\cos\theta(\cos^4\theta + \sin^4\theta)$$

$$- 4\eta''_{13}\eta''_{31}(\eta''_{13} + \eta''_{31})\cos^3\theta\sin^3\theta] + \frac{1}{2} C_{366}[\eta''^3_{11}\sin^2\theta\cos^4\theta$$

$$+ \eta''^2_{11}(\eta''_{13} + \eta''_{31})\cos^3\theta\sin\theta(\cos^2\theta - 2\sin^2\theta) + \eta''_{11}(\eta''_{13} + \eta''_{31})^2$$

$$\cdot \cos^2\theta\sin^2\theta(\sin^2\theta - 2\cos^2\theta) + (\eta''_{13} + \eta''_{31})^3\cos^3\theta\sin^3\theta$$

$$+ 2\eta''_{11}(\eta''^2_{12} + \eta''^2_{21})\cos^2\theta\sin^2\theta + 2(\eta''^2_{12} + \eta''^2_{21})(\eta''_{13} + \eta''_{31})\cos^3\theta\sin\theta]$$

$$+ 2C_{456}[2\eta''^3_{11}\cos^4\theta\sin^2\theta + 2\eta''^2_{11}(\eta''_{13} + \eta''_{31})\cos^3\theta\sin\theta(\cos^2\theta - 2\sin^2\theta)$$

$$- 2\eta''_{11}(\eta''_{13} + \eta''_{31})^2\cos^2\theta\sin^2\theta(\cos^2\theta - \sin^2\theta) - \eta''_{11}(\eta''^2_{12} + \eta''^2_{21})$$

$$\cdot \cos^2\theta\sin^2\theta(2\sin^2(2\phi) + \cos^2(2\phi)) + 2\eta''_{21}(\eta''^2_{13} - \eta''^2_{31})$$

$$\cdot \cos\theta\sin^2\theta\cos(2\phi)\sin(2\phi) + 2\eta''_{12}(\eta''^2_{31} - \eta''^2_{13})\cos\theta\sin^2\theta\cos(2\phi)\sin(2\phi)$$

$$+ 8\eta''_{11}\eta''_{12}\eta''_{21}\cos^2\theta\sin^2\theta(\cos^4\phi + \sin^4\phi - \sin^2\phi\cos^2\phi)$$

$$+ 2\eta''_{11}\eta''_{13}\eta''_{31}(\cos^6\theta + \cos^2\theta\sin^4\theta)\sin^2(2\phi) - 2\eta''_{11}(\eta''^2_{13} + \eta''^2_{31})$$

$$\cdot \cos^4\theta\sin^2\theta\sin^2(2\phi) + \eta''_{11}(\eta''^2_{13} + \eta''^2_{31})\cos^2\theta(\cos^4\theta + \sin^4\theta)\cos^2(2\phi)$$

$$- 2\eta''_{11}(\eta''_{13} + \eta''_{31})^2\cos^2\theta\sin^2\theta(\cos^2\theta - \sin^2\theta)\cos^2(2\phi)$$

$$- 2\eta''_{11}\eta''_{21}\eta''_{13}\cos^4\theta\sin\theta\cos(2\phi)\sin(2\phi) + 2\eta''_{11}\eta''_{21}\eta''_{31}\cos^2\theta\sin^3\theta$$

$$\cdot \cos(2\phi)\sin(2\phi) - 2\eta''_{11}\eta''_{12}\eta''_{31}\sin\theta\cos^4\theta\cos(2\phi)\sin(2\phi)$$

$$+ 2\eta''_{11}\eta''_{12}\eta''_{13}\cos^2\theta\sin^3\theta\cos(2\phi)\sin(2\phi) + 2\eta''_{11}(\eta''_{12} + \eta''_{21})^2$$

$$\cdot \cos^2\theta\sin^2\theta\sin^2(2\phi) - 4\eta''_{11}\eta''_{13}\eta''_{31}\cos^4\theta\sin^2\theta\cos^2(2\phi)$$

$$+ (\eta''_{12} + \eta''_{21})(\eta''^2_{12} + \eta''^2_{21})\sin^2\theta\cos\theta\cos(2\phi)\sin(2\phi)$$

$$- 2(\eta''^2_{12}\eta''_{21} + \eta''_{12}\eta''^2_{21})\cos\theta\sin^2\theta\sin(2\phi)\cos(2\phi) + 2(\eta''_{13} + \eta''_{31})$$

$$\cdot (\eta''^2_{13} + \eta''^2_{31})\cos^3\theta\sin^3\theta\sin^2(2\phi) - 2\eta''_{13}\eta''_{31}(\eta''_{13} + \eta''_{31})$$

$$\cdot \cos\theta\sin\theta(\sin^4\theta + \cos^4\theta)\sin^2(2\phi) - 2\eta''^2_{13}(\eta''_{12} + \eta''_{21})$$

$$\cdot \cos^3\theta\sin^2\theta\sin(2\phi)\cos(2\phi) - (\eta''_{12} + \eta''_{21})(\eta''^2_{13} + \eta''^2_{31})\cos\theta$$

$$\cdot (\cos^4\theta + \sin^4\theta)\cos(2\phi)\sin(2\phi) + 4(\eta''_{12} + \eta''_{21})\eta''_{13}\eta''_{31}$$

$$\cdot \cos^3\theta\sin^2\theta\cos(2\phi)\sin(2\phi) - (\eta''_{12} + \eta''_{21})\eta''^2_{31}\cos^3\theta\sin^2\theta$$

$$\cdot \sin(2\phi)\cos(2\phi) + 2\eta''_{13}\eta''_{31}(\eta''_{12} + \eta''_{21})\cos\theta\sin\theta(\cos^4\theta + \sin^4\theta)$$

$$\cdot \sin(2\phi)\cos(2\phi) + \eta''_{21}\eta''_{13}(\eta''_{12} + \eta''_{21})\cos^3\theta\sin\theta\sin^2(2\phi)$$

$$+ 2\eta''_{12}\eta''_{21}(\eta''_{13} + \eta''_{31})\cos\theta\sin^3\theta\sin^2(2\phi) + 4\eta''_{12}\eta''_{21}(\eta''_{13} + \eta''_{31})$$

$$\cdot \cos\theta\sin\theta(\cos^2\theta - \sin^2\theta)(\cos^4\phi + \sin^4\phi) - 2(\eta''_{12} + \eta''_{21})\eta''_{21}\eta''_{31}$$

$$\cdot \sin^3\theta\cos\theta\sin^2(2\phi) + 2\eta''_{12}\eta''_{31}(\eta''_{12} + \eta''_{21})\sin\theta\cos^3\theta\sin^2(2\phi)$$

$$- 2\eta''_{12}\eta''_{13}(\eta''_{12} + \eta''_{21})\sin^3\theta\cos\theta\sin^2(2\phi)$$

$$- 2(\eta''_{13} + \eta''_{31})(\eta''^2_{12} + \eta''^2_{21})\cos^3\theta\sin\theta\sin^2(2\phi) \; . \qquad \text{(III-6)}$$

In order to obtain a set of equations of motion for the plane waves propagating along the a"-axis direction, it is necessary only to calculate three of the strain derivatives of the elastic energy, i.e., $\partial\phi/\partial\eta''_{11}$, $\partial\phi/\partial\eta''_{21}$ and $\partial\phi/\partial\eta''_{31}$, according to (II-38). Those strain derivatives can be calculated and simplified by taking into account $\eta''_{12} = \eta''_{21}$ and η''_{13} and η''_{31}. They are:

$$\frac{\partial\phi}{\partial\eta''_{11}} = \frac{\partial\phi_2}{\partial\eta''_{11}} + \frac{\partial\phi_3}{\partial\eta''_{11}} \tag{III-7}$$

where

$$\frac{\partial\phi_2}{\partial\eta''_{11}} = (C_{11}\cos^4\theta + 2C_{13}\cos^2\theta\sin^2\theta + C_{33}\sin^4\theta$$

$$+ C_{44}\sin^2(2\theta))\eta''_{11} + (-C_{11}\cos^2\theta\sin(2\theta)$$

$$+ C_{13}\cos(2\theta)\sin(2\theta) + C_{33}\sin^2\theta\sin(2\theta)$$

$$+ C_{44}\sin(4\theta))\eta''_{13} \tag{III-8}$$

$$\frac{\partial\phi_3}{\partial\eta''_{11}} = \frac{1}{2}C_{111}[\eta''^2_{11}\cos^6\theta - 4\eta''_{11}\eta''_{13}\cos^5\theta\sin\theta + 4\eta''^2_{13}\cos^4\theta\sin\theta]$$

$$+ \frac{1}{2}C_{113}[3\eta''^2_{11}\sin^2\theta\cos^4\theta + 4\eta''_{11}\eta''_{13}\sin\theta\cos^3\theta$$

$$\cdot(\cos^2\theta - 2\sin^2\theta) + 4\eta''^2_{13}\cos^2\theta\sin^2\theta(\sin^2\theta - 2\cos^2\theta]$$

$$+ \frac{1}{2}C_{133}[3\eta''^2_{11}\sin^4\theta\cos^2\theta + 4\eta''_{11}\eta''_{13}\cos\theta\sin^3\theta(2\cos^2\theta - \sin^2\theta)$$

$$+ 4\eta''^2_{13}\cos^2\theta\sin^2\theta(\cos^2\theta - 2\sin^2\theta)] + C_{144}[6\eta''^2_{11}\cos^4\theta\sin^2\theta$$

$$+ 2\eta''^2_{12}\cos^2\theta\sin^2\theta + 4\eta''_{11}(\eta''_{12} + \eta''_{21})\cos^3\theta\sin\theta(\cos^2\theta - 2\sin^2\theta)$$

$$+ 2\eta''^2_{13}(\cos^6\theta + 5\cos^2\theta\sin^2\theta\cos(2\theta))$$

$$+ \frac{1}{2} C_{166}[3\eta''^2_{11}\cos^6\theta - 6\eta''_{11}(\eta''_{13} + \eta''_{31})\cos^5\theta\sin\theta$$

$$+ 3(\eta''_{13} + \eta''_{31})^2\cos^4\theta\sin^2\theta + 2(\eta''^2_{12} + \eta''^2_{21})\cos^4\theta]$$

$$+ \frac{1}{2} C_{266}[\eta''^2_{11}\cos^6\theta\cos(2\phi)(\cos^2(2\phi) - 3\sin^2(2\phi))$$

$$+ 12\eta''_{11}\eta''_{13}\cos^5\theta\sin\theta\cos(2\phi)(3\sin^2(2\phi) - \cos^2(2\phi))$$

$$+ 4\eta''^2_{13}\cos^4\theta\sin^2\theta\cos(2\phi)(\cos^2(2\phi) - 3\sin^2(2\phi))$$

$$+ 12\eta''_{11}\eta''_{12}\cos^5\theta\sin(2\phi)(\sin^2(2\phi) - 3\cos^2(2\phi))$$

$$+ 42\eta''^2_{12}\cos^4\theta\cos(2\phi)\sin^2(2\phi) + 24\eta''_{12}\eta''_{13}\cos^4\theta\sin\theta\sin(2\phi)$$

$$\cdot (3\cos^2(2\phi) - \sin^2(2\phi))] + \frac{1}{2} C_{333}[\eta''^2_{11}\sin^6\theta$$

$$+ 4\eta''_{11}\eta''_{13}\sin^5\theta\cos\theta + 4\eta''^2_{13}\sin^4\theta\cos^2\theta]$$

$$+ C_{344}[6\eta''^2_{11}\cos^2\theta\sin^4\theta + 8\eta''_{11}\eta''_{13}\cos\theta\sin^3\theta$$

$$\cdot (2\cos^2\theta - \sin^2\theta) + 2\eta''^2_{12}\sin^4\theta + 2\eta''^2_{13}(5\sin^2\theta\cos^4\theta + \sin^6\theta$$

$$- 6\cos^2\theta\sin^4\theta)] + \frac{1}{2} C_{366}[3\eta''^2_{11}\sin^2\theta\cos^4\theta$$

$$+ 4\eta''_{11}\eta''_{13}\sin\theta\cos^3\theta(\cos^2\theta - 2\sin^2\theta) + 4\eta''^2_{13}\cos^2\theta\sin^2\theta$$

$$\cdot(\sin^2\theta - 2\cos^2\theta)] + 2C_{456}[6\eta''^2_{11}\cos^4\theta\sin^2\theta$$

$$+ 8\eta''_{11}\eta''_{13}\cos^3\theta\sin\theta(\cos^2\theta - 2\sin^2\theta) + 2\eta''^2_{13}(-4\cos^4\theta\sin^2\theta$$

$$+ 4\cos^2\theta\sin^4\theta + \cos^6\theta + 5\cos^2\theta\sin^4\theta\cos(2\phi)$$

$$+ \cos^2\theta\sin^4\theta\sin^2(2\phi) - 2\cos^4\theta\sin^2\theta\sin^2(2\phi)$$

$$- 6\cos^4\theta\sin^2\theta\cos^2(2\phi)) + 2\eta''^2_{12}\cos^2\theta\sin^2\theta$$

$$\cdot (\sin^2(2\phi) - \cos^2(2\phi) + 4\cos^4\phi + 4\sin^4\phi)$$

$$+ 4\eta''_{12}\eta''_{13}\cos(2\phi)\sin(2\phi)\cos^2\theta\sin\theta(\sin^2\theta - \cos^2\theta)] , \qquad \text{(III-9)}$$

$$\frac{\partial\phi}{\partial\eta''_{21}} = \frac{\partial\phi_2}{\partial\eta''_{21}} + \frac{\partial\phi_3}{\partial\eta''_{21}} \qquad \text{(III-10)}$$

where

$$\frac{\partial\phi_2}{\partial\eta''_{21}} = (C_{11}\cos^2\theta - C_{12}\cos^2\theta + 2C_{44}\sin^2\theta)\eta''_{12} \qquad \text{(III-11)}$$

and

$$\frac{\partial\phi_3}{\partial\eta''_{21}} = C_{144}[2\eta''_{11}\eta''_{21}\cos^2\theta\sin^2\theta - 4\eta''_{13}\eta''_{12}\cos^3\theta\sin\theta]$$

$$+ \frac{1}{6} C_{266}[3\eta''^2_{11}\cos^5\theta\sin(2\phi)(\sin^2(2\phi) - 3\cos^2(2\phi))$$

$$+ 12\eta''_{11}\eta''_{13}\cos^4\theta\sin\theta\sin(2\phi)(3\cos^2(2\phi) - \sin^2(2\phi))$$

$$+ 38\eta''_{11}\eta''_{12}\cos^4\theta\cos(2\phi)\sin^2(2\phi) - 2\eta''_{11}\eta''_{12}\cos^4\theta\cos(2\theta)$$

$$\cdot (\cos^4\phi + \sin^4\phi) + 12\eta''^2_{13}\cos^3\theta\sin^2\theta\sin(2\phi)$$

$$\cdot (\sin^2(2\phi) - 3\cos^2(2\phi) - 30\eta''^2_{12}\cos^3\theta\sin^3(2\phi)$$

$$+ 6\eta''^2_{12}\cos^3\theta\sin(2\phi)(\cos^4\phi + \sin^4\phi) - 76\eta''_{12}\eta''_{13}\sin\theta\cos^3\theta\cos$$

$$\cdot (2\phi)\sin^2(2\phi) + 4\eta''_{12}\eta''_{13}\cos^3\theta\sin\theta\cos(2\phi)]$$

$$+ C_{344}[2\eta''_{11}\eta''_{12}\sin^4\theta + 4\eta''_{12}\eta''_{13}\sin^3\theta\cos\theta]$$

$$+ \frac{1}{2} C_{366}[4\eta''_{11}\eta''_{12}\sin^2\theta\cos^2\theta + 8\eta''_{12}\eta''_{13}\sin\theta\cos^3\theta]$$

$$+ 2C_{456}[2\eta''_{11}\eta''_{12}(\cos^2\theta\sin^2\theta + 4\cos^2\theta\sin^2\theta\cos^4\phi$$

$$+ 4\cos^2\theta\sin^2\theta\sin^4\phi) + 2\eta''_{11}\eta''_{13}\cos^2\theta\sin^2\theta\cos(2\phi)\sin(2\phi)$$

$$- 3\eta''_{12}\eta''_{13}\cos^3\theta\sin\theta\sin^2(2\phi) - 4\eta''_{12}\eta''_{13}\cos\theta\sin^3\theta\sin^2(2\phi)$$

$$+ 8\eta''_{12}\eta''_{13}(\cos^4\phi + \sin^4\phi)\cos\theta\sin\theta(\cos^2\theta - \sin^2\theta)]$$

(III-12)

$$\frac{\partial\phi}{\partial\eta''_{31}} = \frac{\partial\phi_2}{\partial\eta''_{31}} + \frac{\partial\phi_3}{\partial\eta''_{31}} \qquad \text{(III-13)}$$

where

$$\frac{\partial\phi_2}{\partial\eta''_{31}} = [-C_{11}\cos^3\theta\sin\theta + C_{13}\cos\theta\sin\theta(\cos^2\theta - \sin^2\theta)$$

$$+ C_{33}\sin^3\theta\cos\theta + 2C_{44}\cos\theta\sin\theta(\cos^2\theta - \sin^2\theta)]\eta''_{11}$$

$$+ [2C_{11}\cos^2\theta\sin^2\theta - 4C_{13}\cos^2\theta\sin^2\theta + 2C_{33}\cos^2\theta\sin^2\theta$$

$$+ 4C_{44}(\cos^4\theta + \sin^4\theta - \sin^2\theta\cos^2\theta)]\eta''_{13} \qquad \text{(III-14)}$$

and

$$
\begin{aligned}
\frac{\partial \phi_3}{\partial \eta''_{31}} = \frac{1}{2} C_{111}[& -\eta''^2_{11}\cos^5\theta\sin\theta + 4\eta''_{11}\eta''_{13}\cos^4\theta\sin\theta \\
& - 4\eta''^2_{13}\cos^3\theta\sin^3\theta] + \frac{1}{2} C_{113}[\eta''^2_{11}\cos^3\theta\sin\theta(\cos^2\theta - 2\sin^2\theta) \\
& + 4\eta''_{11}\eta''_{13}\cos^2\theta\sin^2\theta(\sin^2\theta - 2\cos^2\theta) + 12\eta''^2_{13}\cos^3\theta\sin^3\theta] \\
& + \frac{1}{2} C_{133}[\eta''^2_{11}\cos\theta\sin^3\theta(2\cos^2\theta - \sin^2\theta) \\
& + 4\eta''_{11}\eta''_{13}\cos^2\theta\sin^2\theta(\cos^2\theta - 2\sin^2\theta) - 12\eta''^2_{13}\cos^3\theta\sin^3\theta] \\
& + C_{144}[2\eta''^2_{11}\cos^3\theta\sin\theta(\cos^2\theta - 2\sin^2\theta) + 2\eta''_{11}\eta''_{13}\cos^6\theta \\
& + 10\eta''_{11}\eta''_{13}\sin^4\theta\cos^2\theta - 12\eta''_{11}\eta''_{13}\sin^2\theta\cos^4\theta - 2\eta''^2_{12}\cos\theta\sin^3\theta \\
& + 2\eta''^2_{13}\cos^5\theta\sin\theta + 2\eta''^2_{13}\cos\theta\sin^4\theta + 12\eta''^2_{13}\cos^3\theta\sin^3\theta] \\
& + \frac{1}{2} C_{166}[-3\eta''^2_{11}\cos^5\theta\sin\theta + 12\eta''_{11}\eta''_{13}\cos^4\theta\sin^2\theta \\
& - 12\eta''^2_{13}\cos^3\theta\sin^3\theta - 4\eta''^2_{12}\cos^3\theta\sin\theta] \\
& + \frac{1}{6} C_{266}[3\eta''^2_{11}\cos^5\theta\sin\theta\cos(2\phi)(3\sin^2(2\phi) - \cos^2(2\phi)) \\
& + 12\eta''_{11}\eta''_{13}\cos^4\theta\sin^2\theta\cos(2\phi)(\cos^2(2\phi) - 3\sin^2(2\phi)) \\
& + 12\eta''_{11}\eta''_{12}\cos^4\theta\sin\theta\sin(2\phi)(3\cos^2(2\phi) - \sin^2(2\phi)) \\
& + 12\eta''^2_{13}\cos^3\theta\sin^3\theta\cos(2\phi)(\cos^2(2\phi) + 3\sin^2(2\phi)) \\
& - 42\eta''^2_{12}\cos^3\theta\sin\theta\sin^2(2\phi)\cos(2\phi) - 2\eta''^2_{12}\cos^3\theta\sin\theta \\
& \cdot \cos(2\phi)(\cos^4\phi + \sin^4\phi) + 24\eta''_{12}\eta''_{13}\cos^3\theta\sin^2\theta\sin(2\phi) \\
& \cdot (\sin^2(2\phi) - 3\cos^2(2\phi))] + \frac{1}{6} C_{33}[3\eta''^2_{11}\sin^5\theta\cos\theta
\end{aligned}
$$

$$+ 12\eta''_{11}\eta''_{13}\sin^4\theta\cos^2\theta + \eta''^2_{13}\sin^3\theta\cos^2\theta]$$

$$+ C_{344}[2\eta''^2_{11}\cos\theta\sin^3\theta(2\cos^2\theta - \sin^2\theta)$$

$$+ \eta''_{11}\eta''_{13}\sin^2\theta(10\cos^4\theta + 2\sin^4\theta - 12\cos^2\theta\sin^2\theta)$$

$$+ 2\eta''^2_{12}\sin^3\theta\cos\theta + 4\eta''^2_{13}\cos\theta\sin\theta[\cos^4\theta + \sin^4\theta$$

$$- 3\cos^2\theta\sin^2\theta) + \frac{1}{2} C_{366}[\ {}''^2_{11}\cos^3\theta\sin\theta(\cos^2\theta - 2\sin^2\theta)$$

$$+ 4\eta''_{11}\eta''_{13}\cos^2\theta\sin^2\theta(\sin^2\theta - 2\cos^2\theta)$$

$$+ 12\eta''^2_{13}\cos^3\theta\sin^3\theta + 4\eta''^2_{12}\sin\theta\cos^3\theta]$$

$$+ 2C_{456}[2\eta''^2_{11}\cos^3\theta\sin\theta(\cos^2\theta - 2\sin^2\theta)$$

$$+ \eta''_{11}\eta''_{13}(-8\cos^2\theta\sin^2\theta\cos(2\theta) + \cos^6\theta + 12\cos^2\theta\sin^4\theta\cos^2(2\phi)$$

$$-16\cos^4\theta\sin^2\theta\cos^2(2\phi)) + \eta''^2_{13}\cos\theta\sin\theta\sin^2(2\phi)(12\cos^2\theta\sin^2\theta$$

$$- 6\sin^4\theta - 6\cos^4\theta) + 2\eta''_{11}\eta''_{12}\cos(2\phi)\sin(2\phi)\cos^2\theta\sin\theta$$

$$\cdot (\sin^2\theta - \cos^2\theta) + 4\eta''_{12}\eta''_{13}\cos^3\theta\sin^2\theta\cos(2\phi)\sin(2\phi)$$

$$- 2\eta''^2_{12}\sin^2(2\phi)\sin^3\theta\cos\theta + 4\eta''^2_{12}\cos\theta\sin\theta(\cos^2\theta - \sin^2\theta)$$

$$\cdot (\cos^4\phi + \sin^4\phi)] \qquad \text{(III-15)}$$

If we put these expressions for the strain derivatives of elastic energy into (II-41), we would get a set of wave equations for propagating along the a"-axis direction. It is predicted that the set of wave equations should be nonlinear and a perturbation approach is necessary in order to solve it.

Let us suppose the solutions of these nonlinear wave equations may be expressed in the following forms:

$$U'' = U''_1 + U''_2 + \dots$$
$$V'' = V''_1 + V''_2 + \dots \qquad \text{(III-16)}$$
$$W'' = W''_1 + W''_2 + \dots$$

where U''_1, V''_1, W''_1 and U''_2, V''_2, W''_2 represent the first and second approximation solutions of the displacements, respectively. First, we look at the first approximation equations. Putting (III-16) into (II-41) and using (III-7)-(III-15) and (II-34), we can obtain a set of first approximation wave equations for propagating along a"-axis direction as follows:

$$\rho_0\ddot{U}''_1 = (C_{11}\cos^4\theta + \frac{1}{2}C_{13}\sin^2(2\theta) + C_{33}\sin^4\theta$$
$$+ C_{44}\sin^2(2\theta))\frac{\partial^2 U''_1}{\partial a''^2} + \frac{1}{2}(-C_{11}\cos^2\theta\sin(2\theta)$$
$$+ C_{13}\cos(2\theta)\sin(2\theta) + C_{33}\sin^2\theta\sin(2\theta) + 2C_{44}\sin(2\theta)\cos(2\theta))\frac{\partial^2 W''_1}{\partial a''^2}$$

$$\rho_0\ddot{V}''_1 = \frac{1}{2}(C_{11}\cos^2\theta - C_{12}\cos^2\theta + 2C_{44}\sin^2\theta)\frac{\partial^2 V''_1}{\partial a''^2}$$

$$\rho_0\ddot{W}''_1 = \frac{1}{2}(-C_{11}\cos^2\theta\sin(2\theta) + C_{13}\sin(2\theta)\cos(2\theta)$$
$$+ C_{33}\sin^2\theta\sin(2\theta) + 2C_{44}\sin(2\theta)\cos(2\theta))\frac{\partial^2 U''_1}{\partial a''^2}$$
$$+ \frac{1}{2}[\frac{1}{2}C_{11}\sin^2(2\theta) - C_{13}\sin^2(2\theta) + \frac{1}{2}C_{33}\sin^2(2\theta) + 4C_{44}(\cos^4\theta$$
$$+ \sin^4\theta - \frac{1}{4}\sin^2(2\theta))]\frac{\partial^2 W''_1}{\partial a''^2} \qquad \text{(III-17)}$$

which can be rewritten as:

$$\left.\begin{aligned}
\rho_0 \ddot{U}_1 &= \alpha \frac{\partial^2 U_1''}{\partial a''^2} + \beta_2 \frac{\partial^2 W_1''}{\partial a''^2} \\
\rho_0 \ddot{V}_1 &= \gamma_1 \frac{\partial^2 V_1''}{\partial a''^2} \\
\rho_0 \ddot{W}_1 &= \gamma_2 \frac{\partial^2 W_1''}{\partial a''^2} + \beta_2 \frac{\partial^2 U_1''}{\partial a''^2}
\end{aligned}\right\} . \qquad \text{(III-18)}$$

Here

$$\left.\begin{aligned}
\alpha &= C_{11}\cos^4\theta + \frac{1}{2} C_{13}\sin^2(2\theta) + C_{33}\sin^4\theta + C_{44}\sin^2(2\theta) \\
\gamma_1 &= \frac{1}{2} (C_{11}\cos^2\theta - C_{12}\cos^2\theta + 2C_{44}\sin^2\theta) \\
\gamma_2 &= \frac{1}{4} C_{11}\sin^2(2\theta) - \frac{1}{2} C_{13}\sin^2(2\theta) + \frac{1}{4} C_{33}\sin^2(2\theta) \\
&\quad + 2C_{44}(1 - \frac{3}{4} \sin^2(2\theta)) \\
\beta_2 &= - \frac{1}{2} C_{11}\cos^2\theta\sin(2\theta) + \frac{1}{2} C_{13}\cos(2\theta)\sin(2\theta) \\
&\quad + \frac{1}{2} C_{33}\sin^2\theta\sin(2\theta) + C_{44}\sin(2\theta)\cos(2\theta)
\end{aligned}\right\} .$$

(III-19)

In the above equations the terms containing the coupling coefficient β_2 are called the coupling terms.

Obviously, because the coupling terms are existing in the set of wave equations, it becomes impossible to get the solution of the pure longitudinal vibration even when only the pure longitudinal vibration mode is excited at the surface of the transducer.

In order to seek the direction along which the pure longitudinal mode can propagate, we seek the directions along which the coupling coefficient β_2 vanishes (or is minimum). It is necessary to note that since the elastic constants C_{ij} are contained in the expression for the coupling coefficient, the direction we want to seek can be determined only for a specific crystal rather than in general terms. But the coupling coefficient β_2 is only a function of the angle θ, so it means that under the same angle θ the polar diagram of the coupling coefficient β_2 versus the angle ϕ should be a circle.

From (III-19) we may see immediately that β_2 vanishes when $\theta = n\pi/2$ ($n = 0, 1, 2, \ldots$). This reveals that the pure longitudinal mode can propagate along any radial direction in the original basal plane, as well as in any direction perpendicular to the original basal plane. Moreover, the above condition without coupling coefficient β_2 should work without exception for any hexagonal crystals, which is in coincidence with that in Technical Report No. 22 by J. Philip and M. A. Breazeale (7).

It is important to point out that, besides the above directions, another direction with vanishing β_2 also exists in hexagonal crystals. It is the direction along a cone whose apex angle $(\frac{\pi}{2} - \theta_p)$ is centered about the c-axis (in original coordinate system). However, as mentioned above, the apex angle is different for the different hexagonal crystals. Figure III-2 shows the directions of pure modes.

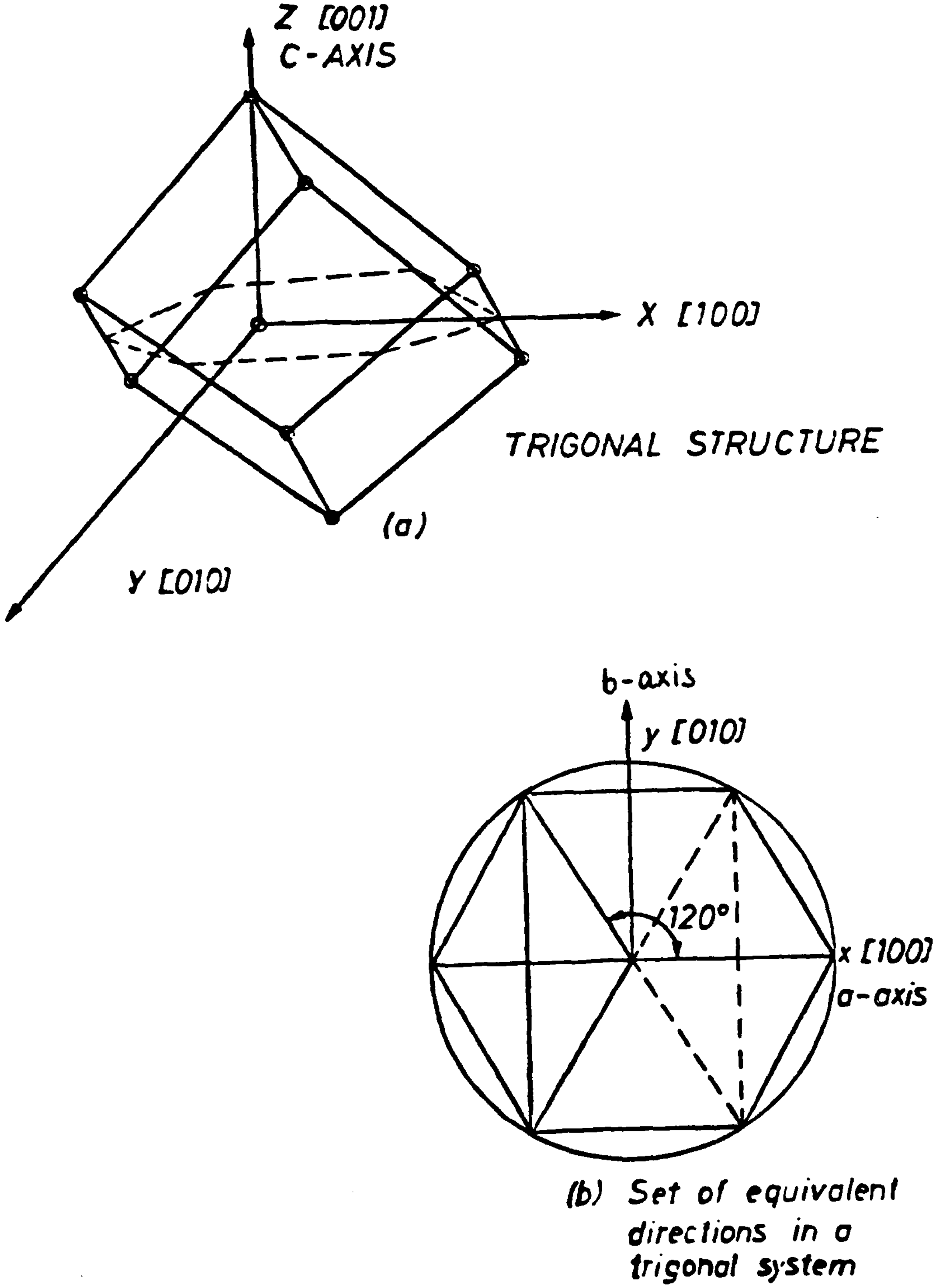

Figure IV-1. The coordinate system for crystals of trigonal symmetry.

$$+ (\eta_{23} + \eta_{32})(\eta_{12}^2 + \eta_{21}^2)$$

$$+ \frac{1}{2} C_{133}\eta_{33}^2(\eta_{11} + \eta_{22}) + C_{134}\eta_{33}\{(\eta_{11} - \eta_{22})(\eta_{23} + \eta_{32})$$

$$+ (\eta_{31} + \eta_{13})(\eta_{12} + \eta_{21})\} + C_{144}\ \{(\eta_{23}^2 + \eta_{32}^2)\eta_{11}$$

$$+ \eta_{22}(\eta_{31}^2 + \eta_{13}^2) + C_{155}\{\eta_{22}(\eta_{23}^2 + \eta_{32}^2) + \eta_{11}(\eta_{31}^2 + \eta_{13}^2)\}$$

$$+ \frac{1}{6} C_{222}\eta_{22}^3 + \frac{1}{6} C_{333}\eta_{33}^3 + C_{344}\eta_{33}\{(\eta_{23}^2 + \eta_{32}^2)$$

$$+ (\eta_{31}^2 + \eta_{13}^2)\} + C_{444}\{\tfrac{1}{6}(\eta_{23} + \eta_{32})^3 - (\eta_{23} + \eta_{32})(\eta_{31}^2 + \eta_{13}^2)\}$$

$$+ \frac{1}{2} (C_{111} + C_{112} - C_{222})\eta_{22}^2\eta_{11}$$

$$+ \frac{1}{2} (-C_{114} - 2C_{124})\eta_{22}^2(\eta_{23} + \eta_{32}) + \frac{1}{4} (-2C_{111} - C_{112} + 3C_{222})\eta_{11}$$

$$(\eta_{12}^2 + \eta_{21}^2) + \tfrac{1}{4}(2C_{111} - C_{112} - C_{222})$$

$$\times\ \eta_{22}(\eta_{12}^2 + \eta_{21}^2) + \tfrac{1}{2}(C_{113} - C_{123})\eta_{33}(\eta_{12}^2 + \eta_{21}^2) + \tfrac{1}{2}(C_{114}$$

$$+ 3C_{124})\eta_{11}(\eta_{31} + \eta_{13})(\eta_{12} + \eta_{21})$$

$$+ \tfrac{1}{2}(C_{114} - C_{124})\eta_{22}(\eta_{31} + \eta_{13})(\eta_{12} + \eta_{21}) + \tfrac{1}{2}(-C_{144} + C_{155})$$

$$(\eta_{23} + \eta_{32})(\eta_{31} + \eta_{13})(\eta_{12} + \eta_{21}) + \ldots \qquad \text{(IV-1)}$$

In this expression C_{111}, C_{112}, C_{113}, C_{114}, C_{124}, C_{144} and C_{222} are combinations of TOE constants given by

The unit cell of a crystal with the trigonal symmetry is shown in Figure IV-1. A drawing of the coordinate frame is also given in the figure. The six SOE constants are C_{11}, C_{12}, C_{13}, C_{14}, C_{33}, C_{44} and the fourteen TOE constants are C_{111}, C_{112}, C_{113}, C_{114}, C_{123}, C_{124}, C_{133}, C_{134}, C_{144}, C_{155}, C_{222}, C_{333}, C_{344}, C_{444}.

Some of the trigonal crystals (for instance, quartz and lithium niobate) exhibit piezoelectricity that affects their nonlinear behavior, and proper attention ultimately must be paid to it in this sort of crystals (10), especially for some strongly coupled piezoelectric trigonal crystals like $LiNbO_3$. In this report, however, we will concentrate on the mechanical properties and neglect the effect of piezoelectricity. Piezoelectricity was treated to some extent in Technical Report No. 23.

For the nonpiezoelectric trigonal crystal, Kaga (11) has given the elastic strain energy density with Brugger definition of third-order elastic constants. After making some corrections (3), the energy density can be expressed as,

$$
\begin{aligned}
\phi = &\frac{1}{2} C_{11}(\eta_{11}^2 + \eta_{22}^2) + C_{12}\eta_{11}\eta_{22} + C_{13}(\eta_{22}\eta_{33} + \eta_{33}\eta_{11}) \\
&+ C_{14}\{(\eta_{11} - \eta_{22})(\eta_{23} + \eta_{32}) + (\eta_{31} + \eta_{13})(\eta_{12} + \eta_{21})\} \\
&+ \frac{1}{2} C_{33}\eta_{33}^2 + C_{44}(\eta_{23}^2 + \eta_{32}^2 + \eta_{31}^2 + \eta_{13}^2) \\
&+ \frac{1}{2} (C_{11} - C_{12})(\eta_{12}^2 + \eta_{21}^2) \\
&+ \frac{1}{6} C_{111}\eta_{11}^3 + \frac{1}{2} C_{112}\eta_{11}^2\eta_{22} + \frac{1}{2} C_{113}(\eta_{11}^2\eta_{33} + \eta_{22}^2\eta_{33}) \\
&+ \frac{1}{2} C_{114}\eta_{11}^2(\eta_{23} + \eta_{32}) + C_{123}\eta_{11}\eta_{22}\eta_{33} + C_{124}\{\eta_{11}\eta_{22}(\eta_{23} + \eta_{32})
\end{aligned}
$$

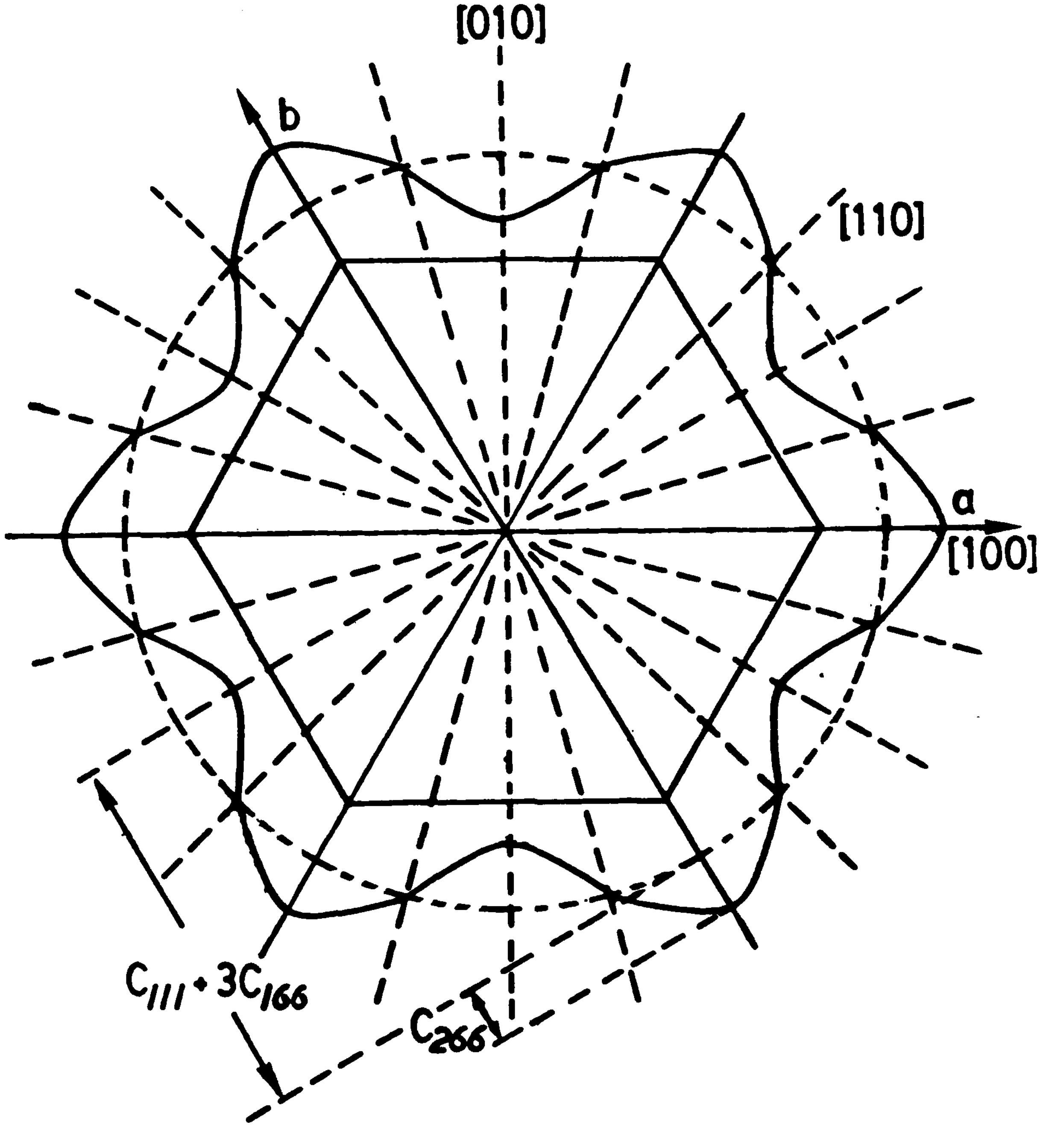

Figure III-5. Magnitude of the K_3 parameter plotted as a function of angle in the basal plane.

negative of the ratio of the nonlinear term to the linear term in the nonlinear wave equation of pure longitudinal mode [Eq. (III-34)]. It is given by

$$\beta = -\frac{\delta}{\alpha} \quad . \tag{III-36}$$

In terms of A_1 and A_2, it can be rewritten as,

$$\beta = \left(\frac{A_2}{A_1^2}\right) \frac{8}{k_L^2 a''} \quad . \tag{III-37}$$

Therefore, by measuring A_1 and A_2 one can determine β which can be used to evaluate the δ's which are combinations of SOE and TOE constants, or the K_3's which are combinations of TOE constants only.

The parameters K_2 and K_3 for the specific directions of pure longitudinal mode also are written in tabular form in Table III-1, in which under K_3 we indicate those specific angles ϕ which lead to magnitudes of the quantity $\cos(2\phi)[\cos^2(2\phi) - 3\sin^2 2\phi]$ of 1, 0, or -1. These would be directions along which measurements will yield the most useful information. This behavior of K_3 in the basal plane was given in graphical form in Figure 2 of Technical Report No. 22, which we reproduce here for convenience (Figure III-5).

IV. PURE MODE PROPAGATION IN TRIGONAL CRYSTALS

There are also two classes of crystals with trigonal symmetry, one with the Hermann-Mauguin symbol $3\bar{3}$ and the other with the symbol 32, 3m, $\bar{3}$m. The first class has seven SOE constants and twenty TOE constants, and the second has six SOE constants and fourteen TOE constants. In this technical report we limit our investigation to the latter.

$$\alpha = K_2$$

and (III-32)

$$\delta = (3K_2 + K_3) \ .$$

Thus,

$$K_3 = \sigma = C_{111}\cos^6\theta + 3C_{113}\sin^2\theta\cos^4\theta + 3C_{133}\sin^4\theta\cos^2\theta$$
$$+ 12C_{144}\cos^4\theta\sin^2\theta + 3C_{166}\cos^6\theta + C_{266}\cos^6\theta\cos(2\phi)$$
$$\cdot (\cos^2(2\phi) - 3\sin^2(2\phi)) + C_{333}\sin^6\theta + 12C_{344}\cos^2\theta\sin^4\theta$$
$$+ 3C_{366}\sin^2\theta\cos^4\theta + 24C_{456}\cos^4\theta\sin^2\theta \ . \qquad \text{(III-33)}$$

Now, we may simplify Eq. (II-41) and, as a matter of fact, write the nonlinear wave equation of pure longitudinal mode as

$$\rho_0\ddot{U}'' = \alpha \frac{\partial^2 U''}{\partial a''^2} + \delta \frac{\partial U''}{\partial a''} \cdot \frac{\partial^2 U''}{\partial a''^2} \ . \qquad \text{(III-34)}$$

The solution to the nonlinear wave equation can be expressed as

$$U'' = U''_1 + U''_2$$
$$= A_1\sin(k_L a'' - \omega t) - A_2\cos2(k_L a'' - \omega t) \ . \qquad \text{(III-35)}$$

Here $A_2 = [\frac{\delta}{8\alpha}](k_L A_1)^2 a''$ is the amplitude of the generated second harmonic wave. The solution (III-35) is in complete analogy to cubic crystals; however, the expressions for α and δ, or alternatively K_2 and K_3 must be examined for hexagonal symmetry.

As in the case of cubic crystals (7), we can define the ultrasonic nonlinearity parameter for hexagonal crystals as the

Now, equating the coefficients on both sides of the equation we get

$$- \omega^2 \rho_0 Ba'' + k_L^2 \alpha Ba'' + k_L \alpha C = - \frac{1}{8} \delta \, k_L^3 A_1^2$$

and (III-26)

$$- \omega^2 \rho_0 Ca'' - k_L \alpha B + K_L^2 \alpha Ca'' = 0 \, .$$

But $\alpha = \rho_0 C_L^2$ and $\omega = C_L k_L$, so Eqs. (III-26) become

$$- C_L^2 k_L^2 \rho_0 Ba'' + k_L^2 \rho_0 C_L^2 Ba'' + k_L \rho_0 C_L^2 C = - \frac{\delta}{8} k_L^3 A_1^2 \qquad \text{(III-27)}$$

and

$$C_L^2 k_L^2 \rho_0 Ca'' - k_L \rho_0 C_L^2 B + k_L^2 \rho_0 C_L^2 Ca'' = 0 \, . \qquad \text{(III-28)}$$

Dividing Eq. (III-27) by $k_L \rho_0 C_L^2$, we get

$$C = - \frac{\delta (k_L A_1)^2}{\rho_0 C_L^2} \, . \qquad \text{(III-29)}$$

Also, dividing Eq. (III-28) by $k \rho_0 C_L{}^2$, we get

$$B = 0 \, . \qquad \text{(III-30)}$$

So, the solution to the wave equation of second approximation (III-22) becomes

$$U_2''(a'',t) = - \left[\frac{\delta (k_L A_1)^2}{8 \rho_0 C_L{}^2}\right] a'' \cos 2(k_L a'' - \omega t) \qquad \text{(III-31)}$$

If we use the same symbols as Breazeale and Ford (9) used before for cubic crystals, we can put

$$\rho_0\ddot{U}_2'' = \alpha \frac{\partial^2 U_2''}{\partial a''^2} + \delta \frac{\partial U_1''}{\partial a''} \frac{\partial^2 U_1''}{\partial a''^2} \qquad \text{(III-21)}$$

or

$$\rho_0\ddot{U}_2'' - \alpha \frac{\partial^2 U_2''}{\partial a''^2} = -\frac{1}{2}\,\delta k_L^3 A_1^2 \sin 2(k_L a'' - \omega t)\ . \qquad \text{(III-22)}$$

Here

$$\delta = 3\alpha + \sigma \qquad \text{(III-23)}$$

and

$$\begin{aligned}\sigma = {} & C_{111}\cos^6\theta + 2C_{113}\sin^2\theta\cos^4\theta + 3C_{133}\sin^4\theta\cos^2\theta \\ & + 12C_{144}\cos^4\theta\sin^2\theta + 3C_{166}\cos^6\theta + C_{266}\cos^6\theta\cos(2\phi) \\ & \cdot((\cos^2(2\phi) - 3\sin^2(2\phi)) + C_{333}\sin^6\theta + 12C_{344}\cos^2\theta\sin^4\theta \\ & + 3C_{366}\sin^2\theta\cos^4\theta + 24C_{456}\cos^4\theta\sin^2\theta\ . \end{aligned} \qquad \text{(III-24)}$$

Taking into account the boundary condition for U_2', that is, $U_2'' = 0$ where $a'' = 0$, we use a trial solution in the following form:

$$U_2'' = Ba''\sin 2(k_L a'' - \omega t) + Ca''\cos 2(k_L a'' - \omega t) \qquad \text{(III-25)}$$

where B and C are the coefficients to be determined. Putting the solution into (III-22), we get

$$\begin{aligned} & 4(-\omega^2\rho_0 Ba'' + k_L^2\alpha Ba'' + k_L\alpha C)\sin 2(k_L a'' - \omega t) \\ & + 4(-\omega^2\rho_0 Ca'' - k_L\alpha B + k_L^2\alpha Ca'')\cos 2(k_L a'' - \omega t) \\ & = -\frac{\delta k^3 A_1^2}{2}\sin^2(k_L a'' - \omega t)\ . \end{aligned}$$

Table III-1. The K_2 and K_3 parameters for hexagonal crystals along the direction of the pure longitudinal mode.

Direction of Wave Propagation θ	$K_2 = \alpha$	$K_3 = (\delta - 3\alpha)$		
			$\cos(2\phi)(\cos^2(2\phi) - 3\sin^2(2\phi))$	ϕ
0	C_{11}	$C_{111} + 3C_{166}$ $+ C_{266}\cos(2\phi)(\cos^2(2\phi) - 3\sin^2(2\phi))$	1	$2n(\frac{\pi}{12}), n=0,2,4,\ldots$
			0	$(2n+1)\frac{\pi}{12}, n=0,1,2,\ldots$
			-1	$2n(\frac{\pi}{12}), n=1,3,\ldots$
$\pi/2$	C_{33}	C_{333}		
θ_p	$C_{11}\cos^4\theta_p + \frac{1}{2} C_{13}\sin^2(2\theta_p) + C_{33}\sin^4\theta_p + C_{44}\sin^2(2\theta_p)$	$C_{111}\cos^6\theta_p + 3C_{113}\sin^2\theta_p\cos^4\theta_p + 3C_{133}\sin^4\theta_p\cos^2\theta_p + 12C_{144}\cos^4\theta_p\sin^2\theta_p + 3C_{166}\cos^6\theta_p + C_{266}\cos^6\theta_p\cos(2\phi)(\cos^2(2\phi) - 3\sin^2(2\phi)) + C_{333}\sin^6\theta_p + 12C_{344}\cos^2\theta_p\sin^4\theta_p + 3C_{366}\sin^2\theta_p\cos^4\theta_p + 24C_{456}\cos^4\theta_p\sin^2\theta_p$		

The coefficient α in Eq. (III-19) may be used to determine the velocity of sound or the slowness of the longitudinal wave in the hexagonal crystals. Because the α is also the same for different angle ϕ, the velocity of sound or the slowness versus the angle ϕ also should be a circle.

Table III-1 describes the relation of α with angle θ at some specific direction along which pure longitudinal waves can propagate in hexagonal crystals.

As soon as the directions of the pure longitudinal mode are determined, it becomes possible to derive the nonlinear longitudinal wave equation whose solution describes growth of the second harmonic of a pure longitudinal wave. The TOE constants can be obtained from the experiment described by the solution to the nonlinear wave equation.

Suppose we have $\beta_2 = 0$ for the specific directions. Thus under the boundary condition of the pure longitudinal vibration at the surface of the transducer, that is $U_1'' = A_1 \sin\omega t$, $V_1'' = W_1'' = 0$ when $a'' = 0$, we may get the solutions of Eq. (III-18) as:

$$\begin{aligned} U_1'' &= A_1 \sin(k_L a'' - \omega t) \\ V_1'' &= W_1'' = 0 \,. \end{aligned} \qquad \text{(III-20)}$$

Here $k_L = \frac{C_L}{\omega}$, $C_L = \sqrt{\frac{\alpha}{\rho_0}}$ is the velocity of longitudinal waves.

Putting Eqs. (III-7)-(III-15) and (II-34) into (II-41) and using (III-16) with (III-20), we can get the second approximation equation of the pure longitudinal mode.

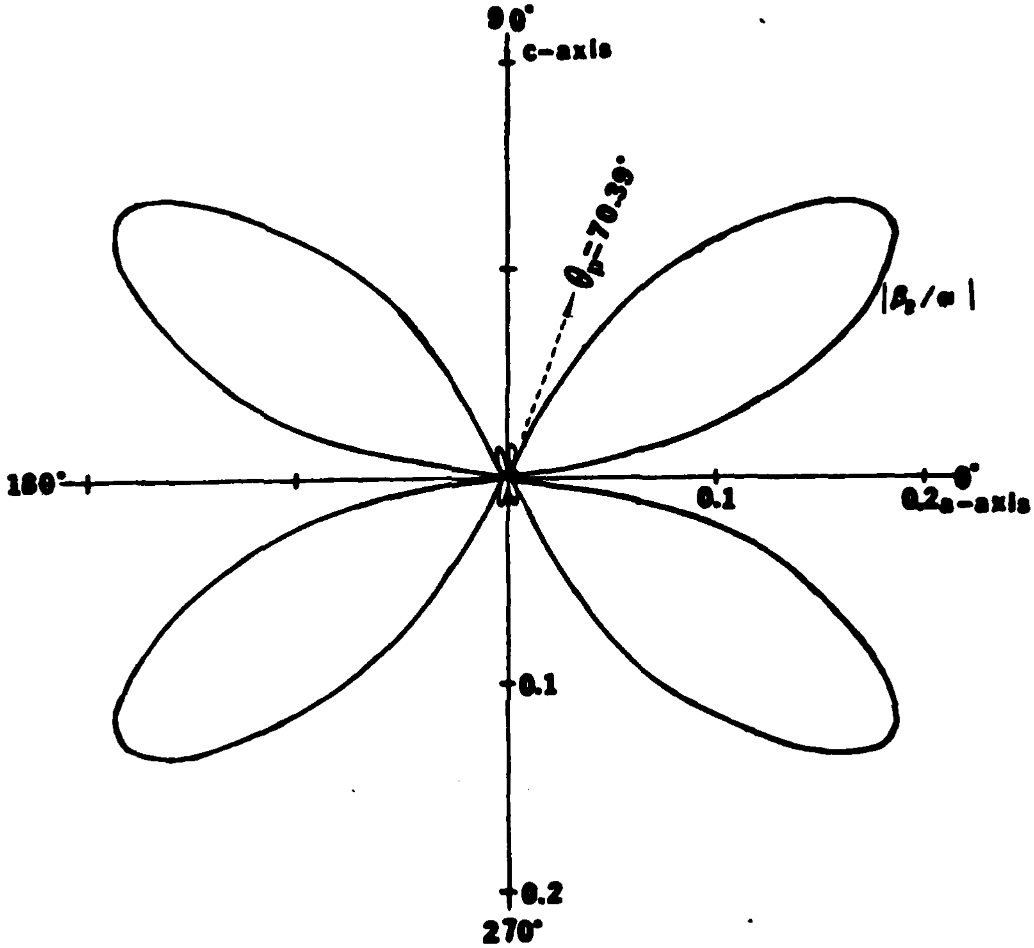

Figure III-4. Coupling coefficient β_2 versus angle θ for ZnO.

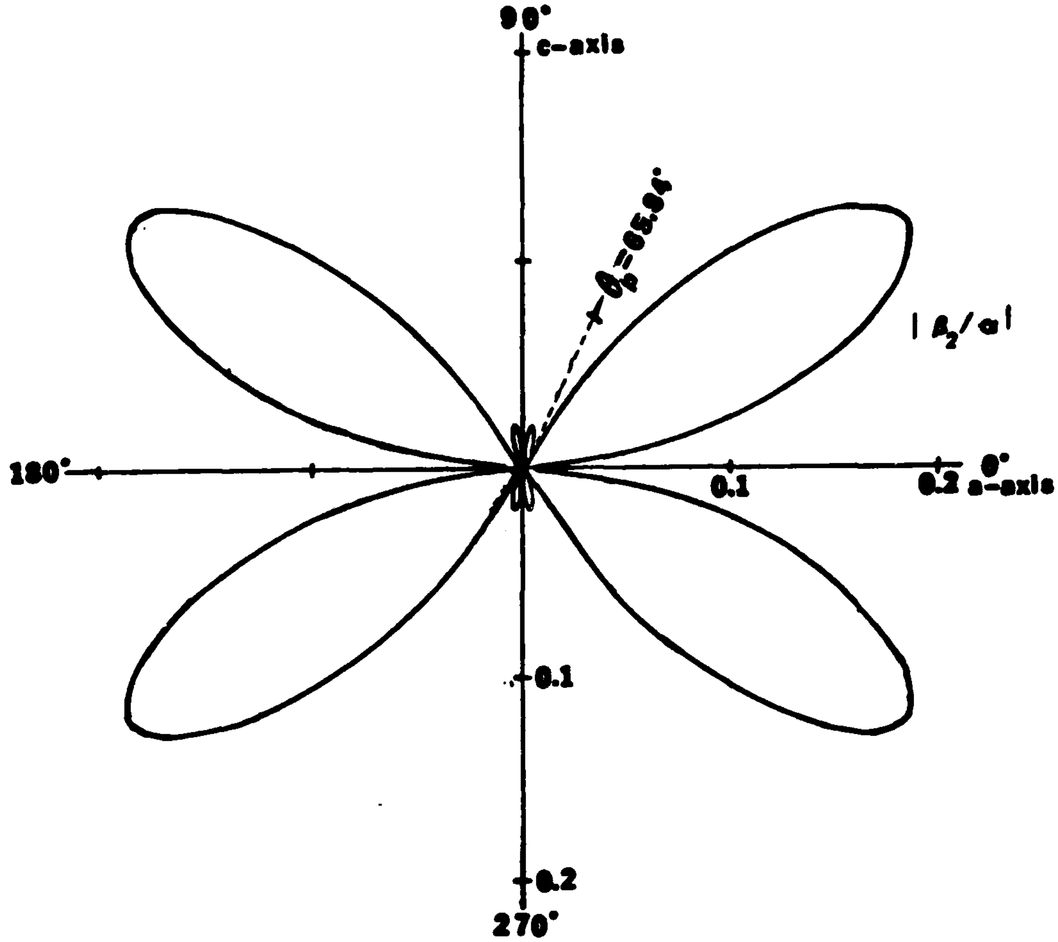

Figure III-3. Coupling coefficient β versus angle θ for CdS.

An estimate of the magnitude of the apex angle can be made for specific crystals. If one ignores the piezoelectric terms, one can make such an estimate for CdS, one of the most interesting hexagonal crystals. The SOE constants of CdS are given by Landold-Börnstein (3) as

$$C_{11} = 87 \quad \text{GPa}$$
$$C_{12} = -54.6 \quad \text{GPa}$$
$$C_{13} = 47.5 \quad \text{GPa}$$
$$C_{33} = 94.1 \quad \text{GPa}$$
$$C_{44} = 14.9 \quad \text{GPa}$$

So we can numerically figure out from (III-19), when $\theta = \theta_p = 65.836°$ or the apex angle $90°-65.836° = 24.164°$, then $\eta\beta_2$ vanishes.

ZnO is another interesting sample. Its SOE constants may be found in the same reference (3). They are:

$$C_{11} = 209 \quad \text{GPa}$$
$$C_{12} = 120 \quad \text{GPa}$$
$$C_{13} = 104 \quad \text{GPa}$$
$$C_{33} = 218 \quad \text{GPa}$$
$$C_{44} = 44.1 \quad \text{GPa}$$

So we can numerically figure out from (III-19), when $\theta = \theta_p = 70.390°$ or the apex angle $90°-70.390° = 29.610°$, then $\eta\beta_2$ vanishes. Figure III-3 and Figure III-4 show the coupling coefficient (β_2/α) versus the angle θ for CdS and ZnO, respectively. In each figure we indicate the pure mode directions along which β_2 vanishes.

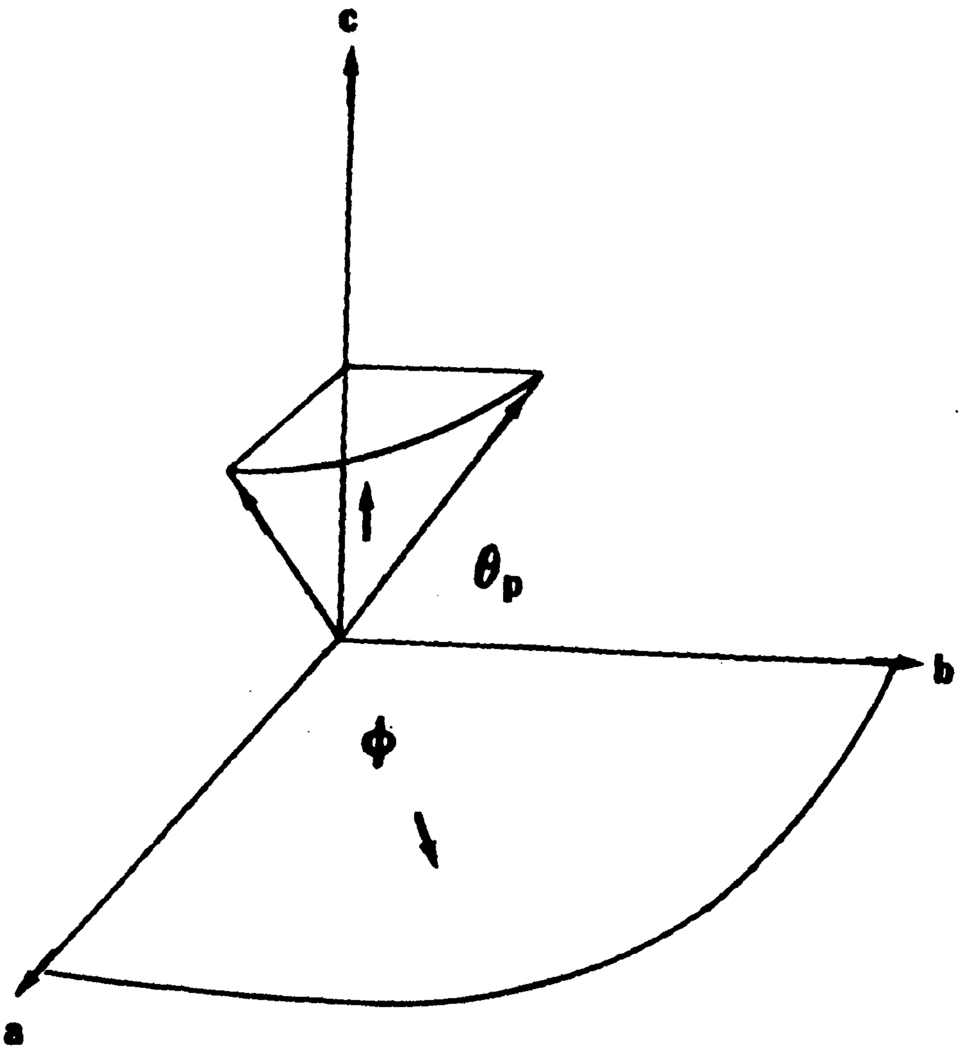

Figure III-2. Pure longitudinal mode directions in hexagonal crystals (indicated by arrows).

$$C_{111} = 4C_{266} + C_{112}$$
$$C_{112} = C_{122} - C_{111} + C_{222}$$
$$C_{113} = 2C_{366} + C_{123}$$
$$C_{114} = 2C_{156} - 3C_{124} \qquad \text{(IV-2)}$$
$$C_{124} = -C_{224} + C_{114}$$
$$C_{144} = -2C_{456} + C_{155}$$
$$C_{222} = 4C_{166} + 2C_{111} + C_{112}$$

If we substitute (II-36) into (IV-1), we would be able to give the complete expression for the strain energy density for the crystal of trigonal symmetry in the twice-rotated, or double primed, coordinate system as was given in the previous section for the crystal of hexagonal symmetry. But we already know that if our only concern is the generation of the second harmonic of a pure longitudinal mode, it is sufficient to write only the terms containing η''^3_{11} in the expression ϕ_3. This simplifies the expressions a great deal.

First, let us look at the expression ϕ_2. By substituting (II-36) into (IV-1), the expression ϕ_2 can be given in the double primed coordinate system as:

$$\phi_2 = \frac{1}{2} C_{11}[\eta''^2_{11}\cos^4\theta + (\eta''^2_{12} + \eta''^2_{21})\cos^2\theta - 2\eta''_{11}(\eta''_{13} + \eta''_{31})\sin\theta\cos^3\theta$$
$$+ (\eta''_{13} + \eta''_{31})^2\cos^2\theta\sin^2\theta] + \frac{1}{2} C_{12}(\eta''^2_{12} + \eta''^2_{21})\cos^2\theta$$
$$+ C_{13}[\eta''^2_{11}\cos^2\theta\sin^2\theta + \eta''_{11}(\eta''_{13} + \eta''_{31})\cos\theta\sin\theta(\cos^2\theta - \sin^2\theta)$$
$$- (\eta''_{13} + \eta''_{31})^2\cos^2\theta\sin^2\theta] + C_{14}[\eta''^2_{11}\sin(2\theta)\cos^2\theta\sin\phi(3\cos^2\phi - \sin^2\phi)$$
$$+ \eta''_{11}(\eta''_{13} + \eta''_{31})\cos^2\theta(\cos^2\theta - 3\sin^2\theta)\sin\phi(3\cos^2\phi - \sin^2\phi)$$

$$+ 3\eta''_{11}(\eta''_{12} + \eta''_{21})\cos^2\theta\sin\theta\cos\phi(\cos^2\phi - 3\sin^2\phi)$$

$$+ \frac{1}{2}(\eta''_{13} + \eta''_{31})^2\cos(2\theta)\sin(2\theta)\sin\phi(\sin^2\phi - 2\cos^2\phi)$$

$$+ (\eta''_{12} + \eta''_{21})^2\cos\theta\sin\theta\sin\phi(\sin^2\phi - 3\cos^2\phi)$$

$$+ (\eta''_{12} + \eta''_{21})(\eta''_{13} + \eta''_{31})\cos\theta(\cos^2\theta - 2\sin^2\theta)(\cos^2\phi - 3\sin^2\phi)\cos\phi]$$

$$+ \frac{1}{2} C_{33}[\eta''^2_{11}\sin^4\theta + 2\eta''_{11}(\eta''_{13} + \eta''_{31})\sin^3\theta\cos\theta$$

$$+ (\eta''_{13} + \eta''_{31})^2\sin^2\theta\cos^2\theta] + C_{44}[2\eta''^2_{11}\cos^2\theta\sin^2\theta$$

$$+ (\eta''^2_{12} + \eta''^2_{21})\sin^2\theta + 2\eta''_{11}(\eta''_{13} + \eta''_{31})\cos\theta\sin\theta$$

$$\times (\cos^2\theta - \sin^2\theta) + (\eta''_{13} + \eta''_{31})^2(\cos^4\theta + \sin^4\theta)$$

$$- 4\eta''_{13}\eta''_{31}\cos^2\theta\sin^2\theta] \ . \qquad \text{(IV-3)}$$

The strain derivatives can be calculated and simplified by taking into account $\eta''_{12} = \eta''_{21}$ and $\eta''_{13} = \eta''_{31}$ as:

$$\frac{\partial\phi_2}{\partial\eta''_{11}} = [C_{11}\cos^4\theta + 2C_{13}\cos^2\theta\sin^2\theta + 2C_{14}\sin(2\theta)\cos^2\theta\sin\phi(3\cos^2\phi - \sin^2\phi)$$

$$+ C_{33}\sin^4\theta + C_{44}\sin^2(2\theta)]\eta''_{11} + [-C_{11}\cos\theta\sin(2\theta)$$

$$+ C_{13}\cos(2\theta)\sin(2\theta) + 2C_{14}\cos^2\theta(\cos^2\theta - 3\sin^2\theta)\sin\phi$$

$$\times (3\cos^2\phi - \sin^2\phi) + C_{33}\sin^2\theta\sin(2\theta) + C_{44}\sin(4\theta)]\eta''_{13}$$

$$+ [6\cos^2\theta\sin\theta\cos\phi(\cos^2\phi - 3\sin^2\phi)]\eta''_{12} \ . \qquad \text{(IV-4)}$$

$$\frac{\partial \phi_2}{\partial \eta''_{21}} = [3C_{14}\cos^2\theta\sin\theta\cos\phi(\cos^2\phi - 3\sin^2\phi)]\eta''_{11} + [C_{11}\cos^2\theta - C_{12}\cos^2\theta$$

$$+ 4C_{14}\cos\theta\sin\theta\sin\phi(\sin^2\phi - 3\cos^2\phi) + 2C_{44}\sin^2\theta]\eta''_{12}$$

$$+ [2C_{14}\cos\theta(\cos^2\theta - 2\sin^2\theta)\cos\phi(\cos^2\phi - 3\sin^2\phi)]\eta''_{13} \,, \qquad \text{(IV-5)}$$

$$\frac{\partial \phi_2}{\partial \eta''_{31}} = [-C_{11}\cos^3\theta\sin\theta + \frac{1}{2} C_{13}\sin(2\theta)\cos(2\theta) + C_{14}\cos^2\theta(\cos^2\theta - 3\sin^2\theta)$$

$$\cdot \sin\phi(3\cos^2\phi - \sin^2\phi) + C_{33}\sin^3\theta\cos\theta + C_{44}\sin(2\theta)\cos(2\theta)]\eta''_{11}$$

$$+ [2C_{14}\cos\theta(\cos^2\theta - 2\sin^2\theta)\cos\phi(\cos^2\phi - 3\sin^2\phi)]\eta''_{12}$$

$$+ [2C_{11}\cos^2\theta\sin^2\theta - 4C_{13}\cos^2\theta\sin^2\theta + 2C_{14}\cos(2\theta)\sin(2\theta)$$

$$\cdot (\sin^2\phi - 2\cos^2\phi)\sin\phi + 2C_{33}\cos^2\theta\sin^2\theta + 4C_{44}$$

$$\cdot (\cos^4\theta + \sin^4\theta - \sin^2\theta\cos^2\theta)]\eta''_{13} \,. \qquad \text{(IV-6)}$$

Carrying out a similar procedure as was done for hexagonal crystals, we get a set of first approximation wave equations for propagation along the a"-axis in trigonal crystals:

$$\rho_0 \ddot{U}''_1 = \alpha \frac{\partial^2 U''_1}{\partial a''^2} + \beta_1 \frac{\partial^2 V''_1}{\partial a''^2} + \beta_2 \frac{\partial^2 W''_1}{\partial a''^2}$$

$$\rho_0 \ddot{V}''_1 = \gamma_1 \frac{\partial^2 V''_1}{\partial a''^2} + \beta_1 \frac{\partial^2 U''^2_1}{\partial a''^2} + \beta_3 \frac{\partial^2 W''_1}{\partial a''^2} \qquad \text{(IV-7)}$$

$$\rho_0 \ddot{W}''_1 = \gamma_2 \frac{\partial^2 W''_1}{\partial a''^2} + \beta_2 \frac{\partial U''^2_1}{\partial a''^2} + \beta_3 \frac{\partial^2 V''_1}{\partial a''^2}$$

where

$$\alpha = C_{11}\cos^4\theta + \frac{1}{2}C_{13}\sin^2(2\theta) + 2C_{14}\sin(2\theta)\cos^2\theta\sin\phi(3\cos^2\phi - \sin^2\phi)$$
$$+ C_{33}\sin^4\theta + C_{44}\sin^2(2\theta)$$
$$\gamma_1 = \frac{1}{2}C_{11}\cos^2\theta - \frac{1}{2}C_{12}\cos^2\theta + 2C_{14}\cos\theta\sin\theta\sin\phi(\sin^2\phi - 3\cos^2\phi)$$
$$+ C_{44}\sin^2\theta$$
$$\gamma_2 = \frac{1}{4}C_{11}\sin^2(2\theta) - \frac{1}{2}C_{13}\sin^2(2\theta) + C_{14}\cos(2\theta)\sin(2\theta)(\sin^2\phi - 2\cos^2\phi)$$
$$\cdot \sin\phi + \frac{1}{4}C_{33}\sin^2(2\theta) + 2C_{44}(1 - \frac{3}{4}\sin^2(2\theta))$$
$$\beta_1 = 3C_{14}\cos^2\theta\sin\theta\cos\phi(\cos^2\phi - 3\sin^2\phi) \qquad \text{(IV-8)}$$
$$\beta_2 = -\frac{1}{2}C_{11}\cos^2\theta\sin(2\theta) + \frac{1}{2}C_{13}\cos(2\theta)\sin(2\theta)$$
$$+ C_{14}\cos^2\theta(\cos^2\theta - 3\sin^2\theta)\sin\phi(3\cos^2\phi - \sin^2\phi)$$
$$+ \frac{1}{2}C_{33}\sin^2\theta\sin(2\theta) + \frac{1}{2}C_{44}\sin(4\theta)$$
$$\beta_3 = C_{14}\cos\theta(\cos^2\theta - 2\sin^2\theta)\cos\phi(\cos^2\phi - 3\sin^2\phi)$$

From the above expressions we can see that there are two coupling terms (β_1 and β_2) existing in the longitudinal wave equation [the first of Eq. (IV-8)]. This means that longitudinal waves excited by the transducer may couple to the transverse waves of both perpendicular and horizontal polarization rather than only one polarization as it does in the hexagonal crystals. So in this case we seek directions along which both of the coupling terms vanish instead of only one term as in the last section.

First, let us look at what happens in the basal plane. When $\theta = 0$, we have

$$\beta_1 = 0$$

and (IV-9)

$$\beta_2 = C_{14}\sin\phi(3\cos^2\phi - \sin^2\phi) .$$

Further,

$$\beta_2 = 0, \text{ when } \phi = n\frac{\pi}{3}, \quad n = 0, 1, 2, \ldots \qquad \text{(IV-10)}$$

This means that both β_1 and β_2 vanish and the pure longitudinal mode can propagate in trigonal crystals only along two special directions, $\phi = 0$ and $\pi/3$, in the basal plane, not all directions like in hexagonal crystals.

Second, we can find another direction of the pure longitudinal mode, which is

$$\theta = (2n + 1)\frac{\pi}{2} \qquad n = 0, 1, 2, 3 \ldots$$

Obviously, all of the directions of the pure longitudinal mode we found above are common for any trigonal crystals without exception no matter what their SOE constants are. It was pointed out (6) that, besides the above directions, in the b-c plane, i.e., $\phi = \pi/2$, one unique direction may also be discovered with vanishing β_1 and β_2. Fortunately, we have already had the possibility to find it quantitatively. It is clear that when $\phi = \pi/2$, β_1 vanishes and β_2 is reduced to

$$\beta_2 = -\frac{1}{2}C_{11}\cos^2\theta\sin(2\theta) + \frac{1}{2}C_{13}\cos(2\theta)\sin(2\theta)$$
$$- C_{14}\cos^2\theta(\cos^2\theta - 3\sin^2\theta) + \frac{1}{2}C_{33}\sin^2\theta\sin(2\theta)$$
$$+ \frac{1}{4}C_{44}\sin(4\theta) \quad \text{(IV-11)}$$

Thus, the direction we seek is dependent on the magnitude of the second order elastic constants of the sample being measured.

The quartz is a sample whose nonlinear mechanical properties are of wide interest. The SOE constants can be found (3) for quartz as:

$$\begin{aligned} C_{11} &= 86.6 \text{ GPa} \\ C_{12} &= 6.7 \text{ GPa} \\ C_{13} &= 12.6 \text{ GPa} \\ C_{14} &= -17.8 \text{ GPa} \\ C_{33} &= 106.1 \text{ GPa} \\ C_{44} &= 57.8 \text{ GPa} . \end{aligned} \quad \text{(IV-12)}$$

The direction with vanishing β_2 can be determined numerically in the b-c plane from (IV-11) as

$$\theta_p = 20.444° .$$

Figure IV-2 shows the diagram of (β_2/α) versus angle θ in the b-c plane for quartz. Lithium niobate might be another interesting sample. Its SOE constants can be written (3) as follows:

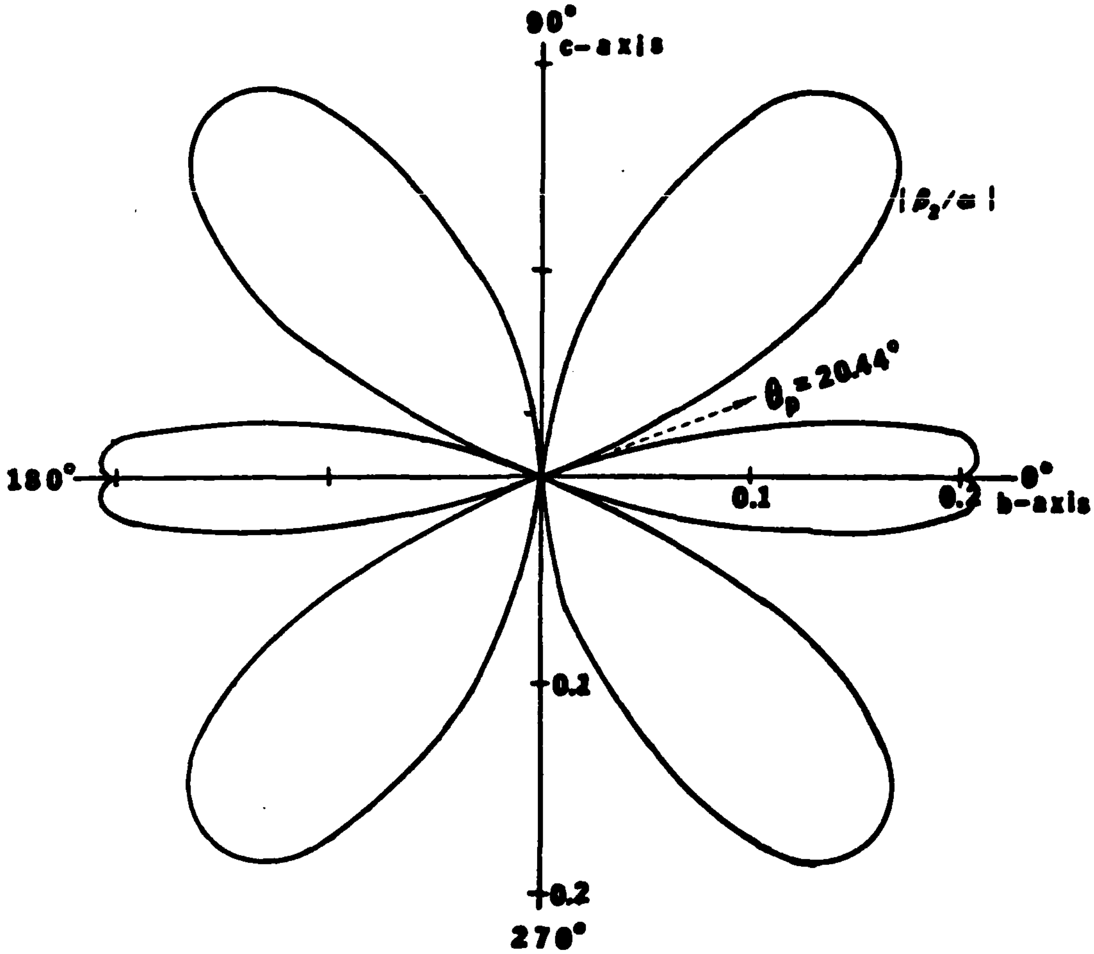

Figure IV-2. Coupling coefficient $|\beta/\alpha|$ versus angle θ for SiO_2 (at $\phi = (2n+1)\pi/2$).

$$C_{11} = 202.0 \text{ GPa}$$
$$C_{12} = 55.0 \text{ GPa}$$
$$C_{13} = 71.0 \text{ GPa}$$
$$C_{14} = 8.3 \text{ GPa} \qquad \text{(IV-13)}$$
$$C_{33} = 242.0 \text{ GP}$$
$$C_{44} = 60.1 \text{ GPa}$$

The angle θ_p with vanishing β_2 gives

$$\theta_p = 69.812° \ .$$

Figure IV-3 shows the diagram of (β_2/α) versus angle θ in the b-c plane for lithium niobate. Finally, we give a summary in Figure IV-4 which shows the direction of pure modes in trigonal crystals in general.

So far we have determined all of the directions of pure longitudinal modes in the crystals of quartz and lithium niobate. In the direction with vanishing β_1 and β_2, the solution of the first approximation wave equation can be worked out with the boundary condition when a" = 0. $U_1 = A_1\sin(\omega t)$ can be written in the same form as (III-20):

$$U_1 = A_1\sin(\omega t - k_L a")$$
$$\text{(IV-14)}$$
$$V_1 = W_1 = 0 \ .$$

Using a similar procedure as in the last section, we may get the second approximation wave equation of the pure longitudinal mode for the trigonal crystals. But, as we already have seen

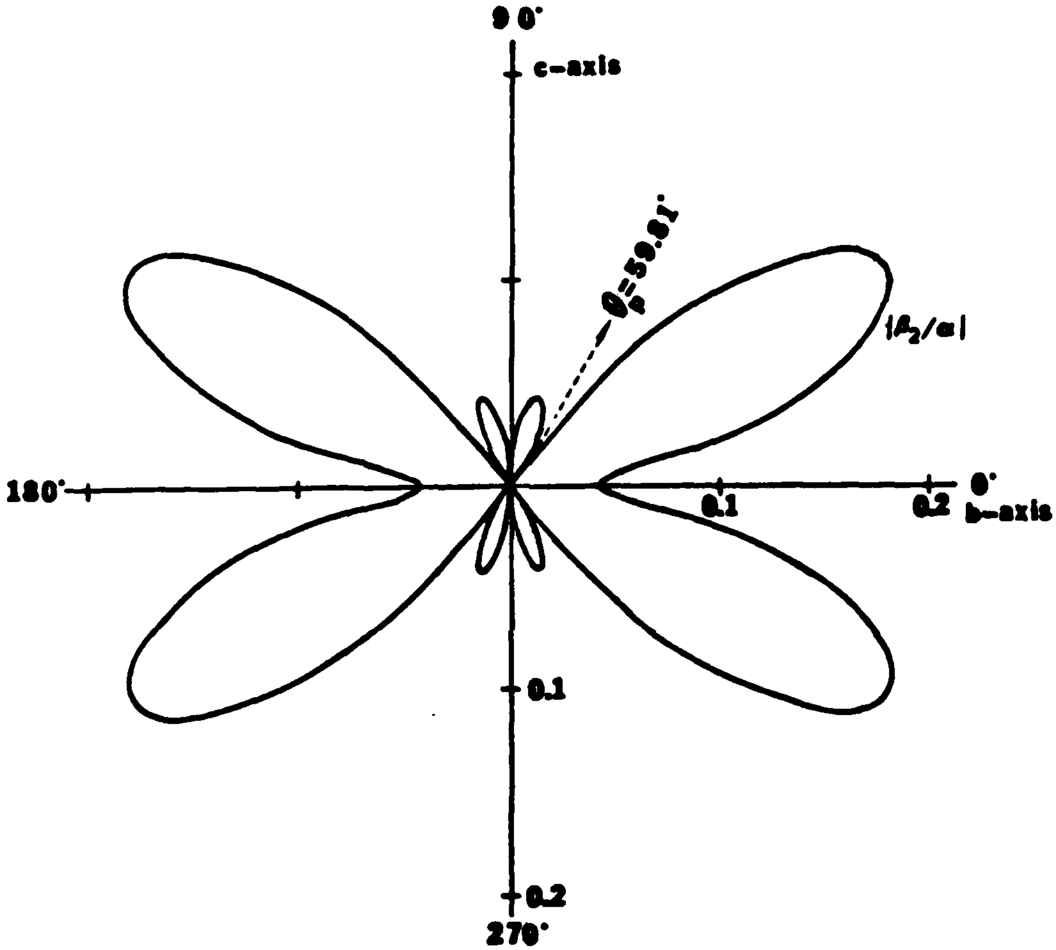

Figure IV-3. Coupling coefficient $|\beta_2/\alpha|$ versus angle θ for $LiNbO_3$ (at $\phi = (2n+1)\pi/2$).

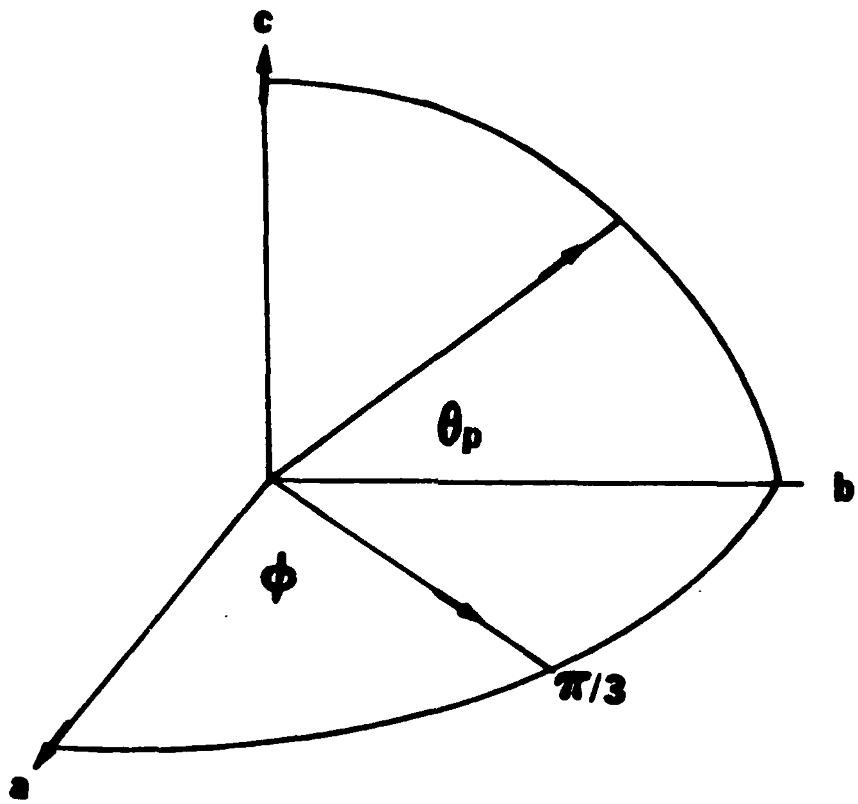

Figure IV-4. Pure longitudinal mode directions in trigonal crystals (indicated by arrow).

above, because of vanishing V_1 and W_1, only the terms involved in η''^3_{11} must be kept in the expression of the part of the strain energy density ϕ_3 in the double primed coordinate system, as well as only the strain derivative of elastic energy $\frac{\partial\phi_3}{\partial\eta''_{11}}$ must be used in the developing of the second approximation wave equation.

Therefore, we need to write the expression for ϕ_3 only with those terms involved in η''^3_{11}. That is:

$$\begin{aligned}
\phi_3 = {} & \frac{1}{6} C_{111}\cos^6\theta\cos^2\phi(\cos^2\phi - 3\sin^2\phi)^2\eta''^3_{11} \\
& + \frac{1}{2} C_{113}\cos^4\theta\sin^2\theta\eta''^3_{11} - C_{114}\cos^5\theta\sin\theta\eta''^3_{11} \\
& + 2C_{124}\sin\theta\cos^5\theta\sin\phi(1 + 2\cos(2\phi))\eta''^3_{11} \\
& + \frac{1}{2} C_{133}\cos^2\theta\sin^4\theta\eta''^3_{11} + 2C_{134}\cos^3\theta\sin^3\theta\sin\phi(4\cos^2\phi - 1)\eta''^3_{11} \\
& - C_{144}\cos^4\theta\sin^2\theta\cos^2\phi\sin^2\phi\eta''^3_{11} + C_{155}\cos^4\theta\sin^2\theta(2 - \frac{1}{4}\sin(2\phi))\eta''^3_{11} \\
& + \frac{1}{6} C_{222}\cos^6\theta\sin^2\phi(1 - 4\cos^2\phi)^2\eta''^3_{11} \\
& + \frac{1}{6} C_{333}\sin^6\theta\eta''^3_{11} + 2C_{344}\cos^2\theta\sin^4\theta\eta''^3_{11} \\
& + \frac{4}{3} C_{444}\cos^3\theta\sin^3\theta\sin\phi(1 - 4\cos^2\phi)\eta''^3_{11} \; . \qquad \text{(IV-15)}
\end{aligned}$$

And, its corresponding strain derivative is

$$\begin{aligned}
\frac{\partial\phi_3}{\partial\eta''_{11}} = {} & \frac{1}{2} C_{111}\cos^6\theta\cos^2\phi(\cos^2\phi - 3\sin^2\phi)^2\eta''^2_{11} \\
& + \frac{3}{2} C_{113}\cos^4\theta \sin^2\theta\eta''^2_{11} - 3C_{114}\cos^5\theta\sin\theta\eta''^2_{11} \\
& + \frac{3}{2} C_{124}\sin\theta\cos^5\theta\sin\phi(1 + 2\cos(2\phi))\eta''^2_{11}
\end{aligned}$$

$$+ \frac{3}{2} C_{133}\cos^2\theta\sin^4\theta\eta_{11}''^3 + 6C_{134}\cos^3\theta\sin^3\theta\sin\phi(4\cos^2\phi - 1)\eta_{11}''^2$$

$$- 3C_{144}\cos^4\theta\sin^2\theta\cos^2\phi\sin^2\phi\eta_{11}''^2$$

$$+ 3C_{155}\cos^4\theta\sin^2\theta(2 - \frac{1}{4}\sin(2\phi))\eta_{11}''^2$$

$$+ \frac{1}{2} C_{222}\cos^6\theta\sin^2\phi(1 - 4\cos^2\phi)^2\eta_{11}''^2$$

$$+ \frac{1}{2} C_{333}\sin^6\theta\eta_{11}''^2 + 6C_{344}\cos^2\theta\sin^4\theta\eta_{11}''^2$$

$$+ 4C_{444}\cos^3\theta\sin^3\theta\sin\phi(1 - 4\cos^2\phi)\eta_{11}''^2 \ . \qquad \text{(IV-16)}$$

Putting (IV-4), (IV-16), and (IV-14) into (II-41) and restricting our attention to the second approximation terms, we can get the second approximation wave equation of the pure longitudinal mode for all of the specific directions mentioned above.

The wave equation is

$$\rho_0\ddot{U}_2'' = \alpha \frac{\partial^2 U_2''}{\partial a''^2} + \delta \frac{\partial U_1''}{\partial a''} \frac{\partial^2 U_1''}{\partial a''^2} \qquad \text{(IV-17)}$$

or

$$\rho_0\ddot{U}_2'' = K_2 \frac{\partial^2 U_2''}{\partial a''^2} - \frac{1}{2} (3K_2 + K_3)k_L^3 A_1^2 \sin^2(\omega t - k_L a'') \ . \qquad \text{(IV-18)}$$

Here

$$K_2 = \alpha = C_{11}\cos^4\theta + \frac{1}{2} C_{13}\sin^2(2\theta) + 2C_{14}\sin(2\theta)\cos^2\theta\sin\phi$$

$$\cdot (3\cos^2\phi - \sin^2\phi) + C_{33}\sin^4\theta + C_{44}\sin^2(2\theta) \qquad \text{(IV-19)}$$

$$K_3 = \delta - 3K_2 = C_{111}\cos^6\theta\cos^2\phi(\cos^2\phi - 3\sin^2\phi)^2$$

$$+ 3C_{113}\cos^4\theta\sin^2\theta - 6C_{114}\cos^5\theta\sin\theta$$

$$+ 3C_{124}\sin\theta\cos^5\theta\sin\phi(1 + 2\cos(2\phi)) + 3C_{133}\cos^2\theta\sin^4\theta$$

$$+ 12C_{134}\cos^3\theta\sin^3\theta\sin\phi(4\cos^2\phi - 1) - 6C_{144}\cos^4\theta\sin^2\theta\cos^2\phi\sin^2\phi$$

$$+ 6C_{155}\cos^4\theta\sin^2\theta(2 - \frac{1}{4}\sin(2\phi)) + C_{222}\cos^6\theta\sin^2\phi(1 - 4\cos^2\phi)^2$$

$$+ C_{333}\sin^6\theta + 12C_{344}\cos^2\theta\sin^4\theta + 8C_{444}\cos^3\theta\sin^3\theta\sin\phi(1 - 4\cos^2\theta) . \quad \text{(IV-20)}$$

The solution of Eq. (IV-18) can be obtained with the boundary condition $U_2'' = 0$ where $a'' = 0$, as

$$U_2(a'',t) = -(3K_2 + K_3)\frac{(k_L A_1)^2}{8K_2}a''\cos 2(\omega t - k_L a'') . \quad \text{(IV-21)}$$

Thus, the solution of the nonlinear wave equation of the pure longitudinal mode can be expressed as

$$\begin{aligned} U'' &= U_1'' + U_2'' \\ &= A_1\sin(\omega t - k_L a'') - A_2\cos 2(\omega t - k_L a'') . \end{aligned} \quad \text{(IV-22)}$$

Here the amplitude of second harmonic is

$$A_2 = (3K_2 + K_3)\frac{(k_L A_1)^2}{8K_2}a'' . \quad \text{(IV-23)}$$

The parameters K_2 and K_3 for the specific directions of a pure longitudinal mode in trigonal crystals are written in tabular form in Table IV-1. The directions specified are the directions along which

Table IV-1. The K_2 and K_3 parameters for trigonal crystals along the direction of the pure longitudinal mode.

Direction of Wave Propagation θ	Direction of Wave Propagation ϕ	$K_2 = \alpha$	$K_3 = (\delta - 3\alpha)$
0	0	C_{11}	C_{111}
0	$\pi/3$	C_{11}	C_{111}
$\pi/2$		C_{33}	C_{333}
θ_p	$\pi/2$	$C_{11}\cos^4\theta_p + \frac{1}{2} C_{13}\sin^2(2\theta_p)$ $- 2C_{14}\sin(2\theta_p)\cos^2\theta_p$ $+ C_{33}\sin^4\theta_p + C_{44}\sin^2(2\theta_p)$	$3C_{113}\cos^4\theta_p\sin^2\theta_p - 6C_{114}\cos^5\theta_p\sin\theta_p$ $3C_{124}\sin\theta_p\cos^5\theta_p + 3C_{133}\cos^2\theta_p\sin^4\theta_p$ $- 12C_{134}\cos^3\theta_p\sin^3\theta_p + 12C_{155}\cos^4\theta_p\sin^2\theta_p$ $+ C_{222}\cos^6\theta_p + C_{333}\sin^6\theta_p + 12C_{344}\cos^2\theta_p\sin^4\theta_p$ $+ 8C_{444}\cos^3\theta_p\sin^3\theta_p$

harmonic generation experiments can be expected to yield useful information about third-order elastic constants in trigonal crystals.

V. CONCLUSION

The results given in this Technical Report can be used to evaluate the third-order elastic constants that determine the amplitude of the second harmonic wave propagating in the listed pure mode directions in crystals of hexagonal and trigonal symmetries.

In addition, we may point out that the theory developed in the early chapters of this report is sufficiently general that one can use it to study the nonlinear behavior of crystals of any symmetry. One even can consider piezoelectric crystals by using the appropriate form of the strain energy ϕ that describes the effect of piezoelectric coupling as well as nonlinear effects. As one goes to crystals of lower symmetry, however, he must be prepared for the equations to become more complicated.

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