Pramāṇa – J. Phys., Vol. 29, No. 4, October 1987, pp. 391–398. © Printed in India.

Elastic nonlinearity parameters of tetragonal crystals

JACOB PHILIP

Department of Physics, Cochin University of Science and Technology, Cochin 682 022, India

MS received 6 May 1987; revised 20 July 1987

Abstract. The equations of motion for the propagation of finite amplitude elastic waves in crystals of tetragonal symmetry have been derived starting from the expression for the elastic strain energy. The equations have been solved for a finite amplitude sinusoidal wave propagating along the pure mode directions which are [100], [110] and [001] for the tetragonal group TI. The solutions corresponding to longitudinal wave propagation yield expressions for the amplitudes of the fundamental and generated second harmonic for these directions in terms of certain combinations of second and third order elastic constants of the medium. The results will aid the experimenter to determine these constants using ultrasonic harmonic generation technique.

Keywords. Elastic nonlinearity; tetragonal symmetry; second harmonic generation; third order elastic constants; pure modes; longitudinal waves.

PACS No. 62·30

1. Introduction

Many important properties of a solid such as thermal expansion, temperature and pressure dependence of elastic constants, temperature dependence of lattice specific heat at high temperatures, ultrasonic attenuation due to phonon viscosity effects etc can be explained only if we treat the solid as a nonlinear elastic medium. This nonlinearity arises from the higher order terms in the energy density expansion of the solid. A finite amplitude elastic wave propagating through such a medium gets distorted as it progresses and higher harmonic waves are generated (Gedroits and Krasilnikov 1963; Breazeale and Thompson 1963). Measurement of the amplitudes of the generated higher harmonic wave offers a unique technique to determine higher order elastic coefficients (Breazeale and Ford 1965). The method has been used to determine combinations of third order elastic constants of a number of crystals of cubic symmetry even down to liquid helium temperatures (Breazeale and Philip 1984).

In an anisotropic medium there are only certain directions along which elastic waves can propagate as pure modes. The specific directions in which pure modes exist have been worked out before for all crystal point groups belonging to different classes (Borgnis 1955; Brugger 1965; Chang 1968). When the nonlinear terms in the strain energy expression are taken into account, it is found that pure transverse modes do not exist; they are always accompanied by longitudinal modes (Goldberg 1961). However, longitudinal waves continue to propagate as pure modes along these directions even under the nonlinear regime.

In this paper we have derived the equations of motion for longitudinal waves propagating along the pure mode directions in crystals of tetragonal symmetry starting from the strain energy expression including the third order terms in strain. Solution of these equations leads to second harmonic terms whose coefficients give amplitudes of the second harmonic of a finite amplitude elastic wave propagating along these directions. Only third order terms in strain have been retained because they do account for most of the nonlinear properties. Moreover, inclusion of fourth and higher order terms make the algebra unwieldy.

Tetragonal crystals have the point group symmetries 4, $\bar{4}$, 4/*m* and 422, 4*mm*, $\bar{4}2m$, 4/*mmm* belonging to TII and TI Laue groups respectively. It is known that the pure mode directions for this symmetry are [100], [010], [110], [1$\bar{1}$0] and [001] (Brugger 1965). The equations of motion for wave propagation along all these directions have been derived and solved for longitudinal case to determine the second harmonic amplitudes in terms of appropriate third order elastic constants. Since most of the experimentally interesting tetragonal crystals belong to the Laue group TI, we have considered only this group in this work. For this group the [100] and [010] directions and the [110] and [1$\bar{1}$0] directions are degenerate so that the only directions we need consider are [100], [001] and [110].

2. Equations of motion along the pure mode directions

In the Lagrangian formulation, elastic strain is described in the undeformed state and the initial co-ordinates of the material particle are taken as independent variables. The Lagrangian strain parameters which are components of the finite strain tensor are given by (Murnaghan 1951)

$$\eta = \tfrac{1}{2}(J^*J - \delta) \tag{1}$$

where J is the Jacobian with matrix elements

$$J_{kl} = \delta x_k / \partial a_l \tag{2}$$

and δ is the unit matrix. If a_i (a, b, c) are the co-ordinates of a point in the unstrained state and x_i (x, y, z) are the corresponding co-ordinates in the strained state, the components of particle displacement are

$$U_i = x_i - a_i \tag{3}$$

where $U_i = U, V, W$. In terms of the displacement derivatives J is given by

$$J = \begin{bmatrix} 1+U_a & U_b & U_c \\ V_a & 1+V_b & V_c \\ W_a & W_b & 1+W_c \end{bmatrix} \tag{4}$$

where $U_a = \partial U/\partial a$, $V_b = \partial V/\partial b$ etc.

If σ is the Cauchy stress tensor, the stress-strain relation can be written as (Murnaghan 1951)

$$\sigma = (\rho/\rho_0) J (\partial \phi / \partial \eta) J^* \tag{5}$$

where ρ_0 and ρ are the undeformed and deformed mass densities respectively and ϕ is the strain energy per unit undeformed volume. Symmetry of the strain components ensures that

$$\partial\phi/\partial\eta_{kl}=\partial\phi/\partial\eta_{lk}. \tag{6}$$

The properties of the crystalline medium enter the theory through the strain energy term ϕ. Deformable media with crystalline structure are, in general, elastically insensitive to rotations and elastic potential energy is a scalar under rotation so that

$$\phi(R^*\eta R)=\phi(\eta), \tag{7}$$

where R is the rotation matrix. In general, ϕ can be written as a sum of terms of different degrees in the elements of η as

$$\phi=\phi_0+\phi_1+\phi_2+\phi_3+\ \dots \tag{8}$$

in which case the relation

$$\phi_j(R^*\eta R)=\phi_j(\eta) \tag{9}$$

holds good with $j=0, 1, 2, 3$ . . . . . In terms of the elastic moduli, equation (8) can be written for a general crystal as (Green 1973)

$$\phi=\phi_0+K_1C_{ij}\eta_{ij}+K_2C_{ijkl}\eta_{ij}\eta_{kl}+K_3C_{ijklmn}\eta_{ij}\eta_{kl}\eta_{mn}+\ \dots \tag{10}$$

where K_n is a factor depending on the definition of elastic constants and $C_{ijkl\dots}$ are the elastic constants of proper order. In Voigt notation they take simpler forms. If the initial energy and deformation of the body are neglected, the first two terms in equation (10) vanish. This expression takes appropriate form for crystals of different symmetry.

In the case of tetragonal crystals belonging to Laue group TI there are 6 second order and 12 third order elastic constants (for group TII there are 7 second order and 16 third order constants). Using Brugger's thermodynamic definition of higher order elastic constants (Brugger 1964), the strain energy expression for tetragonal crystals with Laue group TI becomes (Einspruch and Manning 1964)

$$\begin{aligned}\phi=&\tfrac{1}{2}C_{11}(\eta_{11}^2+\eta_{22}^2)+C_{12}\eta_{12}\eta_{22}+\tfrac{1}{2}C_{33}\eta_{33}^2\\&+C_{13}\eta_{33}(\eta_{11}+\eta_{22})+2C_{44}(\eta_{13}^2+\eta_{23}^2)\\&+2C_{66}\eta_{12}^2+\tfrac{1}{6}C_{111}(\eta_{11}^3+\eta_{22}^3)+\tfrac{1}{6}C_{333}\eta_{33}^3\\&+\tfrac{1}{2}C_{112}\eta_{11}\eta_{22}(\eta_{11}+\eta_{22})+\tfrac{1}{2}C_{113}\eta_{33}(\eta_{11}^2+\eta_{22}^2)\\&+\tfrac{1}{2}C_{113}\eta_{33}^2(\eta_{11}+\eta_{22})+2C_{166}\eta_{12}^2(\eta_{11}+\eta_{22})\\&+2C_{366}\eta_{33}\eta_{12}^2+C_{123}\eta_{11}\eta_{22}\eta_{33}\\&+16C_{456}\eta_{12}\eta_{13}\eta_{23}+2C_{144}(\eta_{11}\eta_{23}^2+\eta_{22}\eta_{13}^2)\\&+2C_{344}\eta_{33}(\eta_{13}^2+\eta_{23}^2)+2C_{155}(\eta_{11}\eta_{13}^2+\eta_{22}\eta_{23}^2).\end{aligned} \tag{11}$$

The principle of conservation of energy along with the definition of J leads to the relation

$$|J| = \rho_0/\rho = (1 + U_a + V_b + W_c). \tag{12}$$

The equations of motion for an elastic medium are restatements of Newton's second law. If we introduce the stress tensor T, which is not symmetric, as

$$T = J\partial\phi/\partial\eta, \tag{13}$$

with nine components, we can write the equations of motion as

$$\partial T_{ij}/\partial a_j = \rho_0 \ddot{U}_i \tag{14}$$

in the Lagrangian frame. So we obtain the equations of motion along the a, b, c axes as

$$\rho\ddot{U} = \partial T_{11}/\partial a + \partial T_{12}/\partial b + \partial T_{13}/\partial c, \tag{15a}$$

$$\rho\ddot{V} = \partial T_{21}/\partial a + \partial T_{22}/\partial b + \partial T_{23}/\partial c, \tag{15b}$$

$$\rho\ddot{W} = \partial T_{31}/\partial a + \partial T_{32}/\partial b + \partial T_{33}/\partial c. \tag{15c}$$

The components of T can be evaluated from (13).

For plane waves propagating along the a-axis, the displacement components are

$$\begin{aligned} U &= U(a, t) \\ V &= V(a, t) \\ W &= W(a, t) \end{aligned} \tag{16}$$

so that the equations of motion (15) for this special case become

$$\begin{aligned} \rho_0\ddot{U} &= \partial T_{11}/\partial a \\ \rho_0\ddot{V} &= \partial T_{21}/\partial a \\ \rho_0\ddot{W} &= \partial T_{31}/\partial a. \end{aligned} \tag{17}$$

Similar expressions hold good for wave propagation along b and c axes.

3. Longitudinal wave propagation along [100] and [001] directions

The equations of motion for plane longitudinal waves propagating along [100], [010] and [001] directions of tetragonal crystals TI have been derived as outlined before. For longitudinal wave along [100] direction, the only nonvanishing displacement component is U. Similarly for [010] and [001] directions, the nonvanishing displacement components are V and W respectively. The lengthy algebra is avoided to save space. The equations of motion for all directions are found to be of the form

$$\rho_0\partial^2 U_i/\partial t^2 - \alpha\partial^2 U_i/\partial a_i^2 = \delta\partial U_i/\partial a_i \partial^2 U_i/\partial a_i^2 \tag{18}$$

where

$$\alpha = C_{11}$$
$$\delta = 3C_{11} + C_{111} \tag{19}$$

for [100] direction with $U_i = U$ and $a_i = a$. Both α and δ have the same values given by equation (19) for [010] direction with $U_i = V$ and $a_i = b$. For the [001] direction,

$$\alpha = C_{33}$$
$$\delta = 3C_{33} + C_{333} \tag{20}$$

with $U_i = W$ and $a_i = c$.

4. Longitudinal wave propagation along [110] direction

In order to derive the equations of motion for the propagation of plane longitudinal waves along the [110] pure mode direction, the strain energy expression given by equation (11) is to be derived with the frame of reference rotated about the [001] direction by $\pi/4$. This can be accomplished with the help of equation (7) with

$$R = \begin{bmatrix} 1/\sqrt{2} & 1/\sqrt{2} & 0 \\ -1/\sqrt{2} & 1/\sqrt{2} & 0 \\ 0 & 0 & 0 \end{bmatrix} \tag{21}$$

for the directions of interest. If primed quantities represent the rotated frame, the modified expression for the strain energy becomes

$$\begin{aligned}
\phi' = {} & \tfrac{1}{4}C_{11}[(\eta'_{11}+\eta'_{22})^2 + 4\eta'^2_{12}] \\
& + \tfrac{1}{2}C_{33}\eta'^2_{33} + \tfrac{1}{4}C_{12}[\eta'_{11}+\eta'_{22})^2 - (\eta'_{22}+\eta'_{12})^2] \\
& + C_{13}\eta'_{33}(\eta'_{11}+\eta'_{22}) + 2C_{44}(\eta'^2_{13}+\eta'^2_{23}) \\
& + \tfrac{1}{2}C_{66}[(\eta'_{11}-\eta'_{22})^2 + 2\eta'_{12}\eta'_{22}] \\
& + \tfrac{1}{4}C_{111}[(\eta'_{11}+\eta'_{22})^3 + 3\eta'^2_{12}(4\eta'_{11}+3\eta'_{22})] \\
& + C_{333}\eta'^3_{33} + \tfrac{1}{4}C_{112}(\eta'_{11}+\eta'_{22})[(\eta'_{11}+\eta'_{22})^2 - 4\eta'^2_{12}] \\
& + \tfrac{1}{2}C_{113}\eta'_{33}[(\eta'_{11}+\eta'_{22})^2 + 4\eta'^2_{12}] \\
& + C_{133}\eta'^2_{33}(\eta'_{11}+\eta'_{22}) + \tfrac{1}{4}C_{166}(\eta'_{11}+\eta'_{22})(\eta'_{11}-\eta'_{22})^2 \\
& + \tfrac{1}{4}C_{366}\eta'_{33}(\eta'_{11}-\eta'_{22})^2 + \tfrac{1}{4}C_{123}\eta_{33}[(\eta'_{11}+\eta'_{22})^2 - 4\eta'^2_{12}] \\
& + \tfrac{1}{2}C_{456}(\eta'_{11}-\eta'_{22})(\eta'^2_{13}-\eta'^2_{23}) + C_{344}\eta'_{33}(\eta'^2_{13}+\eta'^2_{23}) \\
& + \tfrac{1}{2}C_{144}[(\eta'_{11}+\eta'_{22})(\eta'^2_{13}+\eta'^2_{23}) - 4\eta'_{12}\eta'_{13}\eta'_{23}] \\
& + \tfrac{1}{2}C_{155}[(\eta'_{11}+\eta'_{22})(\eta'^2_{13}+\eta'^2_{23}) + 4\eta'_{12}\eta'_{13}\eta'_{23}].
\end{aligned} \tag{22}$$

The corresponding equations of motion have been derived as before. It is found that they have the same form as given by equation (18). For the [110] direction with $U_i = U$ and $a_i = a$,

$$\alpha = \tfrac{1}{2}(C_{11} + C_{12} + 2C_{66})$$
$$\delta = \tfrac{3}{2}(C_{11} + C_{12} + 2C_{66} + \tfrac{1}{6}C_{111} + \tfrac{1}{2}C_{112} + 2C_{166}). \tag{23}$$

About the axis of rotation i.e. the [001] direction in the original frame, the equation and coefficients remain unaltered, given by (20), as expected.

5. Solution of the equations of motion

It is found that for all the directions of interest, the equations of motion for longitudinal wave propagation have the form given by equation (18) which can be rewritten as

$$\rho_0 \ddot{U} - \alpha U_{aa} = \delta U_a U_{aa}. \tag{24}$$

In order to solve this equation, a perturbation approach is usually adopted. The standard trial solution is

$$U = U_1 + U_2 \tag{25}$$

where U_1 and U_2 are approximate solutions of first and second order. Substituting this in (24) and assuming that $U_2 \ll U_1$, one arrives at the solutions after applying proper boundary conditions. The solution is found to be of the form

$$U(a, t) = A_1 \sin(ka - \omega t) - [\delta(kA_1)^2/8\rho_0 C_0^2] a \cos 2(ka - \omega t). \tag{26}$$

In this equation C_0 is the phase velocity of the wave given by $C_0 = (\alpha/\rho)^{1/2}$ and k is the propagation constant given by $k = \omega/C_0$.

In equation (26) we find that the first term on the right hand side is the solution to the linear wave equation so that it represents the fundamental wave with amplitude A_1. The second term contains the 2ω factor and represents the second harmonic component that is generated due to the nonlinearity that has been built into the formulation. It follows that the amplitude of the generated second harmonic is given by

$$A_2 = A_1^2 k^2 a \delta / 8\rho_0 C_0^2. \tag{27}$$

It is customary to write

$$\alpha = K_2$$

and $$\delta = 3K_2 + K_3, \tag{28}$$

where K_2 and K_3 are combinations of second and third order elastic constants tabulated in table 1 for different directions. One can define the nonlinearity parameter as

$$\beta = -\delta/\alpha = -(3K_2 + K_3)/K_2. \tag{29}$$

Table 1. Parameters α and δ for longitudinal wave propagation along pure mode directions in tetragonal crystals.

Direction of wave propagation	$\alpha = K_2$	$\delta = 3K_2 + K_3$
[100]	C_{11}	$3C_{11} + C_{111}$
[001]	C_{33}	$3C_{33} + C_{333}$
[110]	$\frac{1}{2}(C_{11} + C_{12} + 2C_{66})$	$\frac{3}{2}(C_{11} + C_{12} + 2C_{66} + \frac{1}{6}C_{111} + \frac{1}{2}C_{112} + 2C_{166})$

In terms of the amplitudes of the fundamental and second harmonic, the nonlinearity parameter is given by

$$\beta = 8(A_2/A_1^2)(1/k^2 a). \tag{30}$$

6. Conclusion

Table 1 predicts the relative magnitudes of the amplitudes of the second harmonic wave generated when longitudinal waves propagate along the pure mode directions in tetragonal crystals. As expected, the K_2 and K_3 parameters for [100] and [010] directions are the same for the group we have considered. So is the case with [110] and [1$\bar{1}$0] directions. The derived expressions are the ones necessary for an experimenter who intends to determine the combinations of third order elastic constants listed as K_3 parameters in table 1 by ultrasonic harmonic generation technique. What one needs to do is to launch longitudinal ultrasonic waves of known frequency in the specific direction and measure the amplitudes of the fundamental and generated second harmonic after they have travelled through a known distance in the sample and use equation (30) and table 1.

Acknowledgements

The author thanks Dr R Pratap for many fruitful discussions. This work has been supported by the Department of Science and Technology in the form of a Young Scientists research project to the author.

References

Borgnis F E 1955 *Phys. Rev.* **98** 1000

Breazeale M A and Ford 1965 *J. Appl. Phys.* **36** 3486

Breazeale M A and Philip J 1984 in Physical acoustics (eds) W P Mason and R N Thurston (New York: Academic Press) **17** 1

Breazeale M A and Thompson D O 1963 *Appl. Phys. Lett.* **3** 77
Brugger K 1964 *Phys. Rev.* **A133** 1611
Brugger K 1965 *J. Appl. Phys.* **36** 759
Chang Z P 1968 *J. Appl. Phys.* **39** 5669
Einspruch N G and Manning R J 1964 *J. Appl. Phys.* **35** 560
Gedroits A A and Krasilnikov V A 1963 *Sov. Phys. JETP* **16** 1122
Goldberg Z A 1961 *Sov. Phys. Acoust.* **6** 306
Green Jr R E 1973 in Treatise on materials science and technology (ed) H Herman (New York: Academic Press) **3** 73
Murnaghan F D 1951 Finite deformation of an elastic solid (New York: John Wiley)