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ACOUSTICAL AND OPTICAL ACTIVITY IN ALPHA QUARTZ

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Abstract—It is shown that a theory of elastic dielectrics, in which the stored electromechanical energy depends on the polarization gradient, accounts for both acoustical and optical activity. Formulas for the acoustical and optical rotatory powers of α -quartz are derived and the values of new material constants appearing in them are calculated from experimental data.

1. INTRODUCTION

ACOUSTICAL activity (rotation of the direction of mechanical displacement along the path of a transverse, elastic wave) has recently been observed by Pine [1] and by Joffrin and Levelut [2] in α-quartz. The possibility of the phenomenon appears first to have been mentioned by Silin [3]. It was accounted for by one of us [4] on the basis of the theory of elasticity in which the stored energy is a function of the gradient of the strain, in addition to the strain. Portigal and Burstein [5] found an equivalent result by assigning dependence of the elastic stiffness on the wave vector. The purpose of the present paper is to show that both acoustical and optical activity are accounted for in the theory of elastic dielectrics in which the stored electromechanical energy is a function of the polarization gradient [6] in addition to the usual strain and polarization. It has already been shown [7] that the differential equations of the resulting theory, rather than the equations of the classical theory of piezoelectricity, are the correct, long wave, low frequency limit of the finite difference equations of a lattice of shell model atoms if the shell-shell interaction between adjacent atoms is taken into account. It has also been shown that the polarization gradient can account [6] for the surface energy of deformation and polarization and it can also account [7] for an anomaly observed in measurements of electrical capacitance of thin, dielectric films. In the present paper, the field equations are exhibited for the coupled elastic-electric-magnetic system, the problem of shear waves along the trigonal axis of α -quartz is solved, formulas are obtained for the optical and acoustical rotatory powers and numerical values of the new material constants, in the formulas, are calculated from experimental data. Essentially, the theory has it that the appearance of optical activity depends on an interaction between the polarization and the polarization gradient; the appearance of acoustical activity depends on interactions of the strain with both the polarization and the polarization gradient, and is absent if either interaction is missing

—provided, of course, that the phenomenon does not depend on the strain gradient, as assumed, tentatively, in this paper.

2. COUPLED ELASTIC, ELECTRIC AND MAGNETIC FIELDS

The linear equations of an elastic, dielectric continuum, with the contribution of the polarization gradient taken into account, but without the coupling to the magnetic field, may be written as [6]

$$T_{ij,i} = \rho \ddot{u}_j,$$

$$E_{ij,i} + E_j^L + E_j = 0,$$

$$\varepsilon_{ijk} E_{k,j} = 0,$$

$$\varepsilon_0 E_{i,i} + P_{i,i} = 0.$$
(1)

where

$$T_{ij} = \partial W^L/\partial S_{ij}, \quad E_j^L = -\partial W^L/\partial P_j, \quad E_{ij} = \partial W^L/\partial P_{i,i}$$
 (2)

and

$$W^{L} = \frac{1}{2}a_{ij}P_{i}P_{j} + \frac{1}{2}b_{ijkl}P_{j,i}P_{l,k} + \frac{1}{2}c_{ijkl}^{P}S_{ij}S_{kl} + d_{ijkl}P_{j,i}S_{kl} + f_{ijk}P_{i}S_{jk} + f_{ijk}P_{i}P_{k,j},$$
(3)

$$S_{ij} = \frac{1}{2}(u_{i,i} + u_{i,j}). \tag{4}$$

In (1), u_i is the mechanical displacement, P_i is the polarization density, E_i is the Maxwell electric self-field, ρ is the mass density, ε_0 is the permittivity of a vacuum and ε_{ijk} is the alternating tensor. In (3), W^L is the stored energy density of deformation and polarization in which b_{ijkl} , d_{ijkl} and j_{ijk} are constants associated with the polarization gradient, $P_{j,i}$, while a_{ij} , f_{ijk} and c^P_{ijkl} belong to the classical theory of piezoelectricity and are related to the I.R.E. standard [8] symbols for the reciprocal dielectric susceptibility, χ_{ij} , the piezoelectric stress constant, e_{ijk} , and the elastic stiffness at constant electric field, c^E_{ijkl} , according to [9]

$$a_{ij} = \varepsilon_0^{-1} \chi_{ij}, \qquad f_{ijk} = -\varepsilon_0^{-1} \chi_{il} e_{ljk}, \qquad c_{ijkl}^P = c_{ijkl}^E + \varepsilon_0^{-1} \chi_{mn} e_{mij} e_{nkl}. \tag{5}$$

To couple (1) to the equations of the magnetic field, it is only necessary to replace the third of (1) by

$$\varepsilon_{ijk}E_{k,j} + \dot{B}_i = 0 \tag{6}$$

and add the equations

$$\mu_0^{-1} \varepsilon_{ijk} B_{k,j} - \varepsilon_0 \dot{E}_i - \dot{P}_i = 0, \tag{7}$$

$$B_{i,i} = 0, (8)$$

where B_i is the magnetic flux density and μ_0 is the magnetic permeability, assumed to be that of a vacuum.

It is convenient to eliminate B_i by subtracting the curl of (6) from the time derivative of (7), with the result

$$E_{j,ii} - E_{i,ij} = \varepsilon_0 \mu_0 \ddot{E}_j + \mu_0 \ddot{P}_j. \tag{9}$$

The last of (1) and (8) are not independent of (7) and (6), respectively, and may be disregarded for the present purpose.

Thus, (9), along with the first two of (1):

$$T_{ij,i} = \rho \ddot{u}_j,$$

$$E_{ij,i} + E_j^L + E_j = 0,$$

$$E_{j,ii} - E_{i,ji} = \varepsilon_0 \mu_0 \ddot{E}_j + \mu_0 \ddot{P}_j,$$
 (10)

are the field equations governing mechanical and electromagnetic waves, coupled through the constitutive equations:

$$T_{ij} = c_{ijkl}^{P} S_{kl} + f_{kij} P_k + d_{klij} P_{l,k},$$

$$-E_{j}^{L} = f_{jkl} S_{kl} + a_{jk} P_k + j_{jkl} P_{l,k},$$

$$E_{ij} = d_{ijkl} S_{kl} + j_{kij} P_k + b_{ijkl} P_{l,k},$$
(11)

which are obtained from (2) and (3).

For a crystal of class 32 (international) or D_3 (Schoenflies) [10], to which α -quartz belongs, the constitutive equations take the form shown in Fig. 1, in which x_3 has been taken as the trigonal axis and x_1 one of the digonal axes. In Fig. 1, an abridged notation is used for the subscripts attached to the material constants:

$$11 \to 1$$
, $22 \to 2$, $33 \to 3$, $23 \to 4$, $32 \to 7$, $31 \to 5$, $13 \to 8$, $12 \to 6$, $21 \to 9$.

3. PLANE WAVES ALONG THE TRIGONAL AXIS

We consider plane, transverse waves propagating along the x_3 -axis; i.e. u_i , P_i and E_i are functions of x_3 and t only, but u_3 , P_3 and E_3 are zero. Then the field equations (10) reduce to

$$T_{13,3} = \rho \ddot{u}_{1}, \qquad T_{23,3} = \rho \ddot{u}_{2},$$

$$E_{31,3} + E_{1}^{L} + E_{1} = 0, \qquad E_{32,3} + E_{2}^{L} + E_{2} = 0,$$

$$E_{1,33} = \varepsilon_{0} \mu_{0} \ddot{E}_{1} + \mu_{0} \ddot{P}_{1}, \qquad E_{2,33} = \varepsilon_{0} \mu_{0} \ddot{E}_{2} + \mu_{0} \ddot{P}_{2}$$

$$(12)$$

and the constitutive equations (11) reduce to

$$T_{32} = c_{44}^{P} u_{2,3} + f_{14} P_1 + d_{74} P_{2,3},$$

$$T_{31} = c_{44}^{P} u_{1,3} - f_{14} P_2 + d_{74} P_{1,3},$$

$$-E_1^{L} = f_{14} u_{2,3} + \varepsilon_0^{-1} \chi_{11} P_1 + j_{17} P_{2,3},$$

$$-E_2^{L} = -f_{14} u_{1,3} + \varepsilon_0^{-1} \chi_{11} P_2 - j_{17} P_{1,3},$$

$$E_{32} = d_{74} u_{2,3} + j_{17} P_1 + b_{55} P_{2,3},$$

$$E_{31} = d_{74} u_{1,3} - j_{17} P_2 + b_{55} P_{1,3}.$$
(13)

 $\begin{array}{c} S_{22} \\ C_{12} \\ C_{13} \\ C_{14} \\ C_{1$

 $\begin{array}{c|c} S_{11} \\ C_{12} \\ C_{13} \\ C_{14} \\ C_$

Inserting (13) in (12), we have

$$c_{44}^{P}u_{1,33} - f_{14}P_{2,3} + d_{74}P_{1,33} = \rho\ddot{u}_{1},$$

$$c_{44}^{P}u_{2,33} + f_{14}P_{1,3} + d_{74}P_{2,33} = \rho\ddot{u}_{2},$$

$$d_{74}u_{1,33} - 2j_{17}P_{2,3} + b_{55}P_{1,33} - f_{14}u_{2,3} - \varepsilon_{0}^{-1}\chi_{11}P_{1} + E_{1} = 0,$$

$$d_{74}u_{2,33} + 2j_{17}P_{1,3} + b_{55}P_{2,33} + f_{14}u_{1,3} - \varepsilon_{0}^{-1}\chi_{11}P_{2} + E_{2} = 0,$$

$$E_{1,33} = \varepsilon_{0}\mu_{0}\ddot{E}_{1} + \mu_{0}\ddot{P}_{1},$$

$$E_{2,33} = \varepsilon_{0}\mu_{0}\ddot{E}_{2} + \mu_{0}\ddot{P}_{2}.$$
(14)

Now, take

$$u_{1} = A_{1} \sin \zeta(x_{3} - vt), \qquad u_{2} = A_{2} \cos \zeta(x_{3} - vt),$$

$$P_{1} = B_{1} \sin \zeta(x_{3} - vt), \qquad P_{2} = B_{2} \cos \zeta(x_{3} - vt),$$

$$E_{1} = C_{1} \sin \zeta(x_{3} - vt), \qquad E_{2} = C_{2} \cos \zeta(x_{3} - vt)$$
(15)

and substitute these functions in (14) to find

$$(c_{44}^{P} - \rho v^{2})\zeta A_{1} - f_{14}B_{2} + d_{74}\zeta B_{1} = 0,$$

$$(c_{44}^{P} - \rho v^{2})\zeta A_{2} - f_{14}B_{1} + d_{74}\zeta B_{2} = 0,$$

$$d_{74}\zeta^{2}A_{1} - f_{14}\zeta A_{2} + (b_{55}\zeta^{2} + \varepsilon_{0}^{-1}\chi_{11})B_{1} - 2j_{17}\zeta B_{2} - C_{1} = 0,$$

$$d_{74}\zeta^{2}A_{2} - f_{14}\zeta A_{1} + (b_{55}\zeta^{2} + \varepsilon_{0}^{-1}\chi_{11})B_{2} - 2j_{17}\zeta B_{1} - C_{2} = 0,$$

$$\mu_{0}v^{2}B_{1} + (\varepsilon_{0}\mu_{0}v^{2} - 1)C_{1} = 0,$$

$$\mu_{0}v^{2}B_{2} + (\varepsilon_{0}\mu_{0}v^{2} - 1)C_{2} = 0.$$
(16)

Adding and subtracting these equations in pairs, we have

$$(c_{44}^{P} - \rho v^{2})(A_{1} \pm A_{2}) + (d_{74}\zeta \mp f_{14})(B_{1} \pm B_{2}) = 0,$$

$$(d_{74}\zeta \mp f_{14})(A_{1} \pm A_{2}) + (b_{55}\zeta^{2} + \varepsilon_{0}^{-1}\chi_{11} \mp 2j_{17}\zeta)(B_{1} \pm B_{2}) - (C_{1} \pm C_{2}) = 0,$$

$$\mu_{0}v^{2}(B_{1} \pm B_{2}) + (\varepsilon_{0}\mu_{0}v^{2} - 1)(C_{1} \pm C_{2}) = 0.$$
(17)

Thus, there are two solutions, each corresponding to circularly polarized waves [11, p. 222] since the amplitudes must satisfy

$$A_1 = \pm A_2, \qquad B_1 = \pm B_2, \qquad C_1 = \pm C_2$$
 (18)

with either all upper signs or all lower signs. Upon substituting (18) into (15), we see that the upper and lower signs give right and left circular polarization, respectively. The velocities are obtained by setting the determinant of the coefficients of the amplitudes in (17) equal to zero:

$$\begin{vmatrix} c_{44}^{P} - \rho v^{2} & d_{74}\zeta \mp f_{14} & 0 \\ d_{74}\zeta \mp f_{14} & b_{55}\zeta^{2} + \varepsilon_{0}^{-1}\chi_{11} \mp 2j_{17}\zeta & 1 \\ 0 & 1 & \mu_{0}^{-1}v^{-2} - \varepsilon_{0} \end{vmatrix} = 0.$$
 (19)

This is a quadratic equation in v^2 , so that there are two pairs of oppositely circularly polarized waves. Each pair of such waves combines to produce a linearly polarized wave with a rotating direction of polarization [11, p. 222]. Thus, we have two cases of rotary polarization. These may be identified as optical and acoustical by separating out first the electromagnetic part of (19) and then the electromechanical part. The two may, in fact, be considered separately owing to the large ratio of the frequencies (of the order of 10^5) at which the two effects are observed.

4. OPTICAL ACTIVITY

The electromagnetic part of the determinant in (19) is the minor of the upper left element. Thus, the pair of optical velocities is given by

$$\begin{vmatrix} b_{55}\zeta^2 + \varepsilon_0^{-1}\chi_{11} + 2j_{17} & 1\\ 1 & \mu^{-1}v^{-2} - \varepsilon_0 \end{vmatrix} = 0,$$
 (20)

which yields the dispersion formula (cf. [11, p. 426])

$$n_{\pm}^{2} - 1 = (\chi_{11} \mp 2\varepsilon_{0} j_{17} \zeta + \varepsilon_{0} b_{55} \zeta^{2})^{-1}, \tag{21}$$

where n_{+} are the indices of refraction:

$$n_+ = c/v_+ \tag{22}$$

and c is the velocity of light in vacuo:

$$c = (\varepsilon_0 \mu_0)^{-1}. \tag{23}$$

From (21), we have

$$(n_{-}^{2}-1)^{-1}-(n_{+}^{2}-1)^{-1}=4\varepsilon_{0}j_{17}\zeta.$$
(24)

Now, define $n = \frac{1}{2}(n_+ + n_-)$ and assume

$$|n_{+} - n_{-}| \ll n_{+} + n_{-}. \tag{25}$$

Then (24) becomes, approximately,

$$n_{+} - n_{-} = 2(n^{2} - 1)^{2} \varepsilon_{0} j_{17} \zeta_{0}, \qquad (26)$$

where $\zeta_0(=\zeta/n)$ is the wave number in vacuo.

The optical rotatory power, in radians per unit length, is given by [11, p. 222]

$$\theta_{OP} = \frac{1}{2} \zeta_0 (n_- - n_+). \tag{27}$$

Accordingly, from (26) and (27),

$$\theta_{OP} = -(n^2 - 1)^2 \varepsilon_0 j_{17} \zeta_0^2 \tag{28}$$

is the formula for the optical rotatory power in terms of the average index of refraction, n, along the optic axis, the wave length in vacuo, $\lambda_0 (= 2\pi/\zeta_0)$, the fundamental constant ε_0 and the material constant $j_{17} (= j_{132})$ which, as may be seen in (3), measures the interaction between the polarization and the polarization gradient.

5. ACOUSTICAL ACTIVITY

The electromechanical part of the determinant in (19) is the minor of the lower right element, so that we have

$$\begin{vmatrix} c_{44}^P - \rho v^2 & d_{74}\zeta \mp f_{14} \\ d_{74}\zeta \mp f_{14} & b_{55}\zeta^2 + \varepsilon_0^{-1}\chi_{11} \mp 2j_{17}\zeta \end{vmatrix} = 0$$
 (29)

for the equation determining the velocities of the two acoustical waves, as influenced by the quasi-static polarization and polarization gradient. From (29),

$$\rho v_{\pm}^{2} = c_{44}^{P} - (d_{74}\zeta \mp f_{14})^{2} / (b_{55}\zeta^{2} + \varepsilon_{0}^{-1}\chi_{11} \mp 2j_{17}\zeta). \tag{30}$$

In view of (18) and the inequality of v_+ and v_- , the superposition of the two waves results in rotary polarization (acoustical activity) with acoustical rotatory power

$$\theta_{AC} = \frac{1}{2}\omega(v_{-}^{-1} - v_{+}^{-1}),\tag{31}$$

where ω is the circular frequency.

Both waves are dispersive. At the zero frequency (long wave) limit, from (30) and (5),

$$\lim_{\xi \to 0} \rho v_{\pm}^2 = c_{44}^P - \varepsilon_0 f_{14}^2 / \chi_{11} = c_{44}^E, \tag{32}$$

which is the result (without acoustical activity, since $v_+ = v_-$) that would be obtained if the contribution of the polarization gradient were omitted, i.e. if d_{74} , b_{55} and j_{17} were assumed to be zero. As the frequency increases from zero, the absolute velocity difference, $|v_+ - v_-|$, at first increases, so that the acoustical activity appears and increases. With further increase of frequency, the velocity difference again approaches zero, since

$$\lim_{\zeta \to \infty} \rho v_{\pm}^2 = c_{44}^P - d_{74}^2 / b_{55}, \tag{33}$$

so that the acoustical activity diminishes; but this is undoubtedly beyond the range of applicability of the continuum theory. Up to moderately large wave numbers, (30) and (5) give, to the first order in ζ ,

$$v_0/v_{\pm} = 1 \mp (d_{74} - j_{17}e_{14})e_{14}\zeta/c_{44}^E \tag{34}$$

where $v_0^2 = c_{44}^E/\rho$. In this range, the frequency is approximately proportional to the wave number: $\omega = v_0 \zeta$; so that, from (31) and (34),

$$\theta_{AC} = (d_{74} - j_{17}e_{14})e_{14}\rho\omega^2/(c_{44}^E)^2. \tag{35}$$

Thus, at frequencies up to, say, 10^{10} cps, the acoustical rotatory power is approximately proportional to the square of the frequency and depends on the constants ρ , e_{14} and e_{44}^E , which are commonly encountered in piezoelectricity theory, and also on the constants e_{74}^E and e_{17}^E which control the coupling of the polarization gradient with the strain and polarization, respectively.

6. APPLICATION TO OUARTZ

For α -quartz, all the quantities in the formula (28) for optical rotatory power are known except j_{17} . Thus, for left-handed quartz and the sodium D line,

$$\theta_{OP} = -379 \text{ rad./m}$$
 [11, p. 481],
 $\lambda_0 = 5893 \times 10^{-10} \text{ m}$ [11, p. 481],
 $n = 1.5533$ [11, p. 481],
 $\epsilon_0 = 8.854 \times 10^{-12} \text{ F/m}$ [10].

Hence,

$$j_{17} = -\theta_{OP}\lambda_0^2/4\pi^2\varepsilon_0(n^2 - 1)^2 = 0.19 \text{ m}^2/\text{F}.$$
 (36)

With the value of j_{17} known, all quantities in the formula (35) for acoustical rotatory power are known except d_{74} . In particular, Pine [1] finds that the acoustical and optical activities have opposite signs and the acoustical rotary power along the trigonal axis is about 220 rad./m at one gigahertz. Thus, for left handed α -quartz,

$$\theta_{AC} = 220 \text{ rad./m}$$
 [1],
 $\omega = 2\pi \times 10^9 \text{ rad./sec}$ [1],
 $\rho = 2.65 \times 10^3 \text{ kg/m}^3$ [12],
 $c_{44}^E = 57.94 \times 10^9 \text{ Newton/m}^2$ [13],
 $e_{14} = -0.0406 \text{ C/m}^2$ [13].

Hence,

$$d_{74} = \theta_{AC}(c_{44}^E)^2 / e_{14} \rho \omega^2 + j_{17} e_{14} = -174 - 0.0077 \text{ V}.$$
 (37)

The second term, $j_{17}e_{14}$, is negligible in comparison with the first, so that we may drop the dependence of acoustical rotatory power on j_{17} and replace (35) with

$$\theta_{AC} = d_{74}e_{14}\rho\omega^2/(c_{44}^E)^2. \tag{38}$$

Thus, according to this theory, the presence of acoustical activity in α -quartz depends on the existence of the piezoelectric stress constant $e_{14}(=e_{123})$ and the interaction constant $d_{74}(=d_{3223})$ between strain and polarization gradient, whereas the presence of optical activity depends only on the existence of the interaction constant $j_{17}(=j_{132})$ between polarization and polarization gradient.

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Абстракт—Указывается, что теория упругих диэлектриков, в которых сохраняеная электромеханическая энергия зависит от градиента поляризации, отвечает так за активность акустическую, как и оптическую. Определяются формулы для акустической мощности и оптической мощности вращения, для кварца а. Подсчитываются из экспериментальных данных значения новых постоянных материала, сущесмвующих в этих выажениях.