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ELECTROMECHANICAL VIBRATIONS OF CENTROSYMMETRIC CUBIC CRYSTAL PLATES

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R. D. MINDLIN

(Columbia University, New York, U. S. A.)

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Introduction. According to the classical theory of piezoelectricity, there can be no piezoelectric effect in centrosymmetric crystals. Consequently the theory has it that vibrations of a centrosymmetric crystal plate cannot be excited, for example, by applying an alternating voltage drop between electrodes on the opposing faces of the plate. This conclusion is a direct result of the assumption, in the theory, that the stored energy of deformation and polarization is a function of the strain and polarization only [1]. Hence the only possible electromechanical interaction energy is the product of a second rank tensor (strain) and a first rank tensor (polarization) — with a third rank material coefficient (a piezoelectric constant). Since there are no third rank centrosymmetric tensors, there is no piezoelectric effect in centrosymmetric materials.

There is reason to believe, however, that the stored energy of deformation and

polarization should depend not only on the strain and polarization, but also on the polarization gradient. The augmented theory [2] based on such an assumption can find justification on the grounds that: (1) it accommodates the mathematical representation of a surface energy of deformation and polarization [2] which is absent from the classical theory but which has been measured in the laboratory and calculated from atomic considerations; (2) it can account [3] for an apparent anomaly observed in measurements of the electrical capacitance of thin, dielectric films; (3) the resulting equations, rather than the classical ones, are the correct, low frequency limit [3, 5] of the modern dynamical theory of crystal lattices of electronically polarizable atoms.

If the polarization gradient is admitted as a variable in the stored energy function, an additional interaction energy, between the two second rank tensors of strain and polarization gradient, is possible. Since the accompanying material coefficient is fourth rank, it represents a possible electromechanical effect in centrosymmetric materials.

In the following paragraphs, it is shown that, at least in principle, it is possible to excite thickness vibrations in a centrosymmetric cubic crystal plate by applying an alternating voltage drop between electrodes on a pair of (100) faces of the plate. Approximate mode shapes and frequencies are calculated for the case of sodium chloride; for which it is found, also, that the electromechanical coupling coefficient can be as large as one four-hundredth of that of an *X*-cut quartz plate.

1. Field equations and boundary conditions. The linear equations of an elastic dielectric continuum, including the contribution of the polarization gradient to the stored energy, have been derived elsewhere [2] by means of a simple extension of Toupin's [1] variational principle for the classical equations of piezoelectricity. For the present case, we consider a plate of cubic crystal class $m\bar{3}m (O_h)$ [4] bounded by a pair of (100) faces, at $x = \pm h$ on which are deposited electrode films to which are applied voltages $\pm V e^{i\omega t}$. If end effects are neglected, the mechanical and electrical fields are one-dimensional and are governed by the equations [2]

$$\begin{aligned} c_{11}\partial^2 u + d_{11}\partial^2 P &= -\rho\omega^2 u, & d_{11}\partial^2 u + b_{11}\partial^2 P - a_{11}P - \partial\varphi &= 0 \\ \epsilon_0\partial^2\varphi - \partial P &= 0 \end{aligned} \quad (1)$$

where ($\partial = d/dx$), u and P are the x -components of the mechanical displacement and electronic polarization, respectively, and φ is the potential of the Maxwell, electric self-field. Also, ρ is the mass density, c_{11} is an elastic stiffness, ϵ_0 is the permittivity of a vacuum and $\epsilon_0 a_{11}$ is the reciprocal dielectric susceptibility. The remaining two constants, b_{11} and d_{11} , are associated with terms, in the energy density, involving polarization gradient: $1/2 b_{11}$ is the coefficient of a quadratic term and d_{11} is the coefficient of a product of polarization gradient and strain; $\partial P \partial u$. Thus, d_{11} is the additional electromechanical constant without which, as may be seen from (1), the mechanical and electrical fields would not be coupled.

As for the boundary conditions, we suppose that the mass and stiffness of the electrodes are negligible, in comparison with those of the plate, so that the tractions across $x = \pm h$ are zero.

This condition, as shown previously [2], is

$$(c_{11}\partial u + d_{11}\partial P)_{x=\pm h} = 0 \tag{2a}$$

Also, as shown previously [3], we may set

$$(P)_{x=\pm h} = -kV/a_{11}h \quad (0 \leq k \leq 1) \tag{2b}$$

with zero the most likely value of k ; corresponding to continuity of polarization across the crystal-electrode interfaces. Finally, the applied voltage is taken as

$$(\varphi)_{x=\pm h} = \pm V \tag{2c}$$

It may be noted that independent specification of boundary values of polarization and potential is permissible in the augmented theory, but not in the classical theory.

2. Solution. In view of the boundary conditions (2), a solution of (1), with appropriate symmetry and form, is

$$u = A \cos \xi x, \quad P = B_0 + B \cos \xi x, \quad \varphi = C_0 x + C \sin \xi x \tag{3}$$

Upon substituting (3) in (1), we find

$$B_0 = -a_{11}^{-1}C_0, \quad B = \epsilon_0 \xi C \tag{4}$$

$$\frac{A}{C} = -\frac{\epsilon_0 d_{11} \xi^3}{c_{11} \xi^2 - \rho \omega^2} = -\frac{1 + \epsilon_0 a_{11} + \epsilon_0 b_{11} \xi^2}{d_{11} \xi} \tag{5}$$

The second of (5) is a quadratic in ξ^2 :

$$(b_{11}c_{11} - d_{11}^2)\epsilon_0 \xi^4 - [\epsilon_0 b_{11} \rho \omega^2 - (1 + \epsilon_0 a_{11})c_{11}] \xi^2 - (1 + \epsilon_0 a_{11}) \rho \omega^2 = 0 \tag{6}$$

Now, positive-definiteness of the energy density requires

$$b_{11}c_{11} - d_{11}^2 > 0, \quad a_{11} > 0 \tag{7}$$

Hence, one of the roots ξ^2 , of (6), is positive real and the other is negative real; so that one ξ is real and the other is imaginary. Accordingly, from (4) to (7), we may rewrite the solution (3) as

$$\begin{aligned} u &= \alpha_1 C_1 \cos \xi_1 x + \alpha_2 C_2 \operatorname{ch} \xi_2 x \\ P &= -a_{11}^{-1}C_0 + \epsilon_0 \xi_1 C_1 \cos \xi_1 x + \epsilon_0 \xi_2 C_2 \operatorname{ch} \xi_2 x \\ \varphi &= C_0 x + C_1 \sin \xi_1 x + C_2 \operatorname{sh} \xi_2 x \end{aligned} \tag{8}$$

where

$$\alpha_i = -\frac{\epsilon_0 d_{11} \xi_i^3}{c_{11} \xi_i^2 + (-1)^i \rho \omega^2} = \frac{1 + \epsilon_0 a_{11} - (-1)^i \epsilon_0 b_{11} \xi_i^2}{(-1)^i d_{11} \xi_i} \tag{9}$$

Upon substituting (8) in the boundary conditions (2), we find

$$\begin{aligned} (c_{11}\alpha_1 + \epsilon_0 d_{11}\xi_1)\xi_1 C_1 \sin \xi_1 h - (c_{11}\alpha_2 + \epsilon_0 d_{11}\xi_2)\xi_2 C_2 \operatorname{sh} \xi_2 h &= 0 \\ -C_0 + \epsilon_0 a_{11}\xi_1 C_1 \cos \xi_1 h + \epsilon_0 a_{11}\xi_2 C_2 \operatorname{ch} \xi_2 h &= kV/h \\ C_0 h + C_1 \sin \xi_1 h + C_2 \operatorname{sh} \xi_2 h &= V \end{aligned} \tag{10}$$

It may be seen, from (10), that the applied voltage V forces both the mechanical and electrical fields.

Resonance occurs when the determinant of the coefficients of C_0 , C_1 and C_2 in (10), vanishes; or, when

$$\frac{1 + \epsilon_0 a_{11} \xi_1 h \operatorname{ctg} \xi_1 h}{1 + \epsilon_0 a_{11} \xi_2 h \operatorname{cth} \xi_2 h} = \frac{1 + \rho \omega^2 / c_{11} \xi_2^2}{1 - \rho \omega^2 / c_{11} \xi_1^2} \tag{11}$$

in which (9) has been employed.

3. Application of sodium chloride. For sodium chloride, the following are values of the material constants appearing in the solution:

$$\begin{aligned} \epsilon_0^{-1} a_{11}^{-1} &= 4.6 [1, \text{pp. 68, 69}], \\ b_{11} &= 6.88 \times 10^3 \text{ dyn-cm}^4 / \text{coul}^2 [5], \\ c_{11} &= 4.83 \times 10^{11} \text{ dyn/cm}^2 [6, \text{p. 39}], \\ d_{11} &= 4.67 \times 10^7 \text{ dyn-cm/coul} [5], \\ \epsilon_0^{-1} &= 36\pi \times 10^9 \text{ dyn-cm}^2 / \text{coul}^2 [4, \text{p. 68}], \\ \rho &= 2.214 \text{ gm/cm}^3 [6, \text{pp. 39, 88}]. \end{aligned}$$

With these numerical values of the constants, simple approximate expressions for the resonance frequencies and mode shapes can be found.

At low frequencies, we find, from (6),

$$\lim_{\omega \rightarrow 0} \xi_3^{-1} = \left[\frac{b_{11} - d_{11}^2 / c_{11}}{\epsilon_0^{-1} (1 + \epsilon_0 a_{11})} \right]^{1/2} \approx 1.3 \times 10^{-8} \text{ cm.} \quad (12)$$

Considering plate thicknesses of the order of millimeters, (12) indicates an extremely rapid decay, away from the surfaces of the plate, of the exponential parts of the fields, leaving

$$\begin{aligned} u &\approx \alpha_1 C_1 \cos \xi_1 x, & P &\approx -a_{11}^{-1} C_0 + \epsilon_0 \xi_1 C_1 \cos \xi_1 x \\ \varphi &\approx C_0 x + C_1 \sin \xi_1 x \end{aligned} \quad (13)$$

over most of the thickness of the plate. In fact, the decay modulus (12) is less than one-half the nearest neighbor distance, $a = 2.83 \cdot 10^{-8}$ cm,

of the sodium chloride lattice. Also, with (12), the roots of (11) are very nearly roots of

$$\sin \xi_1 h = 0, \quad \xi_1 h \neq 0, \quad (14)$$

or

$$\xi_1 h = n\pi, \quad n = 1, 2, 3, \dots \quad (15)$$

Finally, rewriting (6) in the form

$$\left(1 - \frac{\rho \omega^2}{c_{11} \xi^2} \right) \frac{1 + \epsilon_0 a_{11} + \epsilon_0 b_{11} \xi^2}{\xi^3} - \frac{\epsilon_0 d_{11}^2}{c_{11}} = 0 \quad (16)$$

and noting that

$$\frac{\epsilon_0 d_{11}^2}{c_{11}} \approx 4 \cdot 10^{-17} \text{ cm}^2 \quad (17)$$

we see that the frequencies given by (16) are approximately

$$\omega = \xi_1 (c_{11} / \rho)^{1/2} \quad (18)$$

Hence, from (15),

$$\omega = \frac{n\pi}{h} \left(\frac{c_{11}}{\rho} \right)^{1/2} \quad (19)$$

or

$$f = \frac{\omega}{2\pi} \approx \frac{4.7 n}{2h} \times 10^6 \text{ cyc/sec} , \quad (20)$$

where $2h$ is the thickness of the plate in millimeters. Thus, for a plate one millimeter thick, the lowest resonance ($n = 1$) has a frequency of about 4.7 megacycles per second. It may be noted that this mode has two nodes; the central half of the thickness of the plate and the outer quarters move in opposite directions. This is the mode most likely

to be detected; but the response is very sharp in comparison with that of a piezoelectric crystal such as quartz, as may be seen from a calculation of an electromechanical coupling coefficient.

An electromechanical coupling coefficient may be defined as the square root of the ratio of the strain energy to the total energy – both at zero frequency.

At $\omega = 0$, the solution (8) reduces to [3]

$$u_0 = B_1 \operatorname{ch} \xi_2 x, \quad P_0 = A_2 + B_2 \operatorname{ch} \xi_2 x, \quad \varphi_0 = A_3 x + B_3 \operatorname{sh} \xi_2 x \quad (21)$$

where ξ_2 is given by (12) and

$$\begin{aligned} A_2 &= A_3 / a_{11}, & B_2 &= \varepsilon_0 \xi_2 B_3 = -c_{11} B_1 / d_{11}, & A_3 &= B_3 \varepsilon_0 a_{11} \xi_2 \operatorname{ch} \xi_2 h \\ B_3 &= (1 - k) V (\operatorname{sh} \xi_2 h + \varepsilon_0 a_{11} \xi_2 h \operatorname{ch} \xi_2 h)^{-1} \end{aligned} \quad (22)$$

The strain energy per unit area is

$$W_s = \frac{c_{11}}{2} \int_{-h}^h (\partial u_0)^2 dx = \frac{\varepsilon_0^2 \xi_2^2 d_{11}^2 (1 - k)^2 V^2 (\operatorname{sh} 2\xi_2 h - 2\xi_2 h)}{4c_{11} (\operatorname{sh} \xi_2 h + \varepsilon_0 a_{11} \xi_2 h \operatorname{ch} \xi_2 h)^2} \quad (23)$$

The total energy per unit area is V times the surface charge:

$$W = V (\varepsilon_0 \partial \varphi_0 - P_0)_{x=h} = \frac{\varepsilon_0 \xi_2 (1 + \varepsilon_0 a_{11}) (1 - k) V^2 \operatorname{ch} \xi_2 h}{\operatorname{sh} \xi_2 h + \varepsilon_0 a_{11} \xi_2 h \operatorname{ch} \xi_2 h} + \frac{k V^2 (1 + \varepsilon_0 a_{11})}{h a_{11}} \quad (24)$$

Taking into account the fact that $\xi_2 h$ is very large, the electromechanical coupling coefficient, for the case $k = 0$, is

$$\left(\frac{W_s}{W} \right)^{1/2} \approx \left(\frac{\xi_2 d_{11}^2}{2\varepsilon_0 a_{11}^2 c_{11} (1 + \varepsilon_0^{-1} a_{11}^{-1}) h} \right)^{1/2} \approx 2.4 \cdot 10^{-4} \quad (25)$$

(for $h = 1 \text{ mm}$)

This is to be compared with 9.5×10^{-2} for an X -cut quartz plate [7]. Thus, for $k = 0$, the coefficient for the sodium chloride plate is one four-hundredth that for the quartz plate, resulting in a resonance band width to be much narrower [7]. It is interesting to note that the coupling coefficient (25) is inversely proportional to the square root of the thickness of the plate, whereas the coefficient in the piezoelectric case is independent of the thickness.

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