

## Electrostrictive Effect in Barium Titanate Ceramics

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When a direct current bias is applied to a multicrystalline barium titanate ceramic, an alternating voltage can excite resonances in the ceramic. Four modes of motion have been excited—a longitudinal mode at right angles to the applied field, a radial mode of a circular plate at right angles to the applied field, a thickness longitudinal mode, and a thickness shear mode. The first three are excited when the a.c. field is applied in the same direction as the d.c. polarization, but the fourth is excited when the a.c. field is at right angles to the d.c. polarization. The amount of motion is larger than in magnetostrictive materials, and it appears that barium titanate may be an important electromechanical transducing element.

All of these modes can be accounted for on the basis of a second-order electrostrictive effect. Two electrostrictive constants are involved and these have been evaluated as

$$Q_{12} = -2.15 \times 10^{-12} (\text{cm}^4 / (\text{statcoulombs})^2);$$

$$Q_{11} = +6.9 \times 10^{-12} (\text{cm}^4 / (\text{statcoulombs})^2).$$

Using these constants, the measured electromechanical coupling factors for the four modes are evaluated, and these compare well with the calculated values.

A theoretical explanation of this effect is given which depends on the fact that when a given domain becomes ferro-electric it loses its cubic structure and becomes tetragonal. In this process it expands one percent along the tetragonal axis and contracts one-half percent along the other two axes. In the ceramic piece all directions for the tetragonal axis are equally probable, but an applied field can cause the domains in the direction of the field to grow at the expense of domains perpendicular to the field. This growth is accompanied by an increase in the thickness of the crystal and a decrease in radial dimensions. The measured ratio of 3 to 1, compared to the 2 to 1 ratio observed by x-rays for a single crystal, is accounted for by the nature of the ceramic material which does not join up for all grains. This does not prevent the ceramic from increasing in thickness but does cut down the radial contraction.

Experimental measurements of the electrostrictive effect are given, and it is shown that the displacement is proportional to the square or products of the electric displacements in the ceramic.

### I. INTRODUCTION

WHEN an electric field or electric displacement is applied or generated in a solid dielectric, a change in shape occurs which is proportional to the square of the voltage or electrical displacement. Electrostrictive effects are usually very feeble compared to first-order piezoelectric effects that occur in many crystalline materials. However, in the case of the ferro-electric materials rochelle salt and barium titanate, electrostrictive effects<sup>1,2</sup> may be quite

<sup>1</sup> In the present paper, a strain that is proportional to the square or product of two fields or electric displacements is called an electrostrictive strain. This is contrary in some cases to a usage started by Mueller, who calls the square term a "quadratic piezoelectric effect" when it depends on a strain caused by a spontaneous polarization or an applied field acting on a piezoelectric constant. On this definition the strain in rochelle salt that is proportional to the square of the electric displacement would be a "quadratic piezoelectric effect" because it depends on the orthorhombic crystal becoming monoclinic in the ferro-electric region and generating new piezoelectric constants which give a strain proportional to the spontaneous polarization times the applied electric displacement. The electrostrictive effect in barium titanate is not of this type (see reference 2) and is in every way the analog of a magnetostrictive effect in a ferromagnetic material.

<sup>2</sup> W. P. Mason, *Phys. Rev.* **73**, 1398 (1948).

large and are of considerable interest. The thickness expansion is about as large as can be obtained by the direct piezoelectric effect in rochelle salt and is somewhat larger than can be obtained with magnetostrictive materials. Furthermore, the variations of the properties of barium titanate with temperature are not nearly as large as for rochelle salt. Hence such materials may be of use for various types of transducers. If the alternating variations are small compared to the d.c. polarization, a remanent polarization is sufficient to keep the device operative. However, for the largest displacements or voltages, the remanent polarization may become discharged and it is necessary to put on a steady d.c. biasing voltage to make the device operative.

### II. METHODS FOR MEASURING THE FUNDAMENTAL CONSTANTS

When a constant voltage bias is applied to a multicrystalline barium titanate ceramic, an alternating voltage can excite resonances in the ceramic. There are four effects that have been measured. These are a radial vibration of a disk

of the material,<sup>3</sup> a length vibration of a bar cut from such a disk, a thickness vibration in the direction of the applied field,<sup>4</sup> and a thickness shear mode.<sup>5</sup> The first three motions are excited when the d.c. field is applied in the same direction as the a.c. field, while the fourth is excited when the d.c. polarization is at right angles to the a.c. field. Since, if there are two sets of plates at right angles to each other, the a.c. field cannot be made uniform through its direction of application, this mode has to be excited by exciting a remanent polarization by the d.c. field and then taking off the plating in this direction. In fact for all of these modes the d.c. biasing voltage can be applied and then taken off and the device will still operate by means of the remanent polarization.

A typical method for measuring such resonances is shown by Fig. 1. Here a source of high voltage, such as a high voltage transformer and rectifying tube, with the output connected through high resistances, is put directly on the ceramic piece, while the a.c. voltage is applied through two 4-microfarad condensers in series. At the resonant frequencies of the ceramic, which are usually above 100 kilocycles, the impedances of the condensers are less than 1 ohm while the shunt impedance of the high voltage source being 10 megohms is much higher than the impedance of the ceramic. Hence by this method one obtains a measure of the electrical impedance of the ceramic and can determine the effect of putting a high electrical bias on it.

If one measures the impedance of a freshly made ceramic on which no electrical bias has been placed, the impedance is that of a condenser and no resonances can be excited. However, if one puts a bias of 30,000 volts per centimeter on the ceramic disk of the material, for example, having the dimensions

$$\text{radius } a = 2.5 \text{ cm; thickness } t = 0.025 \text{ cm,} \quad (1)$$

Fig. 2 shows a measurement of the resonant and antiresonant frequencies as a function of the applied voltage, as the voltage bias is decreased to  $-30,000$  volts per cm. Upon reversing the

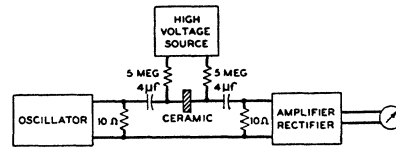


FIG. 1. Measuring circuit for studying resonances and electromechanical coupling factors in barium titanate ceramics.

direction of the bias the ascending curves shown are obtained. It is obvious that we are dealing with a hysteretic material for which the previous history determines the response. Since the electric displacement follows a similar hysteresis curve when plotted against the voltage, it is obvious that the response is determined by the electric displacement rather than the electric field. When the field is reduced to zero a polarization remains and this determines the resonant and antiresonant frequencies of the material.

From the data of Fig. 2 and the measured dielectric constant shown by Fig. 3, one can calculate the electromechanical coupling (which determines the percentage of energy stored in mechanical form to the total input electrical energy), the electrostrictive constant, and the value of the elastic constant controlling the radial vibrations. The method for deriving the fundamental elastic, electrostrictive, and electromechanical coupling constants for radial vibrations is discussed in the appendix. It is shown that the resonant frequency for a material having a Poisson's ratio 0.27, which is near that for

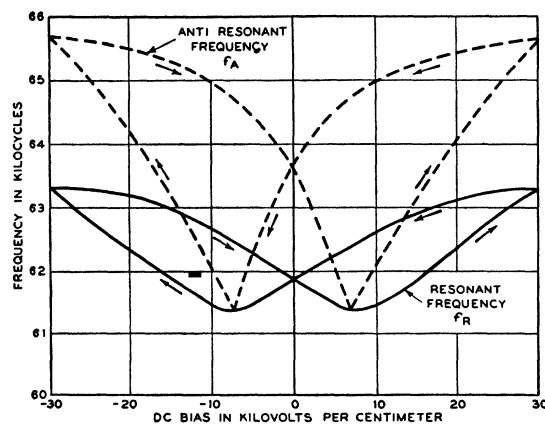


FIG. 2. Resonant and antiresonant frequencies as a function of the biasing voltage for a disk 5 centimeters in diameter.

<sup>3</sup> Shepard Roberts, Phys. Rev. **71**, 890-895 (1947).

<sup>4</sup> W. P. Mason, Phys. Rev. **72**, 869 (1947).

<sup>5</sup> W. L. Cherry, Jr. and Robert Adler, Phys. Rev. **72**, 981 (1947).

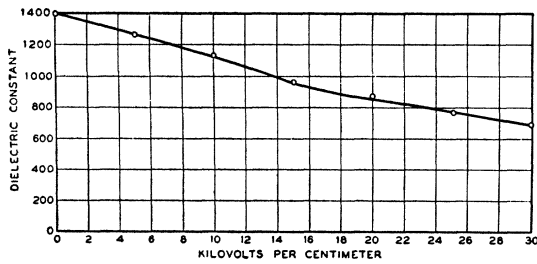


FIG. 3. Dielectric constant at room temperature as a function of the biasing voltage.

barium titanate, is given by the equation

$$f_R = \frac{2.03}{2\pi a} \left( \frac{Y_0}{\rho(1-\sigma^2)} \right)^{\frac{1}{2}}, \quad (2)$$

where  $a$  is the radius,  $Y_0$  = Young's modulus,  $\rho$  = density, and  $\sigma$  = Poisson's ratio. In the disk whose data are given by Fig. 2,  $a = 2.5$  cm,  $\rho = 5.5$ , and  $\sigma = 0.27$ . Hence the value of Young's modulus for zero biasing field is  $1.12 \times 10^{12}$  dyne/cm<sup>2</sup>. The value of Young's modulus is increased slightly with bias, being  $1.18 \times 10^{12}$  at 30,000 volts per centimeter.

It is shown in the appendix that the electromechanical coupling factor  $k$  is determined in terms of the separation of resonance and anti-resonance frequency,  $\Delta f$ , the resonant frequency  $f_R$ , the first root  $R$ , of the frequency determining equation, i.e.,  $R_1 = 2.03$ , and the value of Poisson's ratio by the equation

$$k^2 = \frac{\Delta f}{f_R} \left[ \frac{R_1^2 - (1 - \sigma^2)}{1 + \sigma} + \dots \right]. \quad (3)$$

For  $\sigma = 0.27$ , the value of the factor multiplying  $\Delta f/f_R$  is equal to 2.51. Hence from the data of Fig. 2, the electromechanical coupling factor can be calculated and is shown plotted by Fig. 4. It follows a regular hysteresis loop, indicating that the remanent polarization is annulled when the voltage gradient is about 7500 volts per cm negative. For a smaller initial polarizing field, the coercive field is less.

These results indicate that for an applied d.c. voltage, since the strain is proportional to the square of the electric displacement, if we plot it against the field, the characteristic butterfly loop of a hysteretic material will result as shown by Fig. 5. Actual d.c. measurements with a

bimorph unit with the field applied to only one side give values which follow this type of curve very well. The calculated constant comes very close to that measured by a.c. measurements as discussed in the next section.

Similar a.c. measurements have been made for the thickness longitudinal mode, the thickness shear mode, and the longitudinal length mode, and the coupling factors are shown plotted by Fig. 4. The frequency constant for the longitudinal thickness mode for a zero bias is 2550 kilocycle-millimeters. From this one obtains the elastic constant from the formula

$$f = (1/2t)[(\lambda + 2\mu)/\rho]^{\frac{1}{2}} \quad (4)$$

as

$$(\lambda + 2\mu) = 1.42 \times 10^{12} \text{ dynes/cm}^2. \quad (5)$$

This and the value of Young's modulus

$$Y_0 = 1.13 \times 10^{12} \text{ dynes/cm}^2 \\ = \mu(3\lambda + 2\mu)/(\lambda + \mu) \quad (6)$$

allow one to solve for the two Lamé' elastic constants. These are

$$\lambda = 5.2 \times 10^{11} \text{ dynes/cm}^2; \\ \mu = 4.5 \times 10^{11} \text{ dynes/cm}^2, \quad (7)$$

and from these the values of Poisson's ratio is

$$\sigma = \lambda/2(\lambda + \mu) = 0.27 \quad (8)$$

as quoted above.

To obtain the thickness shear mode, one has to polarize the ceramic in one direction and then remove the plating. An a.c. field perpendicular to this will generate a thickness shear mode with a coupling shown by the single point of Fig. 4.

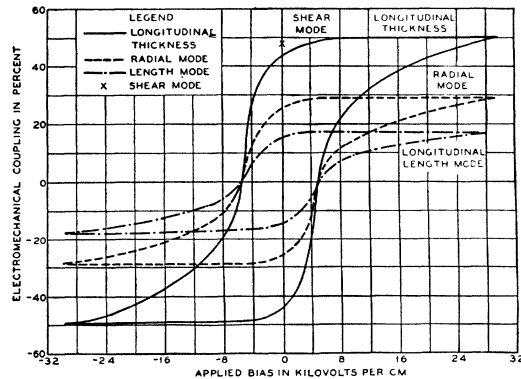


FIG. 4. Electromechanical coupling factors for four modes of motion.

### III. PHENOMENOLOGICAL THEORY OF ELECTROSTRICTIVE EFFECT IN BARIUM TITANATE CERAMICS

Since in a barium titanate ceramic any crystal symmetry is lost by the distribution of crystal axes in all directions, any first-order piezoelectric effects are annulled and all the modes of motion must be due to second-order electrostrictive effects. In the experiments of Section II it was shown that the electromechanical coupling was determined by the electric displacement rather than the electric field, so that we take as the independent variable the stresses and the electric displacements. All the measurements were made under adiabatic conditions so that all constants can be considered as adiabatic.

In terms of a tensor notation the internal energy residing in the body can be expressed in the form

$$dU = T_{ij}dS_{ij} + E_m \frac{dD_m}{4\pi} + TdS, \quad \begin{matrix} i, j = 1, 2, 3 \\ m = 1, 2, 3 \end{matrix} \quad (9)$$

where  $T_{ij}$  are the six stress components,  $S_{ij}$  the six strain components,  $E_m$  the fields, and  $D_m$  the electric displacements,  $T$  the absolute temperature, and  $S$  the entropy. The strain components are defined in the usual tensor form

$$S_{ij} = \frac{1}{2} \left( \frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} \right), \quad (10)$$

where  $u_i$  are the displacements along the  $x_i$  axes. In order to avoid using the factor  $1/4\pi$ , we made the substitution

$$\delta_m = D_m/4\pi. \quad (11)$$

$\delta_m$  is then measured in statcoulombs per square centimeter.

For the present purpose, since we are going to take  $T_{ij}$ ,  $\delta_m$ , and  $S$  as the fundamental variables, we introduce a potential  $H_1$ , called the elastic enthalpy, defined by the equation

$$H_1 = U - S_{ij}T_{ij}. \quad (12)$$

Hence

$$dH_1 = -S_{ij}dT_{ij} + E_m d\delta_m + TdS, \quad (13)$$

and

$$\begin{aligned} S_{ij} &= -\partial H_1 / \partial T_{ij}; & E_m &= \partial H_1 / \partial \delta_m; \\ T &= \partial H_1 / \partial S. \end{aligned} \quad (14)$$

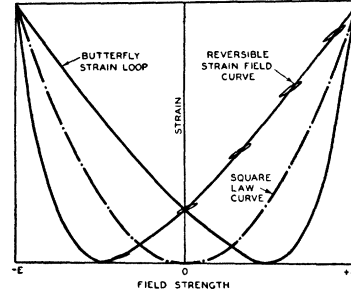


FIG. 5. Strain field relations.

Since for adiabatic conditions  $S$  does not vary, the dependent variables of interest,  $S_{ij}$  and  $E_m$ , can be written in the form

$$S_{ij}(T_{kl}, \delta_n); \quad E_m(T_{kl}, \delta_n). \quad (15)$$

Expanding these functions about the position of zero strain and zero electric field, we have up to second-order terms

$$\begin{aligned} S_{ij} &= \frac{\partial S_{ij}}{\partial T_{kl}} dT_{kl} + \frac{\partial S_{ij}}{\partial \delta_n} d\delta_n + \frac{1}{2!} \left[ \frac{\partial^2 S_{ij}}{\partial T_{kl} \partial T_{qr}} dT_{kl} dT_{qr} \right. \\ &\quad \left. + 2 \frac{\partial^2 S_{ij}}{\partial T_{kl} \partial \delta_n} dT_{kl} d\delta_n + \frac{\partial^2 S_{ij}}{\partial \delta_n \partial \delta_o} d\delta_n d\delta_o \right] + \dots \\ E_m &= \frac{\partial E_m}{\partial T_{kl}} dT_{kl} + \frac{\partial E_m}{\partial \delta_n} d\delta_n + \frac{1}{2!} \left[ \frac{\partial^2 E_m}{\partial T_{kl} \partial T_{qr}} dT_{kl} dT_{qr} \right. \\ &\quad \left. + 2 \frac{\partial^2 E_m}{\partial T_{kl} \partial \delta_n} dT_{kl} d\delta_n + \frac{\partial^2 E_m}{\partial \delta_n \partial \delta_o} d\delta_n d\delta_o \right] + \dots \end{aligned} \quad (16)$$

For the present purpose some of these partial derivatives can be set equal to zero. Since there is no direct piezoelectric effect on account of the uniform distribution of the crystals in all directions

$$\frac{\partial S_{ij}}{\partial \delta_n} = -\frac{\partial^2 H_1}{\partial \delta_n \partial T_{ij}} = -\frac{\partial^2 H_1}{\partial T_{ij} \partial \delta_n} = -\frac{\partial E_m}{\partial T_{ij}} = 0. \quad (17)$$

Furthermore the ceramic can be described as soft electrically but not mechanically. Hence not much change in the elastic constants with stress will occur and  $\partial^2 S_{ij} / \partial T_{kl} \partial T_{qr} = 0$ . There is a slight change of elastic constants with electric displacement as shown by Fig. 2, but it is small

TABLE I. Terms not ruled out by symmetry in the isotropic case.

	$\delta_1^2$	$\delta_1\delta_2$	$\delta_2\delta_1$	$\delta_1\delta_3$	$\delta_2^2$	$\delta_3\delta_1$	$\delta_2\delta_3$	$\delta_3\delta_2$	$\delta_3^2$
$S_{11}$	$Q_{1111}$	0	0	0	$Q_{1122}$	0	0	0	$Q_{1122}$
$S_{12}$	0	$Q_{1111}-Q_{1122}$	$Q_{1111}-Q_{1122}$	0	0	0	0	0	0
$S_{21}$	0	$Q_{1111}-Q_{1122}$	$Q_{1111}-Q_{1122}$	0	0	0	0	0	0
$S_{13}$	0	0	0	$Q_{1111}-Q_{1122}$	0	$Q_{1111}-Q_{1122}$	0	0	0
$S_{22}$	$Q_{1122}$	0	0	0	$Q_{1111}$	0	0	0	$Q_{1122}$
$S_{31}$	0	0	0	$Q_{1111}-Q_{1122}$	0	$Q_{1111}-Q_{1122}$	0	0	0
$S_{23}$	0	0	0	0	0	0	$Q_{1111}-Q_{1122}$	$Q_{1111}-Q_{1122}$	0
$S_{32}$	0	0	0	0	0	0	$Q_{1111}-Q_{1122}$	$Q_{1111}-Q_{1122}$	0
$S_{33}$	$Q_{1122}$	0	0	0	$Q_{1122}$	0	0	0	$Q_{1111}$

and is neglected here. Hence we can set

$$\frac{\partial^2 S_{ij}}{\partial T_{kl} \partial \delta_n} = - \frac{\partial^3 H_1}{\partial T_{kl} \partial \delta_n \partial T_{ij}} = - \frac{\partial^3 H_1}{\partial T_{ij} \partial T_{kl} \partial \delta_n} = - \frac{\partial E_m}{\partial T_{ij} \partial T_{kl}} = 0. \quad (18)$$

This leaves only three second-order partial derivatives, two of which are related, and these we designate as

$$\frac{\partial^2 S_{ij}}{\partial \delta_n \partial \delta_o} = - \frac{\partial^3 H_1}{\partial \delta_n \partial \delta_o \partial T_{ij}} = - \frac{\partial^3 H_1}{\partial T_{ij} \partial \delta_n \partial \delta_o} = - \frac{\partial^2 E_m}{\partial T_{ij} \partial \delta_n} = 2Q_{ij\ no}; \quad (19)$$

$$\frac{\partial^2 E_m}{\partial \delta_n \partial \delta_o} = O_{mno}.$$

The two remaining first-order derivatives of Eq. (16) determine the elastic compliances and dielectric impermeabilities according to the equations

$$\frac{\partial S_{ij}}{\partial T_{kl}} = s^D_{ijkl}; \quad \frac{\partial E_m}{\partial \delta_n} = \beta^T_{mn}, \quad (20)$$

where  $s^D_{ijkl}$  are the elastic compliance constants measured at constant electric displacement and  $\beta^T_{mn}$  are the dielectric "impermeability" constants (inverse of dielectric constants) measured

at constant stress. For the most general case there are 21 components of  $s^D_{ijkl}$  and 6 of the impermeability constants. For the isotropic case considered here, symmetry conditions insure that there are only two elastic compliances and one dielectric impermeability. For the most general case there are 36 components of the electrostrictive tensor  $Q_{ij\ no}$  and 27 for the correction  $O_{mno}$  to the dielectric constant. For the isotropic condition, the off diagonal term of the type

$$Q_{1122} = \frac{\partial^2 S_{11}}{\partial \delta_2^2} \quad \text{and} \quad Q_{2211} = \frac{\partial^2 S_{22}}{\partial \delta_1^2} \quad (21)$$

are obviously equal since the expansion along  $x_1$  for an electric field along  $x_2$  is equal to an expansion along  $x_2$  for a field along  $x_1$ . Hence the tensor is symmetrical and has the same number of components as the fourth rank elastic compliance tensor  $s^D_{ijkl}$ . For the isotropic case symmetry rules out all terms except those shown by Table I. The terms on the left are the strains generated by the products of the electric displacements shown by the top column. Since  $S_{ij} = S_{ji}$  and  $\delta_i \delta_j = \delta_j \delta_i$ , three columns and three rows are redundant. The fourth rank tensor for the elastic compliances will have the same terms with  $Q_{1111}$  replaced by  $s_{1111}$  and  $Q_{1122}$  replaced by  $s_{1122}$ . For the top variable line  $\delta_1^2$  is replaced by  $T_{11}$ ,  $\delta_1 \delta_2$  by  $T_{12}$ , etc.

To simplify the method of writing these equations, the usual one-index matrix symbols are used for the stresses and strains and the usual two-index compliance, electrostrictive, and impermeability constants are used; the electro-

strictive equations become

$$\begin{aligned}
 S_1 &= s_{11}^D T_1 + s_{12}^D (T_2 + T_3) + Q_{11} \delta_1^2 + Q_{12} [\delta_2^2 + \delta_3^2] \\
 S_2 &= s_{12}^D [T_1 + T_3] + s_{11}^D T_2 + Q_{11} \delta_2^2 + Q_{12} [\delta_1^2 + \delta_3^2] \\
 S_3 &= s_{12}^D [T_1 + T_2] + s_{11}^D T_3 + Q_{11} \delta_3^2 + Q_{12} [\delta_1^2 + \delta_2^2] \\
 S_{12} &= \frac{S_6}{2} = (s_{11}^D - s_{12}^D) T_6 + (Q_{11} - Q_{12}) \delta_1 \delta_2 \\
 S_{13} &= \frac{S_5}{2} = (s_{11}^D - s_{12}^D) T_5 + (Q_{11} - Q_{12}) \delta_1 \delta_3 \\
 S_{23} &= \frac{S_4}{2} = (s_{11}^D - s_{12}^D) T_4 + (Q_{11} - Q_{12}) \delta_2 \delta_3 \\
 E_1 &= \delta_1 [4\pi\beta_{11}^T + O_{11}\delta_1] - 2[Q_{11}[\delta_1 T_1 + \delta_2 T_6 + \delta_3 T_5] + Q_{12}[\delta_1(T_2 + T_3) - (T_6\delta_2 + T_5\delta_3)]] \\
 E_2 &= \delta_2 [4\pi\beta_{11}^T + O_{11}\delta_2] - 2[Q_{11}[\delta_2 T_2 + \delta_1 T_6 + \delta_3 T_4] + Q_{12}[\delta_2(T_1 + T_3) - (\delta_1 T_6 + \delta_3 T_4)]] \\
 E_3 &= \delta_3 [4\pi\beta_{11}^T + O_{11}\delta_3] - 2[Q_{11}[\delta_3 T_3 + \delta_1 T_5 + \delta_2 T_4] + Q_{12}[\delta_3(T_1 + T_2) - (\delta_1 T_5 + \delta_2 T_4)]]
 \end{aligned} \tag{22}$$

In this equation an extra term  $O_{11}$  has been added to represent the decrease in dielectric constant with applied field, which as shown by Fig. 3 is considerable.<sup>6</sup> Over a temperature range, the complex dielectric constant varies<sup>7</sup> as shown by Fig. 6. The equations for the various modes can be derived from Eqs. (22).

The simplest mode to consider is a longitudinal mode for a long thin bar generated by a field perpendicular to the length. If we take the thickness as lying along  $z$  while the length is along  $x$ , the equations reduce to

$$\begin{aligned}
 S_1 &= s_{11}^D T_1 + Q_{12} \delta_3^2; \\
 E_3 &= \delta_3 [4\pi\beta_{11}^T + O_{11}\delta_3] - 2Q_{12} \delta_3 T_1.
 \end{aligned} \tag{23}$$

For the case of interest here  $\delta_3$  consists of a part  $\delta_{30}$  due to an applied field or a remanent polarization plus an alternating component due to an applied a.c. voltage. As far as the alternating components go we can write these two equations as

$$\begin{aligned}
 S_1 &= s_{11}^D T_1 + Q_{12} [2\delta_{30}\delta_3]; \\
 E_3 &= \delta_3 [4\pi\beta_{11}^T + O_{11}\delta_{30}] - 2Q_{12} \delta_{30} T_1.
 \end{aligned} \tag{24}$$

To reduce this equation to the standard form<sup>8</sup>

<sup>6</sup> This measurement was made by Gordon Danielson of the Bell Telephone Laboratories.

<sup>7</sup> A. Von Hippel, R. G. Breckinridge, F. G. Chesley, and Lazlo Tisza, *Ind. Eng. Chem.* **38**, 1097 (1946).

<sup>8</sup> See, for example, W. P. Mason, *Phys. Rev.* **70**, 705 (1946).

used in solving piezoelectric crystals, we have to express the stress  $T_1$  in terms of the strain  $S_1$  and field  $E_3$ . By eliminating  $\delta_3$  the alternating part of the electric displacement from the last equation and substituting in the first part of Eqs. (24) we have

$$T_1 = \frac{S_1}{s_{11}^E} - \frac{2Q_{12}\delta_{30}E_3}{(4\pi\beta_{11}^T + O_{11}\delta_{30})s_{11}^E}; \tag{25}$$

$$\delta_3 = \frac{E_3}{4\pi\beta_{11}^{S1} + O_{11}\delta_{30}} + \frac{2Q_{12}\delta_{30}S_1}{s_{11}^D [4\pi\beta_{11}^{S1} + O_{11}\delta_{30}]},$$

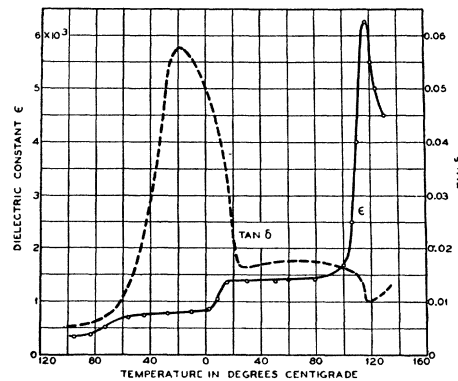


Fig. 6. Dielectric constant for zero bias as a function of the temperature.

where

$$s_{11}^D = s_{11}^E [1 - k^2] \text{ where } k^2 = \frac{4Q_{12}^2 \delta_{30}^2}{[4\pi\beta_{11}^T + O_{11}\delta_{30}]s_{11}^E}.$$

$$4\pi\beta_{11}^{S_1} + O_{11}\delta_{30} = 4\pi\beta_{11}^T + O_{11}\delta_{30} - \frac{4Q_{12}^2 \delta_{30}^2}{s_{11}^D} \\ = (4\pi\beta_{11}^T + O_{11}\delta_{30})(1 - k^2).$$

Substituting the last relation in the last of Eqs. (25), the two equations become

$$T_1 = \frac{S_1}{s_{11}^E} - \frac{2Q_{12}\delta_{30}E_3}{(4\pi\beta_{11}^T + O_{11}\delta_{30})s_{11}^E}; \\ \delta_3 = \frac{E_3}{4\pi\beta_{11}^{S_1} + O_{11}\delta_{30}} + \frac{2Q_{12}\delta_{30}S_1}{s_{11}^E[4\pi\beta_{11}^T + O_{11}\delta_{30}]} \quad (26)$$

These have the same form as the piezoelectric equations (37) of reference (8), and hence the same considerations exist if we set the equivalent piezoelectric constant equal to

$$d_{31}' = \frac{2Q_{12}\delta_{30}}{4\pi\beta_{11}^T + O_{11}\delta_{30}}. \quad (27)$$

This can be evaluated as in the piezoelectric case by measuring the resonant and antiresonant frequencies of the device, the dielectric constant, and the density. The coupling is given by Eq. (50) of reference (8).

$$k^2 = \frac{\pi^2 \Delta f}{4 f_R} \left[ 1 + \frac{(4 - \pi^2) \Delta f}{4 f_R} + \dots \right], \quad (28)$$

while the piezoelectric constant is given by

$$d_{31}' = \frac{2Q_{12}\delta_{30}}{4\pi\beta_{11}^T + O_{11}\delta_{30}} = k \left( \frac{s_{11}^E}{4\pi\beta_{11}^T + O_{11}\delta_{30}} \right)^{\frac{1}{2}},$$

or

$$Q_{12} = \frac{k}{2\delta_{30}} (s_{11}^E (4\pi\beta_{11}^T + O_{11}\delta_{30}))^{\frac{1}{2}}. \quad (29)$$

Measurements have been made for the coupling of a long thin bar as a function of the applied voltage, and the results are shown by Fig. 4. The frequency constant for such a bar is  $2.28 \times 10^5$  kc cm. With a density of 5.5 this corresponds to a compliance constant (inverse of Young's modulus) of  $0.88 \times 10^{-12}$ . The dielectric constant as a

function of voltage is given in Fig. 3. Hence one can calculate the value of  $Q_{12}$ , and the value is approximately

$$Q_{12} = -2.15 \times 10^{-12} \text{ in c.g.s. units.}$$

The negative sign is obtained from expansion measurements which show that the bar contracts in length when a voltage is applied normal to the length.

The same constant  $Q_{12}$  drives the radial mode of a disk, but since this requires a transformation to cylindrical coordinates, the equations are discussed in the appendix. It is there shown that the coupling is  $(2/(1-\sigma))^{\frac{1}{2}}$  times as large as that for the longitudinal mode. This agrees well with the experimental curve of Fig. 4.

The coupling for the thickness mode is also shown by Fig. 4. The effective piezoelectric constant for a thickness mode can be evaluated from Eq. (22) by setting  $S_1 = S_2 = 0$ , since no sidewise motion occurs, and solving for  $T_3$  and  $\delta_3$  in terms of  $S_3$  and  $E_3$ . The resulting equations are

$$T_3 = S_3 c_{11}^E \\ - \frac{2\delta_{30}[Q_{11} - (2s_{12}^D/s_{11}^D + s_{12}^D)Q_{12}]c_{11}^E}{4\pi\beta_{11}^T + O_{11}\delta_{30}} E_3; \quad (30)$$

$$\delta_3 = \frac{E_3}{4\pi\beta_{11}^{S_3} + O_{11}\delta_{30}} \\ + \frac{2c_{11}^E[Q_{11} - (2s_{12}^D/s_{11}^D + s_{12}^D)Q_{12}]\delta_{30}S_3}{[4\pi\beta_{11}^T + O_{11}\delta_{30}]},$$

where

$$c_{11}^E = \frac{c_{11}^D}{1 - k^2}; \\ k^2 = \frac{4\delta_{30}^2[Q_{11} - (2s_{12}^D Q_{12}/s_{11}^D + s_{12}^D)]^2 c_{11}^E}{4\pi\beta_{11}^T + O_{11}\delta_{30}},$$

and

$$4\pi\beta_{11}^{S_3} + O_{11}\delta_{30} = (4\pi\beta_{11}^T + O_{11}\delta_{30})(1 - k^2).$$

Hence the equivalent piezoelectric constant for this case is

$$d_{33}' = \frac{2[Q_{11} - (2s_{12}^D/s_{11}^D + s_{12}^D)Q_{12}]\delta_{30}}{4\pi\beta_{11}^T + O_{11}\delta_{30}}. \quad (31)$$

From the coupling measurements of Fig. 4 one

finds that

$$Q_{11} - \frac{2s_{12}^D}{s_{11}^D + s_{12}^D} Q_{12} = 5.3 \times 10^{-12}. \quad (32)$$

Since  $s_{11}$  and  $s_{12}$  are given in terms of the  $c$  elastic constants by

$$s_{11} = \frac{c_{11} + c_{12}}{c_{11}(c_{11} + c_{12}) - 2c_{12}^2} = \frac{\lambda + \mu}{\mu(3\lambda + 2\mu)}; \quad (33)$$

$$s_{12} = \frac{-c_{12}}{c_{11}(c_{11} + c_{12}) - 2c_{12}^2} = \frac{-\lambda}{2\mu(3\lambda + 2\mu)},$$

we have from the Lamé elastic constants of Eq. (7)

$$s_{11} = 0.88 \times 10^{-12}; \quad s_{12} = -0.236 \times 10^{-12}. \quad (34)$$

Hence the value of  $Q_{11}$  becomes

$$Q_{11} = +6.9 \times 10^{-12} \quad (35)$$

which is of opposite sign and about 3 times as large as  $Q_{12}$ .

The fourth mode of motion that can be generated in a rectangular bar is the thickness shear mode which occurs when the alternating voltage is applied at right angles to the d.c. electric displacement. This mode was tested by taking a bar 5 cm long, 0.5 cm wide, and 0.25 cm thick, applying a voltage of 30,000 volts per cm and using the remanent polarization generated for this case. Since it is difficult to establish an electric displacement along the 0.5-cm direction with plates normal to the large faces, these were dissolved off and the a.c. field applied along the width (0.5-cm direction). The frequency of the measured resonance was 566 kilocycles, which agrees well with the shear elastic constant of Eq. (7). The coupling for the shear mode is higher than that for the thickness longitudinal mode. This is what one expects from Eqs. (22), sixth equation, from which one obtains the equation for a shear mode

$$T_4 = S_4 \mu^E - \frac{2(Q_{11} - Q_{12})\delta_{30}\mu^E}{4\pi\beta_{11}^T + O_{11}\delta_{30}} E_2, \quad (36)$$

$$\delta_2 = \frac{E_2}{4\pi\beta_{11}^S + O_{11}\delta_{30}} - \frac{2(Q_{11} - Q_{12})\delta_{30}\mu^E}{4\pi\beta_{11}^T + O_{11}\delta_{30}} S_4,$$

where

$$\mu^E = \frac{c_{11}^E - c_{12}^E}{2} = \left( \frac{c_{11}^D - c_{12}^D}{2} \right) (1 - k^2);$$

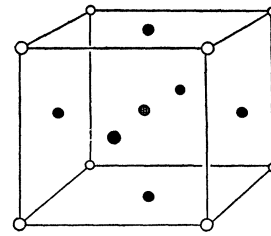
$$k^2 = \frac{4(Q_{11} - Q_{12})^2 \delta_{30} \mu^E}{4\pi\beta_{11}^T + O_{11}\delta_{30}}.$$

Inserting the values given previously for  $Q_{11}$ ,  $Q_{12}$ ,  $\mu$ , and  $\delta_{30}$  (i.e., a remanent polarization equal to 0.85 times that for a 30,000-volt/cm field), one obtains a coupling of 48 percent which agrees well with experiment. Hence the phenomenological theory accounts quantitatively for all the modes of motion observed, and allows one to measure the electrostrictive, elastic, and dielectric constants pertaining to the ceramic.

#### IV. THEORETICAL EXPLANATION OF EFFECT

The ratio of about 2 to 1 between the thickness effect, and the fact that the radial effect is a contraction, allows one to obtain a mechanism for this effect. Barium titanate above 120°C has a cubic structure, having the form shown by Fig. 7. Here eight barium atoms form the corners of the cube. Since each barium atom is shared between eight adjacent cells, this gives a total of one barium atom per cell. Six oxygen atoms occupy the face-centered position on the six sides, and since each is shared between two adjacent cells, this represents a total of three oxygen atoms per cell. Since the titanium atom is much smaller than the other atoms, it is relatively free to move between them.

As the temperature is lowered below 120°C, the titanium atom moves from the center to one of the six positions near the six oxygen atoms. Since the cell was neutral when the titanium was in the center, a dipole moment is introduced by



○ - BARIUM ● - OXYGEN ⊙ - TITANIUM  
UNIT CELL FOR BARIUM TITANATE ABOVE 120°C

FIG. 7. Unit cell for barium titanate above 120°C.



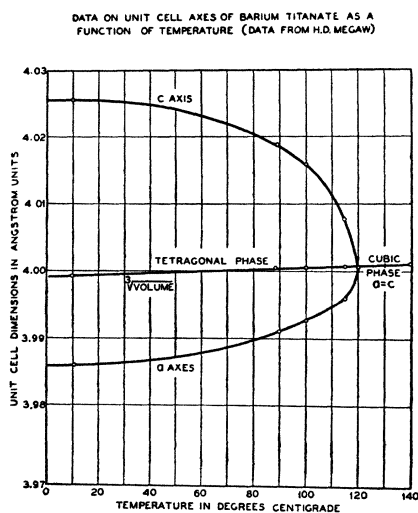


FIG. 8. Unit cell dimensions as a function of the temperature.

the motion of the titanium and the cell acquires a permanent dipole; the domain in which the cell is situated becomes ferro-electric.

When the crystal becomes ferro-electric, the cubic form is lost and the crystal becomes tetragonal with the axis in the direction of the titanium motion 1 percent longer than the other two axes. Figure 8 shows the cell dimensions as a function of temperature, as determined by Miss McGaw.<sup>9</sup> Along the ferro-electric axis the cell dimension increases from 4.0 Å to 4.026 Å, at room temperature, while the other two axes decrease from 4.0 Å to 3.986 Å. The total volume of the unit cell remains unchanged for the crystal, but the axial ratio has changed to 1.01.

For a polycrystalline material the dominant mechanism for producing the electrostrictive effect is the following. As the ceramic material is prepared, all domains (which can exist below 120°C) are equally distributed in all directions and no residual polarization can occur. The effect of a large d.c. field is to change the direction of polarity so that more domains are lined up in the direction of the field rather than in other directions. This change in the direction of a domain occurs not by physically changing the orientation but rather in changing the direction of the ferro-electric axis from one of the six oxygens to another of the six. When the field is

<sup>9</sup> H. D. McGaw, Proc. Roy. Soc. 189, 261-283 (April, 1947).

taken off, the local field caused by the lining up of the domains remains and is sufficient to keep a large share of the domains lined up. Now when domains are lined up in the direction of the field, the plate expands in this direction by  $\frac{2}{3}$  percent times the percentage of domains whose direction of polarization is changed. At the same time the radial dimensions contract. For the crystal, x-ray measurements show that the sidewise contraction is half as much as the thickness expansion. However, for the ceramic, since  $-Q_{12}/Q_{11} = 2.15/6.9 = 0.31$ , the amount of sidewise contraction is less and a volume electrostrictive effect exists. This is probably due to the fact that the crystal domains are not bonded at all points, and a contraction of domains can occur without causing a corresponding contraction in the body, whereas an expansion along the *c* axis carries the material with it whether it is bonded at all points or not. The two effects—the thickness effect and the radial effect—are both of a considerable magnitude.

When a small a.c. field is applied in the presence of a d.c. field or remanent polarization, the following process probably occurs. The a.c. field in itself is too small to reverse any complete domain, but it can cause molecules on the common planes of differently directed domains to change from one domain to another and hence cause one domain to grow at the expense of other domains. If the a.c. field is opposed to the d.c. field, some molecules of the domains directed along the thickness will be lost to other domains directed in different directions and the crystal will become thinner. When the a.c. field is added to the d.c. field, these molecules and more too will be directed in the direction of the field and the plate becomes thicker. Since the change in molecule direction will, in general, lag the applied field, a large dielectric hysteresis occurs just as for rochelle salt, and the mechanical resonances have a poor *Q*. The radial vibration is accounted for by the contraction of the domains in directions perpendicular to the ferro-electric axis, and this process should generate a radial motion about half as large as the thickness motion, which agrees with experiment.

The value of the total increase in thickness, about 5 to 7 parts in  $10^4$  for 30,000 volts per cm applied gives a method for estimating the

number of domains lined up by the d.c. field. Since the plate could expand by  $\frac{2}{3}$  percent if all the domains were lined up, the percent lined up is

$$\frac{5 \text{ to } 7 \times 10^{-4}}{6.6 \times 10^{-3}} = 7.6 \text{ to } 10.6 \text{ percent} \quad (37)$$

over the average value for an isotropic condition.

#### APPENDIX

##### Equation for an Electrostrictive Material in Cylindrical Coordinates

To obtain the equations of motion for a cylindrical plate, Eqs. (22) for rectangular coordinates have to be transformed to cylindrical coordinates. In cylindrical coordinates the variables are the radius vector  $r$ , the angle  $\theta$ , and the dimension along the cylinder designated by  $z$ . In

terms of the  $x, y, z$  rectangular coordinates

$$r^2 = x^2 + y^2; \quad \tan \theta = y/x; \quad z = z. \quad (38)$$

The direction cosines between the  $r, \theta$ , and  $z$  directions and the  $x, y$ , and  $z$  directions are then

$$\begin{array}{c|ccc} & x & y & z \\ \hline r & \cos \theta & \sin \theta & 0 \\ \theta & -\sin \theta & \cos \theta & 0 \\ z & 0 & 0 & 1 \end{array} \quad (39)$$

Making use of the formula for the transformation of a tensor from one coordinate system to another,

$$T_{a,b} = \frac{\partial x_a}{\partial x_k} \frac{\partial x_b}{\partial x_l} T_{kl}, \quad (40)$$

letting  $a, b$  refer to  $r, \theta$ , and  $z$  and noting that  $\partial x_a / \partial x_k$  are the direction cosines of Eq. (39), the stress tensor becomes in cylindrical coordinates

$$\begin{aligned} T_{rr} &= \cos^2 \theta T_{11} + 2 \sin \theta \cos \theta T_{12} + \sin^2 \theta T_{22}, \\ T_{\theta\theta} &= \sin^2 \theta T_{11} - 2 \sin \theta \cos \theta T_{12} + \cos^2 \theta T_{22}, \\ T_{r\theta} &= \sin \theta \cos \theta [T_{22} - T_{11}] + [\cos^2 \theta - \sin^2 \theta] T_{12}, \\ T_{rz} &= \cos \theta T_{13} + \sin \theta T_{23}, \\ T_{\theta z} &= -\sin \theta T_{13} + \cos \theta T_{23}, \\ T_{zz} &= T_{33}. \end{aligned} \quad (41)$$

The strain tensor transforms in a similar manner.

Conversely, the rectangular stress and strain components are related to the cylindrical components by equations of the type

$$S_{ij} = \frac{\partial x_i}{\partial x_a} \frac{\partial x_j}{\partial x_b} S_{ab}, \quad (42)$$

and

$$\begin{aligned} S_{11} &= \cos^2 \theta S_{rr} - 2 \sin \theta \cos \theta S_{r\theta} + \sin^2 \theta S_{\theta\theta}, \\ S_{22} &= \sin^2 \theta S_{rr} + 2 \sin \theta \cos \theta S_{r\theta} + \cos^2 \theta S_{\theta\theta}, \\ S_{33} &= S_{zz}, \\ S_{12} &= \sin \theta \cos \theta (S_{rr} - S_{\theta\theta}) + (\cos^2 \theta - \sin^2 \theta) S_{r\theta}, \\ S_{13} &= \cos \theta S_{rz} - \sin \theta S_{\theta z}, \\ S_{23} &= \sin \theta S_{rz} + \cos \theta S_{\theta z}. \end{aligned} \quad (43)$$

In solving the equations of motion it is necessary to know the values of the strains in terms of the displacements in the  $r, \theta$ , and  $z$  directions. Denoting these by

$$u_r, u_\theta, \text{ and } u_z, \quad (44)$$

Love<sup>10</sup> has shown that the strain components are given by

$$\begin{aligned} S_{rr} &= \frac{\partial u_r}{\partial r}; \quad S_{\theta\theta} = \frac{1}{r} \frac{\partial u_\theta}{\partial \theta} + \frac{u_r}{r}; \quad S_{zz} = \frac{\partial u_z}{\partial z} \\ S_{r\theta} &= \frac{\partial u_\theta}{\partial r} - \frac{u_\theta}{r} + \frac{1}{r} \frac{\partial u_r}{\partial \theta}; \quad S_{rz} = \frac{\partial u_r}{\partial z} + \frac{\partial u_z}{\partial r}; \quad S_{\theta z} = \frac{1}{r} \frac{\partial u_z}{\partial \theta} + \frac{\partial u_\theta}{\partial z}. \end{aligned} \quad (45)$$

<sup>10</sup> Love, *Theory of Elasticity* (Cambridge University Press, London, 1934), fourth edition, p. 56.

The other necessary equations are the Newton's law equations transformed into cylindrical coordinates. From Love's<sup>11</sup> *Theory of Elasticity*, these are

$$\begin{aligned}\rho\ddot{u}_r &= \frac{\partial T_{rr}}{\partial r} + \frac{1}{r} \frac{\partial T_{r\theta}}{\partial \theta} + \frac{\partial T_{rz}}{\partial z} + \left( \frac{T_{rr} - T_{\theta\theta}}{r} \right), \\ \rho\ddot{u}_\theta &= \frac{\partial T_{r\theta}}{\partial r} + \frac{1}{r} \frac{\partial T_{\theta\theta}}{\partial \theta} + \frac{\partial T_{\theta z}}{\partial z} + \frac{2T_{r\theta}}{r}, \\ \rho\ddot{u}_z &= \frac{\partial T_{rz}}{\partial r} + \frac{1}{r} \frac{\partial T_{\theta z}}{\partial \theta} + \frac{\partial T_{zz}}{\partial z} + \frac{T_{rz}}{r}.\end{aligned}\quad (46)$$

These equations give enough relations to solve the case of the electrostrictive disk of barium titanate in radial vibration. Equations (22) give the electrostrictive and elastic equations in rectangular coordinates. Transferring these to cylindrical coordinates by means of the tensor relations of Eqs. (41) and (43), we find

$$\begin{aligned}S_{rr} &= s_{11}^D T_{rr} + s_{12}^D (T_{\theta\theta} + T_{zz}) + Q_{11} \delta_r^2 + Q_{12} [\delta_\theta^2 + \delta_z^2], \\ S_{\theta\theta} &= s_{12}^D [T_{rr} + T_{zz}] + s_{11}^D T_{\theta\theta} + Q_{12} (\delta_r^2 + \delta_z^2) + Q_{11} \delta_\theta^2, \\ S_{zz} &= s_{11}^D T_{zz} + s_{12}^D [T_{rr} + T_{\theta\theta}] + Q_{11} \delta_z^2 + Q_{12} [\delta_r^2 + \delta_\theta^2], \\ S_{rz} &= (s_{11}^D - s_{12}^D) T_{rz} + (Q_{11} - Q_{12}) \delta_r \delta_z, \\ S_{r\theta} &= (s_{11}^D - s_{12}^D) T_{r\theta} + (Q_{11} - Q_{12}) \delta_r \delta_\theta, \\ S_{\theta z} &= (s_{11}^D - s_{12}^D) T_{\theta z} + (Q_{11} - Q_{12}) \delta_\theta \delta_z,\end{aligned}\quad (47)$$

where  $\delta_r$ ,  $\delta_\theta$ ,  $\delta_z$  are the electric displacements divided by  $4\pi$  for the  $r$ ,  $\theta$ , and  $z$  directions. The electric relations become

$$\begin{aligned}E_r &= \delta_r [4\pi\beta_{11}^T + O_{11}\delta_r] - 2[Q_{11}(\delta_r T_{rr} + \delta_\theta T_{r\theta} + \delta_z T_{rz}) + Q_{12}[\delta_r(T_{\theta\theta} + T_{zz}) - (\delta_\theta T_{r\theta} + \delta_z T_{rz})]], \\ E_\theta &= \delta_\theta [4\pi\beta_{11}^T + O_{11}\delta_\theta] - 2[Q_{11}(\delta_\theta T_{\theta\theta} + \delta_r T_{r\theta} + \delta_z T_{\theta z}) + Q_{12}[\delta_\theta(T_{rr} + T_{zz}) - (\delta_r T_{r\theta} + \delta_z T_{\theta z})]], \\ E_z &= \delta_z [4\pi\beta_{11}^T + O_{11}\delta_z] - 2[Q_{11}(\delta_z T_{zz} + \delta_r T_{rz} + \delta_\theta T_{\theta z}) + Q_{12}[\delta_z(T_{rr} + T_{\theta\theta}) - (\delta_r T_{rz} + \delta_\theta T_{\theta z})]].\end{aligned}\quad (48)$$

For the radially vibrating disk

$$u_\theta = 0, \quad (49)$$

and we have also

$$T_{\theta z} = T_{r\theta} = 0. \quad (50)$$

We assume that the thickness along  $z$  is very small compared to the wave-length. Since the stresses on the surface are zero and the thickness very small, we can set

$$T_{zz} = T_{rz} = 0. \quad (51)$$

These are the same assumptions as those made for a longitudinal long thin bar discussed in Eqs. (23) to (29) and hence the results are comparable. Since a field is applied only along the  $z$  direction,  $\delta_r = \delta_\theta = 0$ . The remaining equations then become

$$\begin{aligned}S_{rr} &= s_{11}^D T_{rr} + s_{12}^D T_{\theta\theta} + Q_{12} \delta_z^2, \\ S_{\theta\theta} &= s_{12}^D T_{rr} + s_{11}^D T_{\theta\theta} + Q_{12} \delta_z^2, \\ E_z &= \delta_z [4\pi\beta_{11}^T + O_{11}\delta_z] - 2Q_{12} \delta_z [T_{rr} + T_{\theta\theta}].\end{aligned}\quad (52)$$

<sup>11</sup> See reference 10, p. 90.

To insert in the equation of motion (46) we need to have the stresses expressed in terms of the strains, and for the electrical boundary conditions it is better to use the fields rather than the electric displacements. Furthermore, for small alternating fields superposed on a large d.c. electric displacement, which may be caused by an applied field or a remanent polarization, we can replace  $\delta_z$  by

$$\delta_z = \delta_{z0} + \delta_z e^{i\omega t}, \quad (53)$$

where  $\delta_{z0}$  is the steady electric displacement if a field is applied or the remanent polarization  $P_0$  if the field is taken off. Then solving these three equations simultaneously, the alternating components of stress, strain, and displacement are

given by the equations

$$\begin{aligned} T_{rr} &= \left[ \frac{s_{11}^E}{s_{11}^{E^2} - s_{12}^{E^2}} \right] S_{rr} - \left[ \frac{s_{12}^E}{s_{11}^{E^2} - s_{12}^{E^2}} \right] S_{\theta\theta} - \frac{2Q_{12}\delta_{z0}E_z}{(4\pi\beta_{11}^T + O_{11}\delta_{z0})(s_{11}^E + s_{12}^E)}, \\ T_{\theta\theta} &= \left[ \frac{s_{11}^E}{s_{11}^{E^2} - s_{12}^{E^2}} \right] S_{\theta\theta} - \left[ \frac{s_{12}^E}{s_{11}^{E^2} - s_{12}^{E^2}} \right] S_{rr} - \frac{2Q_{12}\delta_{z0}E_z}{(4\pi\beta_{11}^T + O_{11}\delta_{z0})(s_{11}^E + s_{12}^E)}, \\ \delta_z &= \frac{E_z}{4\pi\beta_{11}^T + O_{11}\delta_{z0}} + \frac{2Q_{12}\delta_{z0}}{4\pi\beta_{11}^T + O_{11}\delta_{z0}} [T_{rr} + T_{\theta\theta}], \end{aligned} \quad (54)$$

where

$$s_{11}^E = \frac{s_{11}^D}{1 - k_l^2}; \quad s_{12}^E = \frac{s_{12}^D}{1 - \frac{s_{11}^D}{s_{12}^D} k_l^2}; \quad k_l^2 = \frac{4Q_{12}^2\delta_{z0}^2}{(4\pi\beta_{11}^T + O_{11}\delta_{z0})s_{11}^E}.$$

$k_l$  is the electromechanical coupling factor for a longitudinal mode given by Eq. (25). We note that since  $1/s_{11}^E = Y_0^E$ , the Young's modulus, and  $-s_{12}^E/s_{11}^E = \sigma$ , the Poisson ratio, that the first two equations of (54) can be simplified to

$$\begin{aligned} T_{rr} &= \left( \frac{Y_0^E}{1 - \sigma^2} \right) [S_{rr} + \sigma S_{\theta\theta}] - \frac{2Q_{12}\delta_{z0}E_z Y_0^E}{(4\pi\beta_{11}^T + O_{11}\delta_{z0})(1 - \sigma)}, \\ T_{\theta\theta} &= \left( \frac{Y_0^E}{1 - \sigma^2} \right) [S_{\theta\theta} + \sigma S_{rr}] - \frac{2Q_{12}\delta_{z0}E_z Y_0^E}{(4\pi\beta_{11}^T + O_{11}\delta_{z0})(1 - \sigma)}. \end{aligned} \quad (55)$$

Now noting that since the plating on the surface is an equipotential surface,  $E_z$  is not a function of  $r$ , then when Eqs. (55) are inserted in the equation of motion, (46), and the relations for a radial motion

$$S_{rr} = \frac{\partial u_r}{\partial r}; \quad S_{\theta\theta} = \frac{u_r}{r} \quad (56)$$

are used, the equation of motion becomes

$$\frac{Y_0^E}{1 - \sigma^2} \left[ \frac{\partial^2 u_r}{\partial r^2} + \frac{1}{r} \frac{\partial u_r}{\partial r} - \frac{u_r}{r^2} \right] = \rho \frac{\partial^2 u_r}{\partial t^2} = -\omega^2 \rho u_r. \quad (57)$$

The last term results for simple harmonic motion.

Since this is a Bessel's equation of the first order, a solution is

$$u_r = A J_1 \left( \frac{\omega r}{v} \right), \quad \text{where} \quad v = \frac{Y_0^E}{(1 - \sigma^2)\rho}. \quad (58)$$

No Bessel's function of the second kind is required since the displacement  $u_r$  vanishes at the center of the disk. At the boundary when  $r = a$  the radius of the disk, the stress  $T_{rr} = 0$ . From (55) we have

$$0 = \frac{Y_0^E A}{1 - \sigma^2} \left[ \frac{\omega}{v} J_0 \left( \frac{\omega a}{v} \right) - \frac{(1 - \sigma) J_1(\omega a/v)}{a} \right] - \frac{2Q_{12}\delta_{z0}E_z Y_0^E}{(4\pi\beta_{11}^T + O_{11}\delta_{z0})(1 - \sigma)}. \quad (59)$$

Hence

$$A = \frac{2Q_{12}\delta_{z0}E_z(1 + \sigma)}{[4\pi\beta_{11}^T + O_{11}\delta_{z0}] \left[ \frac{\omega}{v} J_0 \left( \frac{\omega a}{v} \right) - \frac{(1 - \sigma) J_1(\omega a/v)}{a} \right]}. \quad (60)$$

From this data we can evaluate the stresses  $T_{rr}$  and  $T_{\theta\theta}$  as

$$\begin{aligned}
 T_{rr} &= -\frac{2Q_{12}\delta z_0 Y_0^E E_z}{(4\pi\beta_{11}^T + O_{11}\delta z_0)(1-\sigma)} \left[ 1 - \frac{\left[ \frac{\omega}{v} J_0\left(\frac{\omega r}{v}\right) - \frac{(1-\sigma)J_1(\omega r/v)}{r} \right]}{\left[ \frac{\omega}{v} J_0\left(\frac{\omega a}{v}\right) - \frac{(1-\sigma)J_1(\omega a/v)}{a} \right]} \right], \\
 T_{\theta\theta} &= -\frac{2Q_{12}\delta z_0 E_z Y_0^E}{(4\pi\beta_{11}^T + O_{11}\delta z_0)(1-\sigma)} \left[ 1 - \frac{\left[ \frac{\sigma\omega}{v} J_0\left(\frac{\omega r}{v}\right) + \frac{(1-\sigma)J_1(\omega r/v)}{r} \right]}{\left[ \frac{\omega}{v} J_0\left(\frac{\omega a}{v}\right) - \frac{(1-\sigma)J_1(\omega a/v)}{a} \right]} \right].
 \end{aligned}
 \tag{61}$$

The next step in the solution is to obtain the electrical impedance by a.c. methods. This can be obtained by substituting the values of  $T_{rr}$  and  $T_{\theta\theta}$  in the last of Eqs. (54) and integrating over the surface of the crystal. Since the value of  $\delta_z$  at the surface is equal to the surface charge this will evaluate the total charge  $Q$  and we have

$$Q = \int_0^{2\pi} d\theta \int_0^a \delta_z r dr = \frac{E_z \pi a^2}{(4\pi\beta_{11}^T + O_{11}\delta z_0)} + \frac{2Q_{12}\delta z_0}{(4\pi\beta_{11}^T + O_{11}\delta z_0)} \int_0^{2\pi} d\theta \int_0^a r [T_{rr} + T_{\theta\theta}] dr.
 \tag{62}$$

Introducing the value of  $T_{rr}$  and  $T_{\theta\theta}$  from Eq. (61) this integral becomes

$$Q = \frac{E_z \pi a^2}{(4\pi\beta_{11}^T + O_{11}\delta z_0)} \left[ 1 - \frac{8Q_{12}\delta z_0 Y_0^E}{(4\pi\beta_{11}^T + O_{11}\delta z_0)(1-\sigma)} \left[ 1 - \frac{(1+\sigma) \int_0^a r^2 J_0(\omega r/v) dr}{a^2 \left( \frac{\omega}{v} J_0(\omega a/v) - \frac{(1-\sigma)}{a} J_1(\omega a/v) \right)} \right] \right].
 \tag{63}$$

Performing the integration and employing the substitution

$$\frac{1}{(4\pi\beta_{11}^T + O_{11}\delta z_0)} \left[ 1 - \frac{8Q_{12}^2 \delta z_0^2 Y_0^E}{(4\pi\beta_{11}^T + O_{11}\delta z_0)(1-\sigma)} \right] = \frac{1}{4\pi\beta_{11}^{RC} + O_{11}\delta z_0},
 \tag{64}$$

where  $\beta_{11}^{RC}$  is the radially clamped impermeability constant, i.e., the impermeability when the plate is prevented from moving radially, we have

$$Q = \frac{E_z \pi a^2}{(4\pi\beta_{11}^{RC} + O_{11}\delta z_0)} \left[ 1 + \frac{k^2}{(1-k^2)} \frac{(1+\sigma)J_1(\omega a/v)}{(\omega a/v)J_0(\omega a/v) - (1-\sigma)J_1(\omega a/v)} \right],
 \tag{65}$$

where the coefficient of coupling for a radial mode becomes

$$k_r^2 = \frac{8Q_{12}^2 \delta z_0^2 Y_0^E}{(4\pi\beta_{11}^T + O_{11}\delta z_0)(1-\sigma)}.
 \tag{66}$$

Comparing this with the longitudinal mode, given by Eq. (25), we see that the radial mode has a higher coupling by the factor

$$(2/1-\sigma)^{\frac{1}{2}}.
 \tag{67}$$

Since the admittance of the plate is equal to the current into the plate divided by the voltage across

the plate, and for simple harmonic motion

$$i = dQ/dt = j\omega Q, \quad (68)$$

the impedance of the electrostrictive plate is given by

$$\frac{1}{Z} = \frac{i}{E} = \frac{j\omega\pi a^2}{(4\pi\beta_{11}^{RC} + O_{11}\delta_{z0})l_t} \left[ 1 + \frac{k^2}{1-k^2} \frac{(1+\sigma)J_1(\omega a/v)}{(\omega a/v)J_0(\omega a/v) - (1-\sigma)J_1(\omega a/v)} \right]. \quad (69)$$

The resonant frequency occurs when

$$(\omega a/v)J_0(\omega a/v) - (1-\sigma)J_1(\omega a/v) = 0. \quad (70)$$

For a value of  $\sigma = 0.27$  found from the elastic measurements, this equation has the lowest root

$$(\omega a/v) = 2.03 = R_1. \quad (71)$$

Hence the frequency is given by the equation

$$f_R = (2.03/2\pi a)(Y_0^E/\rho(1-\sigma^2))^{1/2}. \quad (72)$$

The antiresonant frequency occurs when the expression in brackets in Eq. (69) reduces to zero. This occurs at a frequency somewhat above the resonant frequency. To determine the frequency separation  $\Delta f$ , between resonance and antiresonance, we develop the function  $J_0(\omega a/v)$  and  $J_1(\omega a/v)$  in a MacLaurin's series about the root  $R_1$ . This gives

$$\begin{aligned} J_0(\omega a/v) &= J_0(R_1) + \frac{\partial}{\partial f} [J_0(\omega a/v)]_{(\omega a/v)=R_1} \Delta f + \dots \\ &= J_0(R_1) - \frac{2\pi a}{v} J_1(R_1) \Delta f + \dots, \end{aligned} \quad (73)$$

$$\begin{aligned} J_1(\omega a/v) &= J_1(R_1) + \frac{\partial}{\partial f} [J_1(\omega a/v)]_{(\omega a/v)=R_1} \Delta f + \dots \\ &= J_1(R_1) + \frac{2\pi a}{v} \left[ J_0(R_1) - \frac{J_1(R_1)}{R_1} \right] \Delta f + \dots \end{aligned}$$

Inserting these values in Eq. (69) and setting the numerator equal to zero, the frequency separation  $\Delta f$  becomes

$$\frac{\Delta f}{f_R} = \frac{(k^2/(1-k^2))(1+\sigma)}{R_1^2 - (1-\sigma^2)}. \quad (74)$$

Hence solving for the coupling factor  $k^2$ , we find to a first approximation

$$k^2 = \frac{\Delta f}{f_R} \left[ \frac{R_1^2 - (1-\sigma^2)}{1+\sigma} \right]. \quad (75)$$

For  $\sigma = 0.27$  the value of the factor multiplying  $\Delta f/f_R$  is equal to 2.51. Comparing this to the factor for a longitudinal crystal given by Eq. (28), which is  $\pi^2/4 = 2.47$ , it is seen that the same equations are very nearly applicable. By using Eq. (75) the coupling can be evaluated by measuring the separation of resonant and antiresonant frequencies and the frequency  $f_R$ . The resonant frequency and the density give the elastic constant by employing Eq. (72). From the measured value of  $k_r$ , the measured value of the dielectric constant, which is the inverse of

$$\epsilon_{11} = \frac{1}{\beta_{11}^T + \frac{O_{11}\delta_{z0}}{4\pi}}, \quad (76)$$

one can determine  $Q_{12}\delta_{z0}$  from Eq. (66). By taking a known value of field for which  $\epsilon_{11}$  has been measured,  $\delta_{z0}$  is determined by the equation

$$\delta_{z0} = \frac{E_z}{(4\pi\beta_{11}^T + O_{11}\delta_{z0})} = \frac{E_z\epsilon_{11}}{4\pi}.$$

By observing how the coupling varies around a hysteresis loop, one can evaluate the remanent polarization.