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PROPOSAL FOR THE INVESTIGATION OF
THE EFFECTS OF STRESS ON POLARIZED
FERROELECTRIC CERAMICS

Robert W. Timme

Naval Research Laboratory
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An investigation of the effects of multidimensional sustained stresses on polarized ferroelectric ceramics is proposed. The ceramics to be studied will be those lead zirconate titanate types I, II, and III normally used in underwater sound transducers. Characterization of the ceramics will be accomplished by measuring the elements of the piezoelectric constant tensors d and g and the dielectric constant tensor ε as a function of stress magnitude, dimensionality, and orientation.

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Abstract

An investigation of the effects of multidimensional sustained stresses on polarized ferroelectric ceramics is proposed. The ceramics to be studied will be those lead zirconate titanate Types I, II, and III normally used in underwater sound transducers. Characterization of the ceramics will be accomplished by measuring the elements of the piezoelectric constant tensors d and g and the dielectric constant tensor ϵ as a function of stress magnitude, dimensionality, and orientation.

Problem Status

This is a proposal for a new program.

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PROPOSAL FOR THE INVESTIGATION OF THE EFFECTS OF STRESS ON POLARIZED FERROELECTRIC CERAMICS

Introduction

Most of the underwater sound transducers used by the U. S. Navy contain polarized ferroelectric (piezoelectric) ceramics as their acoustic elements. The advantages of these ceramics include good piezoelectric activity, high dielectric constant, adaptability to various shapes and sizes, and relatively low cost; however, these advantages are offset to some extent by the tendency for the ceramic characteristics to change with time, to be dependent on past history, and to be affected by temperature and stress.

Statement of the Problem

Transducers are designed according to specifications generated by the fleet. Operating conditions to which the transducer will be exposed may vary from 0°C to 40°C and from atmospheric pressure to 110 MPa. The transducer may be pressure and temperature cycled or deployed permanently at some position. The designer attempts to satisfy the specifications by an appropriate choice of ceramic type and configuration. He usually must base his decisions on values of ceramic characteristics known only at room temperature and pressure and with no thought of history effects.

The problem is that the piezoelectric and dielectric constants are functions of temperature, stress, cycling, and time. Thus, the free-field voltage sensitivity, the transmitting current response, or the impedance of a transducer may be exactly as needed at room temperature and pressure, but may be completely unacceptable under the intended operating conditions.

A complete characterization of the piezoelectric ceramics with respect to all the possible environmental parameters would be a very complex and time consuming, albeit ultimately desirable, undertaking. Therefore, this proposal is concerned only with an investigation of the effects of sustained stress on piezoelectric ceramics with all other variables remaining constant. The effects of stress that will be considered depend upon the stress dimensionality, magnitude, and orientation with respect to the direction of polarization.

The stresses experienced by a ceramic acoustic element are generated by the action of the hydrostatic pressure of the underwater environment

upon the particular geometry of the element. The stresses can be hydrostatic, as for ceramic plates, disks, rods, and free-flooded cylinders or spheres. They can be three dimensional, but not hydrostatic, for plates, disks, and flooded cylinders under a bias stress, and for thick-walled capped cylinders and spheres. The stresses can be two-dimensional for thin-walled cylinders and spheres, and one-dimensional for shielded plates, disks, and thin-walled cylinders. The stresses always are compressive.

Solution of the Problem

At present, transducer designers rely heavily upon past experience and a cut-and-try approach. An example of this is described in NRL Report 7644 [1] which traces the design and development of a hydrophone having a sensitivity of -149 dB re 1 V/ μ Pa and experiencing an operational hydrostatic pressure of 46 MPa. The high sensitivity could best be achieved by a capped thin-walled cylinder, but yet the walls could not be too thin or substantial depolarization and, hence, loss of sensitivity, would result. Because few data on the effects of two-dimensional stresses on the polarized ceramic were available, it was necessary to measure the sensitivity as a function of pressure for several configurations. Although a workable solution was found, there was no guarantee of optimization, which illustrates the main problem with the cut-and-try method. Transducers can be designed eventually, but each design usually is good for only that one particular set of conditions.

A better approach to the problem would be to understand how stresses individually and collectively act to affect the polarization, and therefore the piezoelectric and dielectric characteristics, of the ceramic.

Any acoustic element, whether it be in the shape of a plate, cylinder, or sphere, can be divided up mathematically into a number of small unit cubes with the appropriate boundary conditions. These cubes must be so small that there will be no stress gradients. If the response of these unit elements is known for the various combinations of multidimensional stress, then the effects of the stress on any arbitrary shape can be predicted by an appropriate summing procedure.

This form of solution is not original; for example, it is presently used in fields ranging from atomic physics (where the linear combination of atomic orbitals is used to describe the wave function of a molecule) to electrical engineering (complex circuit analysis); however, the extent of its usefulness is predicated upon the completeness and accuracy with which individual unit elements can be characterized.

Technical Description of the Program

Objective

The objective of this program is to characterize the effects of multi-dimensional sustained stresses on polarized ferroelectric ceramics.

The ceramics to be investigated will be those lead zirconate titanate types normally used in underwater sound transducers. They are designated Types I, II, and III in accordance with MIL-STD-1376 (SHIPS) [2]. Characterization will be accomplished by measuring the elements of the piezoelectric constant tensors d and g and the dielectric constant tensor ϵ as a function of stress magnitude, dimensionality, and orientation.

The data obtained will be analyzed and presented in tables, charts, and graphs that can be directly applied to transducer design and also used for correlation with the theory of domain or dipole switching.

Discussion

Ferroelectric ceramics are polycrystalline materials that have a crystal structure nonsymmetric in such a way as to produce a permanent electrical dipole moment. Interaction between the dipole moments of neighboring unit cells results in a domain with aligned dipoles. Initially, the many domains of an entire body are randomly orientated so that no net polarization exists. An externally applied electric field will align many of the domains and thus create a body with a remanent polarization that has many of the piezoelectric properties of certain natural crystals. A stress acting alone cannot polarize a ferroelectric ceramic because of the "gerade" nature of the stress tensor. However, a sufficiently large stress can depolarize a previously poled ceramic by making random domain alignment in the plane perpendicular to the stress more energetically favorable.

A detailed theory describing the interaction of stress and the domain structure has not been formulated. The work to date has been empirical with limited observations of one-dimensional [3-9], two-dimensional [10,11], and three-dimensional stresses [12]. The one-dimensional work is fairly good and limited only in the magnitude of stress application, to the accuracy of the data, and to a few orientations; however, the two- and three-dimensional work is completely inadequate to cover the possible combinations of stress.

To decide which stresses are significant and which effects are to be measured, one should consider first what stresses are present in a ceramic transducer element deployed far beneath the ocean surface. Let the pressure at some given depth be designated p . For a simple plate or disk or for a free-flooded cylinder or sphere, where all surfaces are exposed to the water, the stress will be equivalent to the hydrostatic pressure p . Often, a bias stress is applied either parallel or perpendicular to the direction of polarization for a ceramic stack or a free-flooded cylinder. This stress then would be three-dimensional, but not hydrostatic. The effects of the hydrostatic versus three-dimensional stresses would naturally be different. A capped cylinder would experience the stresses [13] given in Eqs. (1-3):

$$T_r = p \frac{1 - (a/r)^2}{1 + (a/b)^2}, \quad (1)$$

$$T_\theta = p \frac{1 + (a/r)^2}{1 - (a/b)^2}, \quad (2)$$

$$T_z = p \frac{1}{1 - (a/b)^2}. \quad (3)$$

The stresses T_r , T_θ , and T_z are compressive and are in cylindrical coordinate notation indicating, respectively, radial, circumferential, and axial orientation. The inner radius of the cylinder is a and the outer radius is b . From these equations it can be seen that a three-dimensional stress can be present in a thick-walled cylinder, but, for sufficiently thin walls, the stress can be approximated as two dimensional. In either case, T_θ and T_z may be much larger than the hydrostatic pressure p . One should note also that T_r and T_θ are not constant throughout the cylinder wall, but are functions of radial position. If the ends of the cylinder are shielded from the hydrostatic pressure rather than capped, then $T_z = 0$ and only a two-dimensional stress is present in a thick wall and approximately a one-dimensional stress in a thin wall. The stresses in a sphere [14] are given by

$$T_r = p \frac{1 - (a/r)^3}{1 - (a/b)^3}, \quad (4)$$

$$T_\theta = T_\phi = p \frac{1 + \frac{1}{2}(a/r)^3}{1 - (a/b)^3}. \quad (5)$$

In these equations, the stresses T_r , T_θ , and T_ϕ are in spherical coordinates indicating, respectively, radial, circumferential, and azimuthal orientation. Much the same observations can be made here as was done for the cylinder in that, depending upon the wall thickness, either two- or three-dimensional stresses must be considered and that T_θ and T_ϕ always are greater than the hydrostatic pressure.

From these remarks it is evident that a unit volume of a ceramic acoustic element of a transducer may be subjected to stresses much larger than the hydrostatic pressure and to multidimensional stresses, depending upon the element's configuration.

It is proposed in this program that the one-dimensional work be repeated and extended to higher stress magnitudes and that the measurements

be made with greater accuracy. The orientations that will be investigated are listed below and illustrated in Fig. 1.

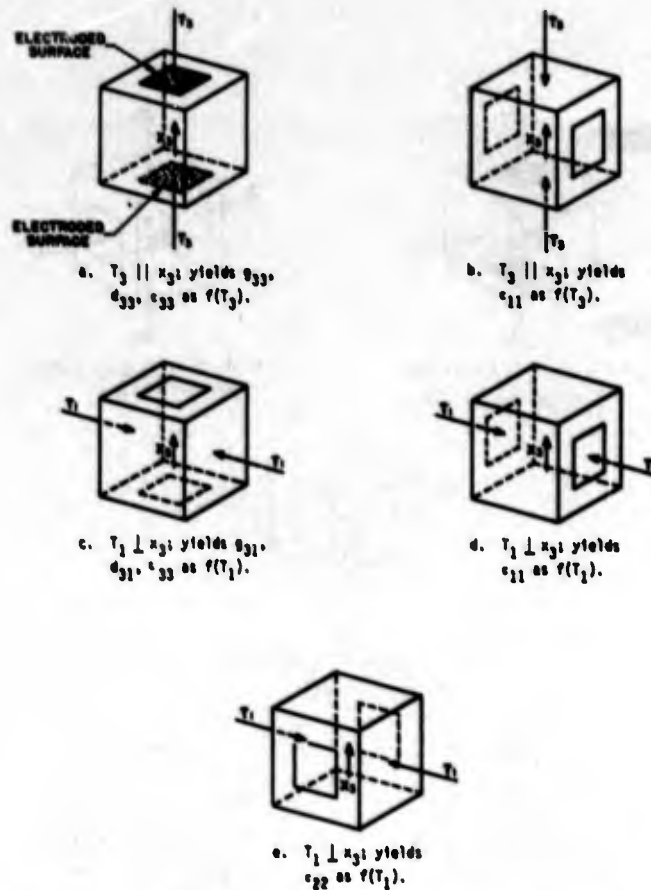


Fig. 1. Five cases of one-dimensional stress orientation and parameters to be determined.

a. Stress parallel (T_{\parallel}) to the polar axis (x_3) and measurements of g_{33} , d_{33} , and ϵ_{33} as a function of stress ($T_3 = T_{\parallel}$).

b. Stress parallel to the polar axis and measurements of ϵ_{11} as a function of stress T_3 .

c. Stress perpendicular (T_{\perp}) to the polar axis and measurements of g_{31} , d_{31} , and ϵ_{33} as a function of stress ($T_1 = T_{\perp}$).

d. Stress perpendicular to the polar axis and measurements of ϵ_{11} as a function of stress T_1 .

e. Stress perpendicular to the polar axis and measurements of ϵ_{22} as a function of stress T_1 .

After processing, the data will be presented in graphs of the various parameters as functions of stress magnitude and orientation for the three types of ceramic material.

The two-dimensional orientations are listed below and illustrated in Fig. 2.

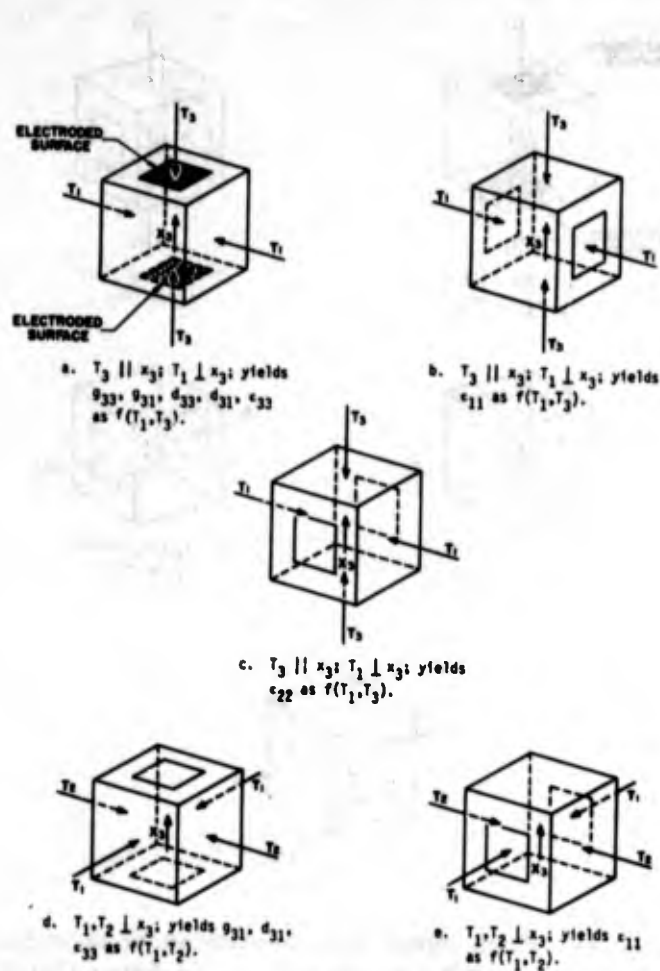


Fig. 2. Five cases of two-dimensional stress and parameters to be determined.

a. One stress parallel ($T_{\parallel} = T_3$) and one stress perpendicular ($T_{\perp} = T_1$) to the polar axis and measurements of g_{33} , g_{31} , d_{33} , d_{31} , and ϵ_{33} for combinations of stresses T_1 and T_3 .

b. One stress parallel and one stress perpendicular to the polar axis and measurements of ϵ_{11} for combinations of stresses T_1 and T_3 .

c. One stress parallel and one stress perpendicular to the polar axis and measurement of ϵ_{22} for combinations of T_1 and T_3 .

d. Both stresses perpendicular (T_1 and T_2) to the polar axis x_3 and measurements of g_{31} , d_{31} , and ϵ_{33} for combinations of stresses T_1 and T_2 .

e. Both stresses perpendicular to the polar axis and measurements of ϵ_{11} for combinations of stresses T_1 and T_2 .

These data will be presented in graphs containing families of curves for the various parameters as functions of magnitude and orientation of stresses. An example of this presentation is given in Fig. 3.

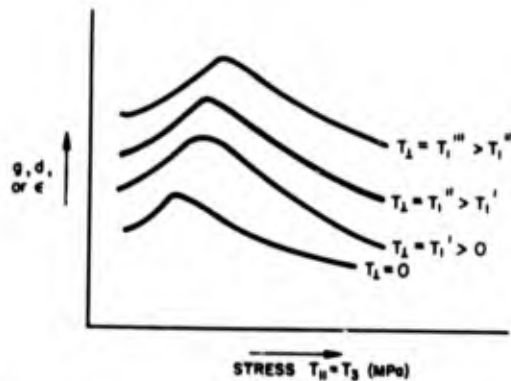


Fig. 3. Example presentation of the effects of two-dimensional stresses on piezoelectric ceramic.

The three-dimensional orientations are listed below and illustrated in Fig. 4.

a. Two stresses perpendicular (T_1 and T_2) and one stress parallel (T_3) to the polar axis and measurements of g_{33} , g_{31} , d_{33} , d_{31} , and ϵ_{33} for combinations of stresses T_1 , T_2 , T_3 .

b. Two stresses perpendicular (T_1 and T_2) and one stress parallel (T_3) to the polar axis and measurements of ϵ_{11} for combinations of stresses T_1 , T_2 , T_3 .

These data also will be presented in graphical form.

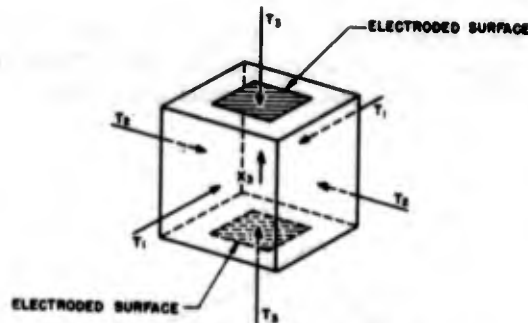
The stress magnitudes will be limited in general to values that produce a reduction of piezoelectric or dielectric constants to about 25% of their initial values. This change is expected to be sufficient to allow correlation with theory and application to transducer design.

Approach

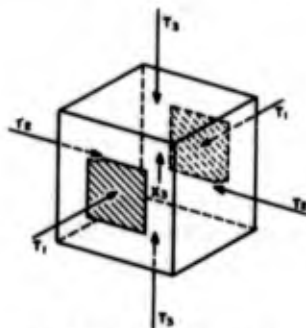
This proposed investigation will be divided into three phases:

- Phase 1. Effects of one-dimensional stresses
- Phase 2. Effects of two-dimensional stresses
- Phase 3. Effects of three-dimensional stresses

The division is natural and progresses from the least to the more difficult to accomplish. Each phase will begin with a study and optimization



a. $T_3 \parallel X_3; T_1, T_2 \perp X_3; \text{yields } \epsilon_{33}, d_{33}, \epsilon_{31}, d_{31}, d_{3l} \text{ as } f(T_1, T_2, T_3).$



b. $T_3 \parallel X_3; T_1, T_2 \perp X_3; \text{yields } \epsilon_{11} \text{ as } f(T_1, T_2, T_3).$

Fig. 4. Two cases of three-dimensional stress orientation and parameters to be determined.

of experimental procedure. It will include such items as size of sample, application of stress, and measurement techniques for parameters. The second part of each phase will be the collection of data on Types I, II, and III ceramic. The third part will be data reduction, analysis, and presentation.

It is estimated that Phase 1 will require six man-months; Phase 2, twelve man-months; and Phase 3, eighteen man-months--for a total program time of three man-years.

Utilization of Study Data

Applications

The data will have immediate application to the problems of transducer design because the graphs and charts obtained should indicate which type

of ceramic would be applicable for a particular stress orientation; however, the more basic application would be to interpret the data into a consistent theory explaining the actions of stresses on the structure of the ceramic and thereby predicting changes in the parameters of interest.

The ultimate goal is to understand the effects of stress on ceramic well enough to predict the sensitivity of an acoustic element as a function of pressure. This application can be illustrated with the following example.

A capped radially polarized ceramic cylinder often is used as an acoustic element in a transducer. As seen in Eqs. (1-3), the stresses generated by hydrostatic pressure are a function of radial position, hence stress gradients exist that differ for every cylinder of different wall thickness. If the cylinder is divided up into a number of concentric cylinders (a number sufficiently large that the stress gradient in the wall of each subcylinder is negligible) and then each subcylinder is divided into a number of small unit volumes, as shown in Fig. 5, then one could go to a graph similar to Fig. 3, where the effects of two orthogonal stresses, each perpendicular to the direction of polarization, and a given stress parallel to the direction of polarization are given, and find a value of the piezoelectric constant g_{33} or g_{31} . The sensitivity of that i -th unit volume then could be calculated, and repeating this for N unit volumes, the sensitivity M of the entire cylinder could be known:

$$M = \sum_{i=1}^N M_i \quad (6)$$

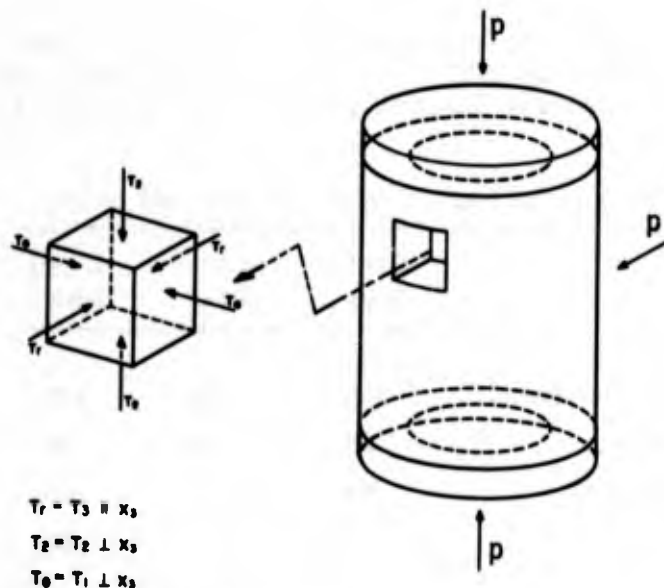


Fig. 5. Resolution of a cylinder into unit volume elements.

A computer program could be used to predict the sensitivity of a configuration of any dimensions as a function of pressure based upon the knowledge of the effects of stress upon a unit volume.

Future Studies

The program proposed here is essentially a collection of data and a basic study of stress interaction with ceramics. The example of application discussed above would be the result of a future study. Sophistications in the unit sensitivity summing doubtlessly would be necessary because of internal constraints and interactions.

The dependence of ceramic parameters upon stress is only one facet of the overall problem. The parameters are also functions of temperature, pressure cycling, and time (aging). A complete representation including all these variables should be achieved eventually.

Program Management and Personnel

This is a proposed program to be implemented by the Underwater Sound Reference Division, Code 8200, of NRL. The principal investigator responsible for the division's effort will be Dr. R. W. Timme.

Periodic reviews by the most competent personnel in Code 8200 as well as by the sponsoring agency will be the principal management tool. Written progress reports will be submitted to the sponsoring agency on a quarterly basis. The written reports will be supplemented by oral presentations and reviews as appropriate. Publications and patent applications expected during the investigation will describe significant results.

Time and Costs

The work described in this proposal is expected to span a period of three years at the level of effort of one scientific man-year plus necessary supply and service support per year. An estimate of expenses is given in Table I.

Table I. Estimated cost of proposed study

	First Year	Second Year	Third Year
Salaries (and overhead)	36K	38K	40K
Materials	5K	5K	5K
Equipment	8K	4K	4K
Travel	1K	2K	2K
Total	\$50K	\$49K	\$51K

References

1. R. W. Timme, R. L. Davidson, and A. C. Tims, "Expendable Hydrophone for Sonobuoy Application," NRL Report 7644, 17 Sep 1973 [AD-766 786].
2. MIL-STD-1376 (SHIPS), "Piezoelectric Ceramic for Sonar Transducers," 21 Dec 1970.
3. H. H. A. Krueger and J. Berlincourt, "Effects of High Static Stress on the Piezoelectric Properties of Transducer Materials," J. Acoust. Soc. Amer. 33, 1339-1344 (1961).
4. G. E. Martin, "Effects of Static Stress on the Dielectric, Elastic, and Piezoelectric Properties of Ceramics," unpublished report.
5. A. M. El'gard, "Effect of Uniaxial Mechanical Stresses on the Dielectric and Piezoelectric Properties of Polarized Ferroelectrics," Soviet Phys.--Sol. State 6, 1984-1990 (1965).
6. R. K. Linde, "Depolarization of Ferroelectrics at High Strain Rates," J. Appl. Phys. 38, 4839-4842 (1967).
7. H. H. A. Krueger, "Stress Sensitivity of Piezoelectric Ceramics Part 1: Sensitivity to Compressive Stress Parallel to the Polar Axis," J. Acoust. Soc. Amer. 42, 636-645 (1967).
8. H. H. A. Krueger, "Stress Sensitivity of Piezoelectric Ceramics Part 3: Sensitivity to Compressive Stress Perpendicular to the Polar Axis," J. Acoust. Soc. Amer. 43, 583-591 (1968).
9. R. R. Whymark and K. J. Triebes, "Effect of Large-Amplitude Static and Dynamic Stress on the Mechanical Loss of Lead Zirconate Titanate Transducer Ceramic," J. Acoust. Soc. Amer. 45, 587-591 (1969).
10. R. F. Brown, "Effect of Two-Dimensional Mechanical Stress on the Dielectric Properties of Poled BaTiO₃ and Pb(Zi,Ti)O₃," Can. J. Phys. 39, 741-753 (1961).
11. R. F. Brown and G. W. McMahon, "Material Constants of Ferroelectric Ceramics at High Pressures," Can. J. Phys. 40, 672-674 (1962).
12. R. Y. Nishi and R. F. Brown, "The Behavior of Piezoceramic Projector Materials Under Hydrostatic Pressure," J. Acoust. Soc. Amer. 36, 1292-1296 (1964).
13. Derivation given by R. A. Langevin, "The Electro-Acoustic Sensitivity of Cylindrical Ceramic Tubes," J. Acoust. Soc. Amer. 26, 421-427 (1954).
14. S. P. Timoshenko and J. N. Goodier, *Theory of Elasticity* (McGraw-Hill, New York, 1970) 3rd ed., p. 395.