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Strain Dependence of Longitudinal Piezoelectric, Elastic, and Dielectric Constants of X-Cut Quartz*

R. A. Graham
Sandia Laboratories, Albuquerque, New Mexico 87115
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Experimental measurements of the piezoelectric current from impact-loaded *X*-cut quartz are utilized to determine the finite-strain piezoelectric and elastic constitutive relations for uniaxial strain η_1 from 2.4×10^{-3} to 4.3×10^{-2}. The measurements are analyzed to provide values for the linear piezoelectric stress constant e_{11}, the direct-effect nonlinear constant $\partial e_{11}/\partial\eta_1$, the strain dependence of the permittivity, and the second-, third-, and fourth-order longitudinal elastic constants. It is found that consitutive relations developed to describe nonlinear responses at small strains describe the response of *X*-cut quartz for the large strains employed in the present investigation. The results show that $e_{11}=(0.1711 \pm 0.00094)$ C m^{-2}, $\partial e_{11}/\partial\eta_1 = -2.64\pm 0.048$ C m^{-2}, and $\epsilon_{11}^{-1}\,\partial\epsilon_{11}/\partial\eta_1 = -0.46\pm 0.13$. The elastic constants are $c_{11}=(0.868\pm 0.0095)\times 10^{12}$ dyn cm^{-2}, $c_{111}=(-3.0\pm 0.3)\times 10^{12}$ dyn cm^{-2}, and $c_{1111} = +(75\pm 25)\times 10^{12}$ dyn cm^{-2}. Data from previous authors are analyzed to obtain values for the nonlinear piezoelectric constants at 573 and 79 °K.

I. INTRODUCTION

This paper reports measurements of nonlinear elastic and piezoelectric constitutive relations for *X*-cut α quartz.[1] Elastic shock-compression responses are utilized to measure the piezoelectric stress constant e_{11}, the strain dependence of e_{11}, the strain dependence of the permittivity, and the strain dependence of the longitudinal elastic constants. The measurements are accomplished for elastic compressions of from 2.4×10^{-3} to 4.3×10^{-2}. The present work is the first quantitative experimental determination of a nonlinear piezoelectric constitutive relation.

An elastic shock wave is introduced into each sample by subjecting the *X*-cut quartz disk to a precisely controlled planar impact. As a result of the impact, a shock wave traverses the disk causing a current to flow in a low-impedance resistive circuit connecting electrodes on the faces of the disk. The current pulse is a result of the direct piezoelectric effect[2]; as such, the current provides a direct measure of the piezoelectric polarization of the shock-loaded sample. Since *X*-cut quartz remains elastic to strains of 4.3×10^{-2}, the contributions of the nonlinear piezoelectric and elastic constants are large and may be readily detected.

Various nonlinear acoustic effects in elastic solids were recently reviewed by Zarembo and Krasil'nikov.[3] Of particular interest are the nonlinear interactions of acoustic and microwave electric fields in piezoelectric solids which lead to unique electrical responses at microwave frequencies.[4] Although the microwave experiments have demonstrated the existence of the nonlinear interactions, the nonlinear piezoelectric constants have not yet been measured. Acoustic second-harmonic-generation experiments in *X*-cut quartz have shown the existence of nonlinear piezoelectric responses; however, the experiments provided only an order-of-magnitude estimate for the nonlinear piezoelectric constant.[5,6]

Third-order elastic constants characteristic of the unstrained state have been determined for a number of solids.[3,7] The present investigation extends these measurements by several orders of magnitude in strain amplitude. Recently, third- and fourth-order longitudinal elastic constants were determined for sapphire and fused quartz in a manner similar to that employed in the present investigation.[8]

Previous investigations of short-circuited piezoelectric currents from shock-loaded X-cut quartz have served to delineate the principal features of the piezoelectric response for stresses from 2.6 to 300 kbar.[9–13] In this stress range, drastically different current-vs-time pulses are observed which range from those which can be described in terms of linear elastic constitutive relations to those which must be described with highly nonlinear inelastic and electrical properties. The principal observations of these earlier investigations were that X-cut quartz exhibits (i) a very large elastic limit under shock loading,[10,14,15] (ii) a small increase in piezoelectric constant with compression,[11] and (iii) distortion of current-time waveforms due to shock-induced conductivity in the elastic range.[12,13]

This paper is organized in the following way. Nonlinear constitutive relations for elastic and piezoelectric solids are presented in Sec. II. The experimental arrangement is then described in Sec. III. An electrostatic model is developed in Sec. IV to relate the measured displacement currents to the strain-induced piezoelectric polarization. Following the presentation of results in Sec. V, the various piezoelectric elastic constants and dielectric constants are compared to related work in Sec. VI. Data from previous authors are analyzed to obtain the temperature dependence of the nonlinear piezoelectric constant. Finally the principal results of the investigation are summarized in Sec. VII.

II. NONLINEAR CONSTITUTIVE RELATIONS

Nonlinear constitutive equations describing the elastic response of materials are frequently developed from expansions of strain energy in powers of a finite-strain measure. When truncated after the first or second nonlinear term, the equations have been successfully used to describe nonlinear effects observed at small strains.[16] The applicability of these equations to the description of solids at the large elastic compressions employed in the present investigation remains to be demonstrated. In this regard, the quantitative values obtained for the nonlinear coefficients in the present experiments provide an explicit measure of the applicability of the expansions to the large strains employed.

While general concepts in nonlinear elasticity are well established, the extension of the theory to include electrical effects is nontrivial and is the subject of active investigation by Toupin,[17] Eringen,[18] and Tiersten.[19] Toupin[17] first pointed out that considerations of invariance indicate that the use of local polarization, not the macroscopic electric field, leads to rotationally invariant equations. Since the present investigation is restricted to one-dimensional measurements, rotational invariance need not be invoked, and the interpretation of the measurements will be based on constitutive equations which employ the electric field.[20] It should be noted that the experimental data themselves will be used to justify the form of the piezoelectric constitutive relation.

A. Nonpiezoelectric Elastic Solids

Following Thurston,[16] finite deformations are described in terms of a coordinate system (a_1, a_2, a_3) which identifies a material particle and a coordinate system (x_1, x_2, x_3) which identifies a spatial position. The x_i are spatial or Eulerian coordinates and the a_i are material or Lagrangian coordinates. The displacement d_i may be written

$$d_i = x_i - a_i\,,\quad i = 1, 2, 3\,. \tag{1}$$

Lagrangian or material strains η_{jk} are then defined as the difference of the squares of the lengths of line elements as

$$2\eta_{jk}\,da_j\,da_k = dx_i\,dx_i - da_i\,da_i\,, \tag{2}$$

where the Einstein summation is used. The constitutive relation will be developed from an expansion of internal energy $\overline{U}$ at constant entropy s. It is convenient to define a thermodynamic tension

$$t_{km} = \rho_0\left(\frac{\partial \overline{U}}{\partial \eta_{km}}\right)_s$$

and elastic constants of the form

$$c^s_{ijkl} = \left(\frac{\partial t_{ij}}{\partial \eta_{kl}}\right)_s\,. \tag{3}$$

Expanding the internal energy $\overline{U}(\eta, s)$ about the unstrained state, we find

$$[\overline{U}(\eta, s) - \overline{U}(0, s)] = \tfrac{1}{2}c^s_{ijkl}\eta_{ij}\eta_{kl} + \tfrac{1}{6}c^s_{ijklmn}\eta_{ij}\eta_{kl}\eta_{mn} + \tfrac{1}{24}c^s_{ijklmnpq}\eta_{ij}\eta_{kl}\eta_{mn}\eta_{pq} + \cdots\,. \tag{4}$$

Hence,

$$t_{ij} = c^s_{ijkl} + \tfrac{1}{2}c^s_{ijklmn}\eta_{ij}\eta_{kl} + \tfrac{1}{6}c^s_{ijklmnop}\eta_{ij}\eta_{kl}\eta_{mn} + \cdots + \text{higher-order terms (h. o. t.)}\,. \tag{5}$$

Since the present experiments are concerned only with longitudinal response in uniaxial strain, Eq. (5) is specialized with more compact notation to yield

$$t_1 = c^s_{11}\eta_1 + \tfrac{1}{2}c^s_{111}\eta_1^2 + \tfrac{1}{6}c^s_{1111}\eta_1^3 + \cdots + \text{h. o. t.}\,, \tag{6}$$