Second- and third-order piezoelectric stress constants of lithium niobate as determined by the impact-loading technique*

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Determination of the e_{22}, e_{33}, and e_{15} second-order piezoelectric stress constants, several third-order piezoelectric stress constants, and the c_{11}^D and c_{33}^D elastic stiffness constants are reported for lithium niobate from experiments with input strains from 7×10^{-4} to 8×10^{-3} produced by the elastic impact-loading method. Measurements of the e_{33} constant were made on a large number of samples to establish sample uniformity. The differences were found to be less than 1%. The present value of e_{33} is higher than that reported in previous work and appears to call for a revision of the accepted value along with that of the elastic constant c_{33}^E. The third-order piezoelectric stress constants are readily detectable, but the values determined in the present investigation are limited in accuracy due to the relatively low strains which could be applied to the samples before conductivity became excessive.

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I. INTRODUCTION

The purpose of this paper is to report the results of an experimental determination of certain second- and third-order piezoelectric stress constants and several second-order elastic stiffness constants for lithium niobate. The measurements were made with the impact-loading technique.

The piezoelectric properties of lithium niobate are of importance in the operation of a wide variety of piezoelectric devices including transducers, delay lines, filters, and resonators which may operate in either bulk- or surface-wave modes. (For examples of typical devices see Ref. 1.) Although bulk-wave piezoelectric devices are influenced principally by second-order piezoelectric constants, surface-wave devices which operate at microwave frequencies may produce localized strains of the order of 10^{-4} and induce responses influenced by third-order piezoelectric constants. Proposed devices for convolution and correlation of signals at microwave frequencies use the third-order piezoelectric response as the principal mode of operation.[2] Piezoelectric resonators which operate at large fields are also influenced by third-order piezoelectric constants. Furthermore, the determination of third-order elastic constants in piezoelectric solids requires knowledge of the third-order piezoelectric constants.

We have been interested in the use of quartz and lithium niobate piezoelectric gauges[3–5] for precise time-resolved measurements of impulsive loads which produce compressive stresses of the order of 1 GPa (10 kbar) and strains which are typically 10^{-2}. At these large strains output signals may be significantly influenced by nonlinear piezoelectric, dielectric, and elastic response.

Although knowledge of nonlinear piezoelectric constants is of increasing importance, only limited measurements of nonlinear piezoelectric constants have been made and there are no standard techniques for their determination. The first experiment which involved the effect of nonlinear piezoelectric constants was performed by Hruska[6] who detected a change in resonant frequency of quartz resonators as the magnitude of the biasing electric field was increased.

Various techniques which have been used to determine nonlinear piezoelectric constants involve either indirect or direct piezoelectric effects. Those techniques which utilize the indirect effect are (i) strain and/or electric field dependence of ultrasonic-wave velocities or resonant frequencies[7,8] and (ii) pressure derivatives of ultrasonic-wave velocities under different electrical boundary conditions.[9,10] Those techniques which utilize the direct effect are (iii) interaction of microwave-frequency acoustic waves,[11,12] (iv) stress-induced piezoelectric polarization measurements under static loading,[13] (v) strain-induced piezoelectric polarization measurements under impact loading,[14,15] and (vi) pressure-induced hydrostatic piezoelectric polarization measurements under hydrostatic loading.[16] Accuracy of the interpretation of ultrasonic-wave velocity and resonant frequency measurements is hampered by the separation of third-order elastic and piezoelectric contributions, and the most accurate nonlinear piezoelectric constants have been determined with techniques (v) and (vi).

The second-order piezoelectric constants of lithium niobate have been widely investigated; nevertheless, there is a wide variation in the reported values of the d_{33} and e_{33} piezoelectric constants. The most likely explanation for the different values obtained for these constants is a variation in properties due to the effects of ferroelectric domains. The possibility that there are variations in material properties from sample to sample makes it important to determine if present crystal-growth techniques yield crystals with reproducible properties. The impact-loading method is destructive and numerous samples are used; hence, investigation of material reproducibility follows naturally with that technique.

Lithium niobate has been described as a "frozen-in" ferroelectric since the application of an electric field does not rotate in the direction of polarization at temperatures remote from the Curie temperature of 1475 K. The remanent polarization has been found to be unchanged by application of hydrostatic pressure to 2 GPa,[16] but the effect of uniaxial strain on remanent polarization has not been determined.

Section II presents the thermodynamic definitions of material constants which are to be used to interpret the data. In Sec. III the relation between the current pulses from impact-loaded samples and the piezoelectric and dielectric constants is given. The experimental configurations used for the present measurements are then shown in Sec. IV. The results of the measurements are presented in Sec. V and discussed in Sec. VI. Finally, the significance of the present measurements is discussed in Sec. VII.

II. THERMODYNAMIC DEFINITION OF MATERIAL CONSTANTS

Large strains and electric fields are encountered in the present experiments and nonlinear constitutive relations are required for the interpretation of the experimental observations. The formation of nonlinear piezoelectric constitutive relations has been considered by numerous authors,[8,11,17–28] but, as yet, there is no generally accepted notation or definition of terms.

The electrical enthalpy, $H_2=U-E_jD_j$, is a function of material strain η_{ij}, electric field E_j, and entropy θ. U is the internal energy and D_j is the electrical displacement. The material strain is related to the displacement by the relation[26] $2\,da_j\,da_k\eta_{jk}=dX_i\,dX_i-da_i\,da_i$ where X is the location of a mass element in a spatial coordinate system and a is the location of a mass element in a material coordinate system. The thermodynamic tension t_{ij} is taken to be the derivative of the internal energy with respect to the strain at constant entropy, and the electric displacement D_j is taken to be the derivative of the electrical enthalpy with respect to the electric field at constant entropy and strain. Accordingly, to third-order in energy,

$$t_{ij}=\rho_0\left(\frac{\partial U}{\partial \eta_{ij}}\right)=c^E_{ijkl}\eta_{kl}-e_{kij}E_k+\tfrac{1}{2}C^E_{ijklmn}\eta_{kl}\eta_{mn}-\tfrac{1}{2}f_{klij}E_kE_l-\tfrac{1}{2}e_{ijklm}E_m\eta_{kl} \qquad \text{(1a)}$$

and

$$D_i=\frac{-\partial H_2}{\partial E_i}=e_{ijk}\eta_{jk}+\epsilon^{\eta}_{ij}E_j+\tfrac{1}{2}e_{ijklm}\eta_{jk}\eta_{lm}+\tfrac{1}{2}f_{ijkl}E_j\eta_{kl}+\tfrac{1}{2}\epsilon^{\eta}_{ijk}E_jE_k, \qquad \text{(1b)}$$

where ρ_0 is the density at zero strain, c^E_{ijkl} is the second-order elastic stiffness constant at constant field, c^E_{ijklmn} is the third-order elastic stiffness constant at constant electric field, e_{kij} is the second-order piezoelectric stress constant, e_{ijklm} is the third-order piezoelectric stress constant, ϵ^{η}_{ij} is the dielectric permittivity at constant strain, ϵ_{ijk} is the third-order dielectric permittivity, and f_{klij} is the electrostrictive constant. Strains and thermodynamic tensions are taken to be positive in tension. Transformations between thermodynamic tension and stress and between finite strain and compression under uniaxial strain conditions are given in Ref. 14. Small pyroelectric contributions to electric displacement due to isentropic heating are treated in Sec. VI.

Theories which seek solutions in the uncoupled approximation do not include the contributions of piezoelectric stiffening terms in Eq. (1a), while theories which seek solution in the weak-coupling approximation take the uncoupled solutions and add contributions due to the piezoelectric stiffening to account for the electromechanical coupling effects. In the present investigation the data are interpreted in terms of the weak-coupling approximation.

The present investigation was conducted on Z-, Y-, and 36°-rotated–Y-cut (hereafter called the rotated cut) samples. The second-order longitudinal piezoelectric stress constants for the Z- and Y-cut samples are e_{33} and e_{22}, respectively, whereas the rotated cut is characterized by a combination of the four piezoelectric constants. For lithium niobate, a crystal of symmetry $3m$, the rotated linear piezoelectric constant is[29]

$$e'_{22}=2e_{15}\cos^2\theta\sin\theta+e_{22}\cos^3\theta+e_{33}\sin^3\theta+e_{31}\cos^2\theta\sin\theta, \qquad (2)$$

where $\theta=36°$ for the rotated cut of the present investigation.

Experiments on the Y- and rotated-cut samples involve wave propagation along nonspecific directions in the crystal. When isotropic solids are subjected to a planar impact loading, shock waves propagate through the samples with particle motion directed along the direction of propagation. When anisotropic crystals are subjected to the same loading, this uniaxial strain condition is encountered only if the loading is applied along certain specific directions in the crystal.[30] In the present investigation only the Z-cut samples are loaded along a specific direction. Nevertheless, numerical calculations by Johnson[31] indicate that analysis of the piezoelectric response in terms of uniaxial strain for impact loading along the Y and 36°-rotated–Y directions should be correct to a suitable approximation since the amplitude of the quasi-longitudinal wave is

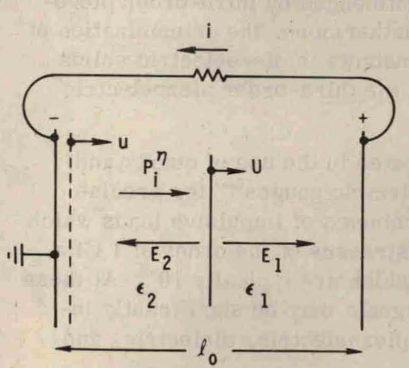

FIG. 1. The electrical conditions in one-dimensional regions of a shock-loaded piezoelectric disk are as shown. The impact was applied to the left face of the disk, and at any time before wave transit time the shock front divides the sample into two characteristic regions within which the conditions are uniform. The electrodes are assumed to be connected with an electrical short circuit. The positive coordinate direction is from left to right.