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STRAIN DEPENDENCE OF THE PIEZOELECTRIC POLARIZATION OF *z*-CUT LITHIUM NIOBATE*

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Measurements of the piezoelectric polarization of *z*-cut lithium niobate are accomplished for dynamic compressive strains of from 8.9×10^{-4} to 6×10^{-3}. Values for the linear piezoelectric constant, e_{33} and its strain derivative are determined. The logarithmic strain derivatives of the longitudinal piezoelectric constants of *x*-cut quartz and *z*-cut lithium niobate are found to be approximately equal.

THE PRESENT paper reports measurements of the piezoelectric polarization of *z*-cut lithium niobate ($LiNbO_3$) at compressive elastic strains, η_3, from 8.9×10^{-4} to 6×10^{-3}. Values for the linear piezoelectric stress constant, e_{33}, and the nonlinear constant $e_{333} = \partial^2 P_\eta / \partial \eta_3^2$, are derived from the measurements; the nonlinear constant is measured for the first time. The present work extends recent measurements of the linear and nonlinear constants of *x*-cut quartz[1] to a solid with significantly larger piezoelectric and dielectric constants.

Dynamic elastic strains are applied to specimen disks of *z*-cut lithium niobate by the precisely controlled impact of projectiles at velocities which range from 20 to 150 msec^{-1}. The impacts produce plane, longitudinal, elastic shock-waves which travel through the disks and produce piezoelectric currents in low impedance resistive circuits connecting electrodes on the faces of the disks. A vapor-plated guard-ring electrode configuration ensures that the electrical measurements are representative of regions of the specimen which are subjected to uniaxial strain, uniaxial polarization and uniaxial electric field along the *z* axes of the disks. Except for modifications noted below, the technique is the same as they employed to investigate *x*-cut quartz and details of the technique are fully reported in previous papers.[1,2]

The present experiments utilize the impact of *x*-cut quartz projectiles upon the *z*-cut lithium niobate samples. Determination of the strain imparted to the samples requires knowledge of the impact velocity and the nonlinear constitutive relations of the projectile and sample. Although the third-order elastic constants of lithium niobate have not been measured, consideration of the strain amplitude, the uncertainty of the third-order constant of *x*-cut quartz,[1] and the impact velocity shows that the experimental error in the strain is small compared to the experimental accuracy (1–1/2 per cent) of the current pulse amplitude.

Since each experiment is destructive, it is necessary to evaluate the reproducibility of the sample material. The investigation has included study of samples from seven different boules of 'Transducer Grade' lithium niobate as specified by Crystal Technology, Inc. The disks were oriented, cut, lapped and polished to precise dimensional tolerance by the Valpey–Fisher Corporation. The experimental results showed no sample to sample variations.

An electrostatic model has been developed[1] to relate the observed current to the piezoelectric polarization, P_η. In the absence of conductivity, P_η is related to the jump in current, i_i, produced by the impact by the relation

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$$P_\eta = \frac{\alpha t_0}{A(1-\gamma)} i_i \,, \qquad (1)$$

where η is the Lagrangian strain, α is the ratio of the strained to unstrained permittivity taken to be unity in the present case, γ is the linear strain, A is the area of the charge collecting electrode, and t_0 is the time for the shock wave to traverse the disk.

It should be noted that, unlike techniques which rely upon electromechanical coupling effects on wave speeds to determine piezoelectric constants, the present technique utilizes the direct piezoelectric effect to provide a direct measure of the piezoelectric polarization. Under these conditions, the magnitude of the nonlinear contribution depends explicitly on the magnitude of the strain.

The current-time traces for experiments conducted at strains greater than 6×10^{-3} showed effects of shock-induced conductivity.[3] Because the electrostatic model used to develop equation (1) presumes that the conductivity is zero, the observed current–time records for strains greater than this could not be used to derive accurate values for the piezoelectric polarization. This limiting maximum strain value severely limits the contribution of the nonlinear constant and correspondingly large errors are obtained.

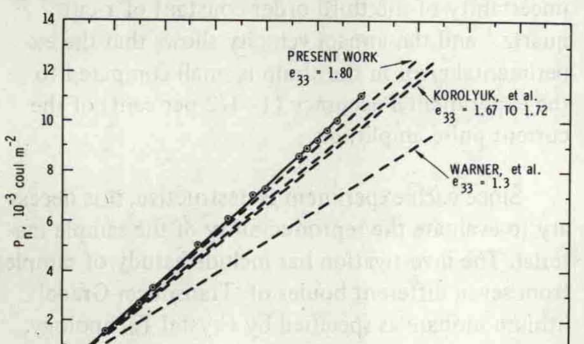

FIG. 1. The experimentally observed piezoelectric polarization is plotted at various compressive Lagrangian strains. The various dashed lines represent extrapolations to large strain of the linear piezoelectric constant, e_{33}, observed by various authors.

The observed piezoelectric polarization vs. Lagrangian strain data are shown in Fig. 1 along with extrapolations of linear constants determined by previous investigations. The most obvious result from inspection of the figure is that there is substantial disagreement between the various measurements as to the magnitude of the e_{33} constant. The various values are tabulated in Table 1.

Table 1. Linear piezoelectric stress constants of lithium niobate

	e_{33}, C m^{-2}
Present work	1.80 ± 0.025
Warner *et al.*[4]	1.3
Smith *et al.*[5]	1.33
Korolyuk *et al.*[6]	1.67 and 1.72

± indicates standard error

The linear constant determined from our experiment is about 6 per cent higher than the values reported by Korolyuk *et al.* This difference is probably within the experimental error of the two measurements. On the other hand, the value of e_{33} determined by Warner *et al.* differs from the present value by about 30 per cent and their value was recently confirmed by Smith and Welsh. The various values obtained from the e_{33} constant by the various investigators are cause for considerable concern.

Even though the present technique is not as widely applied to the measurement of piezoelectric constants as the techniques used by the previous investigators, the present technique provides a sensitive and direct method for determining longitudinal piezoelectric constnants. The e_{11} constant determined for *x*-cut quartz by the present technique[1] is in excellent agreement with the established value of Bechmann[7] and the errors with the e_{11} constant indicate that the present technique provides the most accurate value ever determined for that constant. Crystal growth conditions and incomplete poling of lithium niobate samples have been known to cause sample-to-sample variations in properties.[8–10] With the recent advances in crystal growth technique and material characterization,[10,11] the present author believes that the most likely cause for the various e_{33} values is the use of multidomain crystals. If that is the case, the preferred value is the largest obtained.

Table 2. piezoelectric constants

	e_{xx} ($C\ m^{-2}$)	$\partial e_{xx}/\partial \eta_x$ ($C\ m^{-2}$)	$\partial \ln e_{xx}/\partial \eta_x$
x-cut quartz[1]	0.1711 ± 0.00094	2.62 ± 0.048	15.3 ± 0.3
z-cut lithium niobate	1.80 ± 0.025	18 ± 6	10 ± 3

± indicates standard error except for nonlinear constants of lithium niobate which are maximum experimental errors. *x* and *xx* denote 1 and 11 for quartz and 3, and 33 for lithium niobate.

The linear and nonlinear constants of lithium niobate are compared to corresponding values obtained for *x*-cut quartz in Table 2. The nonlinear constant has a large (40 per cent) standard error; however, consideration of the maximum experimental error gives a maximum error of ± 30 per cent for this constant. It should be noted that even though the linear constants differ by an order of magnitude, the logarithmic derivatives with strain have approximately the same value. Thus, the magnitude of the nonlinear constants appears to depend upon the value of the linear constant. This observation is in agreement with recent observations by Lim *et al.*[12] of the electromechanical coupling associated with nonlinear interaction of acoustic surface waves which involve contributions from a combination of tensor components. These authors found that materials with lower linear coupling constants exhibited lower nonlinear coupling constants. The present value for the nonlinear constant is also in good agreement with an estimate by Thompson and Quate,[13] who utilized a microwave acoustic technique.

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Auszug – Es werden Messungen der piezoelektrischen Polarisierung von Lithiumniobat-z-Schnitt für dynamische Druckspannungen von 8, 9 $\times 10^{-4}$ bis 6 $\times 10^{-3}$ durchgeführt. Es werden Werte der linearen piezoelektrischen Konstante e_{33} und ihre Spannungsableitungen ermittelt. Es wurde festgestellt, dass die logarythmischen Spannungsableitungen der longitudinalen piezoelektrischen Konstanten des x-Schnitt Quartzes und z-Schnitt Lithiumniobats ungefähr gleich sind.