

## Shear elastic nonlinearity of electro-elastic crystals\*

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**Abstract** Shear elastic nonlinearity is examined through the second harmonic generation (SHG) of transverse waves in cubic, hexagonal and trigonal crystals. An experiment is carried out for Z-cut LiNbO<sub>3</sub>. Its third-order elastic constant  $C_{444}$  is determined.

**Keywords:** shear elasticity, nonlinearity of crystal

The SHG of transverse waves can be a powerful tool to investigate the shear elastic nonlinear properties of materials. The polarization can be different for a transverse wave propagating along a given direction, which can help to probe varieties of processes that are polarization sensitive. Moreover, the geometrical nonlinearity does not contribute to the SHG of a pure mode transverse wave; therefore, it is a direct manifestation of the material nonlinear properties.

### 1 SHG of transverse waves in crystal

The rotationally invariant state function for an electro-elastic crystal can be expressed as Taylor's series<sup>[1]</sup>:

$$\psi = \frac{1}{2} c_{ijkl} \eta_{ij} \eta_{kl} - e_{ijk} W_i \eta_{jk} - \frac{1}{2} \chi_{ij} W_i W_j + \frac{1}{6} c_{ijklmn} \eta_{ij} \eta_{kl} \eta_{mn} - \frac{1}{2} e_{mijk} W_m \eta_{ij} \eta_{kl} - \frac{1}{2} b_{ijkl}^s W_i W_j \eta_{kl} - \frac{1}{6} \chi_{ijk} W_i W_j W_k \quad (1)$$

where  $\eta_{ij}$  is Green finite strain tensor

$$\eta_{ij} = \frac{1}{2} (u_{i,j} + u_{j,i} + u_{k,j} u_{k,i}) \quad (2)$$

and  $W$  is a material form electric field which related to ordinary spatial electric field  $E$  by:

$$W_i = E_j x_{i,j} \quad (3)$$

In Eq. (1) terms up to cubic order are preserved. The material constants  $c_{ijkl}$  and  $c_{ijklmn}$  are the second- and third-order elastic constants,  $e_{ijk}$  are linear piezoelectric constants,  $\chi_{ij}$  and  $\chi_{ijk}$  are the second- and third-order dielectric susceptibility,  $b_{ijkl}^s$  are the electrostrictive constants, and  $e_{mijk}$  are the third-order piezoelectric coefficients. For a plane wave propagating along an arbitrary direction, a working coordinate system (**abc**) can be constructed through a rotational transformation as shown in Fig. 1. Axis **a** is corresponding to wave propagation direction, **b** and **c** are polarization directions.

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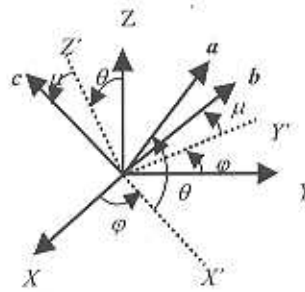


Fig.1 Constitutive and working coordinate system.

Obviously, the vector  $W$  ( $W_x, W_y, W_z$ ) in  $(XYZ)$  can be expressed as a vector. Constitutive and working  $W'$  ( $W'_a, W'_b, W'_c$ ) in  $(abc)$  by a simple transformation,

$$[W] = [\alpha]^T \cdot [W'] \quad (4)$$

Also the strain tensor  $\eta$  in the  $(XYZ)$  system can be represented by the strain tensor  $\eta'$  in  $(abc)$  through

$$[\eta] = [\alpha]^T \cdot [\eta'] \cdot [\alpha] \quad (5)$$

where  $[\alpha]$  is the coordinate transformation matrix,  $[\alpha]^T$  is its transpose. Thus, the scale state function can be expressed as a function of  $W'$  and  $\eta'$  by substituting Eq. (4) and (5) into Eq. (1). Under this case the state function can be written as

$$\psi = \psi_2 + \psi_3 = \frac{1}{2} c'_{LKL} \eta'_{L1} \eta'_{K1} + \frac{1}{6} c'_{MKLMN} \eta'_{L1} \eta'_{K1} \eta'_{MN} \quad (6)$$

without considering piezoelectricity. Thus the bilinear equation of motion for a plane wave propagation along  $a$ -direction is expressed as:

$$\rho_0 \ddot{u}_i = \frac{\partial}{\partial a} \left[ \frac{\partial \psi_2}{\partial \eta'_{i1}} + \left( \frac{\partial u_1}{\partial a} \frac{\partial \psi_2}{\partial \eta'_{11}} + \frac{\partial \psi_3}{\partial \eta'_{11}} \right) \right] \quad (i = 1, 2, 3) \quad (7)$$

Here we refer to the  $a$ -direction as direction 1. For a pure mode transverse wave in the working system  $(abc)$  Eq. (7) gives:

$$\rho_0 \ddot{u}_i - M_2 u_{i,11} = M_3 u_{i,1} u_{i,11} \quad (i = 2 \text{ or } 3) \quad (8)$$

where  $u_i$  is the particle displacement component of a pure mode wave. The calculations show that  $M_2$ , the coefficient before the bilinear term  $u_{i,1} u_{i,11}$  in the equation, involves only the third-order elastic constants or their combination for transverse waves. This is different from the case of longitudinal waves where  $M_2$  involves both the second and the third-order elastic constants<sup>[2]</sup>. This can be explained as follows. Without losing generality, we may set  $u_1 = u_3 = 0$  and  $u_2 = u_2(a, t)$ . For this case we have  $\eta'_{11} = \frac{1}{2} u_{2,1}^2$ ,  $\eta'_{21} = \eta'_{12} = \frac{1}{2} u_{2,1}$  and others are zero from Eq. (2). It is seen that  $\eta'_{12}$  (or  $\eta'_{21}$ ) is always an linear strain for a pure mode transverse wave.  $M_3$  is actually the coefficient of the quadratic terms in  $\frac{\partial \psi_2}{\partial \eta'_{21}}$  and  $\frac{\partial u_2}{\partial a} \frac{\partial \psi_2}{\partial \eta'_{11}}$ . For the pure mode direction, the term  $\eta'_{11}$  ( $\eta'_{12} + \eta'_{21}$ ) must not appear in the

expression of  $\psi_2$ . Thus  $\frac{\partial \psi_2}{\partial \eta'_{21}} \propto \eta'_{21}$  will not generate quadratic term of  $u_{2,1}$  since there is no nonlinear term in  $\eta'_{21}$ .

Because  $\frac{\partial \psi_2}{\partial \eta'_{11}} \propto \eta'_{11} = \frac{1}{2} u_{2,1}^2$ ,  $\frac{\partial u_2}{\partial a} \frac{\partial \psi_2}{\partial \eta'_{11}}$  will generate cubic terms in  $u_{2,1}$  but not square terms. Therefore, no second-order elastic constants can be involved in  $M_3$  for a pure mode transverse wave. This distinctive feature makes the SHG of transverse waves a straight manifestation of shear nonlinear properties of materials. Analogous to the case of longitudinal waves, a nonlinearity parameter  $\beta_r$  is defined as:

$$\beta_r = \frac{M_3}{M_2} \quad (9)$$

which characterizes SHG of transverse waves and is a description of the shear nonlinear property of the material.

We have calculated  $M_2$  and  $M_3$ , hence the nonlinear parameter of SHG of transverse waves for cubic, hexagonal ( $622$ ,  $6mm$ ,  $\bar{6}m2$ ,  $6/mmm$ ) and trigonal ( $32$ ,  $3m$ ,  $\bar{3}m$ ) crystals. It is found that  $M_3=0$  for most high symmetry directions of the crystals, which means the SHG of transverse waves is prohibited in those wave propagation directions. The non-zero  $M_3$  and corresponding  $M_2$  are listed in table [1] of Ref.[3] and [4] for these crystals. Piezoelectric corrections are listed in Table [2] of Ref.[4]. Obviously the solution of Eq. (8) is

$$u_r = A_1 \sin(\omega t - ka) + A_2 \cos[2(\omega t - ka)] \quad (10)$$

where  $k$  in fundamental wavenumber.  $A_2$  is second harmonic wave amplitude,

$$A_2 = \frac{1}{8} k^2 L \beta_r A_1^2 \quad (11)$$

where  $L$  is wave propagation distance. Thus  $\beta_r$  can be experimentally determined if  $A_1$  and  $A_2$  are measured, i. e.

$$\beta_r = \frac{8}{k^2 L} \left( \frac{A_2}{A_1^2} \right) \quad (12)$$

## 2 Experiment

The experimental setup is the same as that given in Ref. 5. A 5 MHz toneburst signal from an arbitrary waveform generator is amplified and filtered, then applied to the transmitting transducer centered at 5 MHz. The received waveform from the receiving transducer centered at 10 MHz is recorded by a digital oscilloscope and the data are transferred to a computer where the waveform is analyzed. The sample is Z-cut LiNbO<sub>3</sub> (Valpey-Fisher, Hopkinton) with length 2 cm. To measure absolute amplitude of the fundamental and the second harmonic waves the receiving transducer is calibrated by the procedure given in<sup>[6]</sup>. To examine the possible nonlinearity of the measurement system itself, an aluminum sample is measured first. It is found that the second harmonic is at an undetectable level of the measurement system for the aluminum sample as shown in Fig. 2(a). The result confirms that the SHG of transverse waves is prohibited in an isotropic solid.

Figure 3 gives the relation between the measured second harmonic amplitude  $A_2$  and the square of the fundamental amplitude  $A_1^2$ . The straight line is expected from Eq. (11). From the slope of the straight line  $\beta_r$  can be determined.

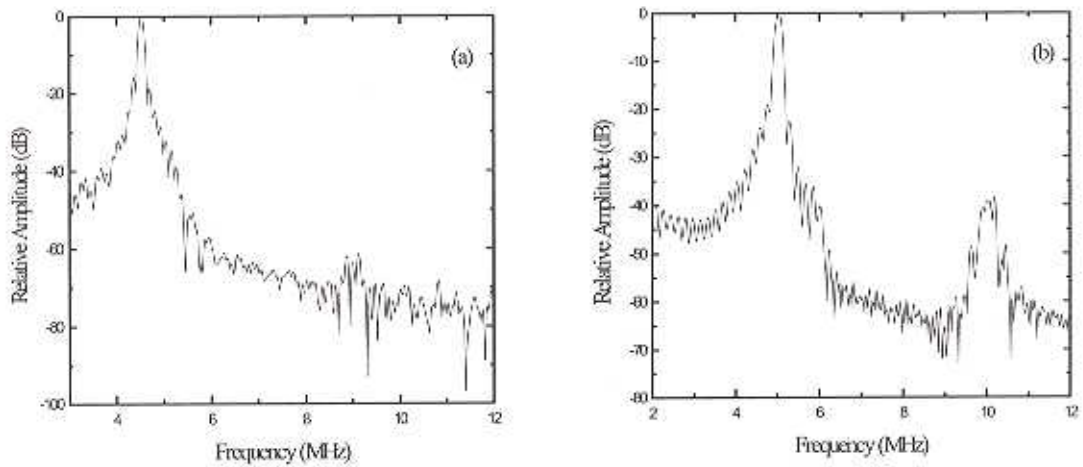


Fig.2 Frequency spectra of received signal (a) for aluminum; (b) for *X*-cut LiNbO<sub>3</sub>.

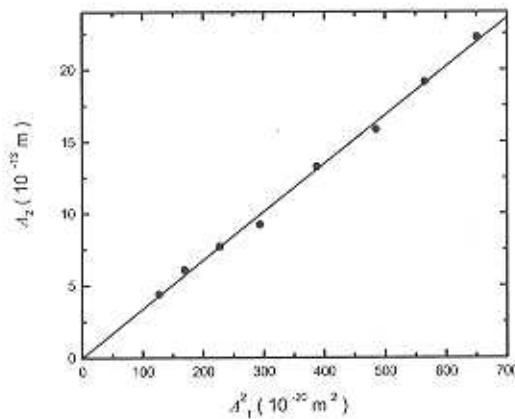


Fig.3  $A_1^2$  Vs.  $A_2$ .

It is known that for the transverse wave propagating along the *Z*-direction with polarization at the *X*-direction in 3m crystal,  $M_2=c_{44}$ ,  $M_3=c_{444}$ <sup>[3,4]</sup> and does not couple to the piezoelectric effect<sup>[4]</sup>. The measured  $\beta_3$  and the calculated third-order elastic constant  $c_{444}$  are listed in Table 1. The value of  $c_{444}$  is larger than that reported in Ref. 7 where the constant is measured by stress derivative of the sound velocity of small amplitude acoustic wave and a standard error of 0.4 is reported. The present measurement is more direct.

Table 1 Measured  $\beta$  and  $c_{444}$ .

$\beta$	$c_{44}$	$c_{444}$	
		This work	Ref.[6]
1.7	0.6	-1.02	-0.3

Unit of  $c_{44}$  and  $c_{444}$ :  $10^{11}$  N/m<sup>2</sup>

### 3 Conclusion

The calculations show that the shear nonlinearity of crystals has some characteristics which distinguish itself from the longitudinal nonlinearity. First, self-interaction of a transverse wave to generate its own second harmonic wave is permitted only in some special wave propagation directions for anisotropic electro-elastic solids. Second, the geometrical nonlinearity originated from the finite strain does not contribute to SHG of transverse waves. Therefore, the second-order elastic constants of the materials are not involved in the nonlinear parameter of the second harmonic generation.

The experiment carried out for Z-cut LiNbO<sub>3</sub> demonstrates that the shear nonlinear properties of materials can be investigated by the SHG of the transverse waves with the aid of the calibrated receiving transducer centered at the harmonic frequency to measure the amplitude of acoustic waves.

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